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Kennya Thayres dos Santos Lima

**Development of gelatin and gelatin-paperboard materials incorporated with
modified curcumin for food packaging applications**

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Kennya Thayres dos Santos Lima

Development of gelatin and gelatin-paperboard materials incorporated with modified curcumin for food packaging applications

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Orientador(a): Prof.(a) Germán Ayala Valencia, Dr.(a)

Coorientadoras: Prof.(a) Alcilene Rodrigues Monteiro Dr.(a); Prof.(a) Jéssica de Matos Fonseca, Dr.(a)

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CAMPUS UNIVERSITÁRIO REITOR JOÃO DAVID FERREIRA LIMA - TRINDADE
FLORIANÓPOLIS – SC
ppgeal.posgrad.ufsc.br TELEFONE: +55 48 3721-4063 / E-MAIL: ppgeal@contato.ufsc.br

Kennya Thayres dos Santos Lima

Development of Gelatin and Gelatin-Paperboard Materials Incorporated with Modified Curcumin for Food Packaging Applications

O presente trabalho em nível de doutorado foi avaliado e aprovado por banca examinadora composta pelos seguintes membros:

Prof. Dr. German Ayala Valencia - Presidente - UFSC
Prof^a. Dr^a. Leiliane Teles César - Membro Externo - IFCE
Prof. Dr. Sergio Yesid Gomez Gonzalez - Membro Externo - UFSC
Dr. Raul Remor Dalsasso - Membro Interno -

Certificamos que esta é a **versão original e final** do trabalho de conclusão que foi julgado adequado para obtenção do título de doutora em Engenharia de Alimentos.



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Prof^a. Dr^a. Jaciane Lutz Ienczak
Coordenadora do Programa



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Prof. Dr. Germán Ayala Valência
Orientador

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RESUMO

A embalagem é essencial para a preservação e proteção de alimentos, e os avanços recentes têm se concentrado no uso de materiais sustentáveis, como papel e biopolímeros. Apesar de suas limitações de barreira, o papel pode ser aprimorado com revestimentos biopoliméricos. Revestimentos e filmes de gelatina, por exemplo, são eficazes na proteção contra oxidação e podem ser enriquecidos com compostos bioativos, como a curcumina, que possui propriedades antioxidantes. No entanto, a eficácia da curcumina é limitada por sua baixa solubilidade e biodisponibilidade, problemas que podem ser resolvidos pela modificação de sua estrutura, aumentando suas propriedades funcionais para a preservação de alimentos. Este estudo aborda o desenvolvimento de filmes de gelatina ativa contendo curcumina modificada (MC) por precipitação antissolvente e sua aplicação para reduzir o escurecimento da banana; e investigou o desenvolvimento de embalagens de papel bicamada revestidas com gelatina e MC. Filmes de gelatina com diferentes concentrações de MC (0,0, 0,1, 0,3, 0,5 e 1%) foram produzidos pelo método de fundição; e soluções formadora de filme contendo 0 e 1% de MC foram usadas para revestir papel kraft (85,47 g/m²) para produzir embalagens bicamada. O MC apresentou maior solubilidade em água e atividade antioxidante em comparação à curcumina natural. As análises estruturais (FTIR, Raman e XRD) dos filmes confirmaram que o MC foi bem incorporado sem alterar a espessura, permeabilidade, molhabilidade e propriedades mecânicas do filme. A microscopia eletrônica de varredura (MEV) mostrou uma distribuição uniforme de MC e os filmes resultantes apresentaram atividade antioxidante melhorada (testes DPPH e ABTS) em função da concentração de MC. Filmes de gelatina contendo 1% de MC reduziram o escurecimento da banana durante o armazenamento desta fruta a 25°C e 75% UR por 6 dias. Os papéis revestidos (comparados aos papéis não revestidos) apresentaram maior selabilidade, espessura, gramatura e estrutura superficial contínua, além de menor rigidez e maior flexibilidade, especialmente na presença de MC. A permeabilidade ao vapor de água aumentou com o revestimento, enquanto a permeabilidade ao oxigênio foi reduzida, e o ângulo de contato indicou maior afinidade pela água e menor oleofilicidade. A análise de cor revelou que a adição de MC intensificou os tons vermelho-alaranjados do material. Essas descobertas destacam o potencial da combinação de MC, filmes de gelatina e papéis revestidos, que oferece uma alternativa atraente, sustentável e antioxidante para embalagens de alimentos, garantindo a preservação dos alimentos.

Palavras-chave: curcumina modificada; embalagens de alimentos; celulose; extensão de vida útil; precipitação antissolvente; papel.

RESUMO EXPANDIDO

Introdução

A embalagem é essencial para a preservação, segurança e qualidade dos alimentos, protegendo contra umidade, luz, oxigênio e contaminação microbiana, além de facilitar o manuseio e fornecer informações sobre o produto. A sustentabilidade tem se tornado cada vez mais importante, incentivando o uso de materiais recicláveis e biodegradáveis. O papel é uma opção renovável e econômica, mas precisa de revestimentos de biopolímeros, como a gelatina, para melhorar suas propriedades de barreira. A incorporação de compostos bioativos, como a curcumina, aumenta a atividade antioxidante, especialmente quando sua solubilidade é melhorada por modificações. Estudos anteriores exploraram filmes de gelatina com curcumina e papel revestido com biopolímeros, mas a combinação de gelatina com curcumina modificada (MC) em embalagens bicamada de papel ainda não foi investigada.

Objetivos

Este estudo tem como objetivo desenvolver e caracterizar materiais à base de papel kraft revestido com gelatina incorporada com MC para aplicações em embalagens de alimentos. Especificamente, busca-se analisar e classificar de forma abrangente diferentes biopolímeros e métodos de revestimento para papel ou papelão, avaliando as propriedades físico-químicas e as possíveis aplicações alimentícias desses materiais. Além disso, o estudo objetiva produzir curcumina modificada, investigar suas propriedades, desenvolver filmes de gelatina com diferentes concentrações de MC, investigar suas propriedades e avaliar sua eficácia como embalagens antioxidantes para bananas. Outro objetivo trata-se do desenvolvimento e avaliação de materiais bicamada à base de papel e gelatina incorporada com MC, visando a produção de embalagens alimentícias.

Metodologia

A metodologia da revisão da literatura envolveu uma busca bibliográfica na base SCOPUS. Inicialmente, foram pesquisados os termos “paper AND food AND packaging” dos últimos dez anos. Em seguida, uma pesquisa mais específica foi realizada com os termos “biopolymer AND coating AND paper AND packaging”, resultando em 116 artigos, dos quais apenas 20 abordavam o desenvolvimento de revestimentos biopoliméricos em papel ou papelão com potencial aplicação em alimentos; esses 20 estudos foram selecionados para a revisão. MC foi obtida por precipitação antissolvente. As propriedades de curcumina e MC foram caracterizadas quanto à morfologia, tamanho de partícula, estrutura cristalina, ligações químicas, solubilidade, cor, atividade antioxidante e atividade de água. Filmes de gelatina com diferentes concentrações de MC (0,0; 0,1; 0,3; 0,5 e 1%) foram produzidos pelo método de casting, e caracterizados quanto à espessura, morfologia, estrutura cristalina, ligações químicas, absorção e solubilidade em água, propriedades ópticas, ângulo de contato com a água, permeabilidade ao vapor de água, propriedades mecânicas e atividade antioxidante. Para aplicação em embalagens, bananas frescas foram embaladas com os filmes com melhor atividade antioxidante, incluindo controles, e armazenadas a 25 °C e 75% UR por 6 dias, sendo avaliadas, textura, perda de massa e aparência visual. Embalagens bicamada foram produzidas revestindo o papel kraft (85,47 g/m²) com soluções formadoras de filme de gelatina sem e com 1% de MC, seguidas de secagem a 25 °C. Os papeis não revestidos (PA)

e revestidos (PG e PC, com gelatina e com MC 1%, respectivamente) foram caracterizados quanto à morfologia e microestrutura, espessura, gramatura, ângulo de contato com água e óleo, propriedades de barreira a vapor de água e oxigênio, cor (CIELab e ΔE), propriedades mecânicas (módulo de elasticidade, resistência à tração e alongamento), atividade antioxidante (DPPH e ABTS) e capacidade de selagem térmica.

Resultados e Discussão

A partir da revisão da literatura, foi possível observar que o uso de revestimentos biopoliméricos em papel e papelão melhora propriedades como hidrofobicidade, resistência a óleos e algumas características mecânicas, preenchendo os poros da matriz celulósica. Revestimentos compostos e multicamadas, incluindo nanopartículas e bioativos, conferem propriedades antibacterianas, antioxidantes e barreira a gases, embora a permeabilidade ao vapor de água e oxigênio ainda seja um desafio. As pesquisas futuras devem focar em técnicas como reticulação, extrusão reativa e incorporação de nanomateriais para melhoria de barreiras e selagem térmica, mantendo a biodegradabilidade do material. A partir da análise dos resultados, foi possível observar que a modificação de MC por precipitação antissolvente promoveu redução do tamanho de partículas, mudança na morfologia para agulhas dendríticas e redução da cristalinidade, aumentando a solubilidade em água, e intensificando a coloração e a atividade antioxidante. As análises FTIR e Raman indicaram alterações nos grupos C=O e fenólicos, refletindo interações moleculares distintas nos polimorfos. A incorporação de MC em filmes de gelatina não alterou a espessura, estrutura cristalina, ligações químicas, absorção de água, solubilidade ou propriedades mecânicas, mas conferiu coloração uniforme, maior absorção de luz visível e expressiva melhoria (491%) de atividade antioxidante, tornando os filmes ativos. Na aplicação prática, filmes contendo 1% MC reduziram o escurecimento de cascas de banana durante o armazenamento, sem alterar massa ou dureza dos frutos, evidenciando o potencial do MC para embalagens de alimentos ativas. Também foi possível observar que a aplicação de revestimentos de gelatina, com ou sem MC, alterou significativamente as propriedades do papel, formando uma camada contínua que eliminou poros e aumentou espessura e gramatura, sem diferenças significativas causadas por MC; todos os papéis revestidos permaneceram hidrofóbicos, com menor interação com óleo, e apresentaram redução da permeabilidade ao vapor de água e viabilidade na barreira ao oxigênio, destacando a limitação de PA e a eficácia do revestimento. A cor foi intensamente alterada por MC, aumentando tons vermelhos e amarelos de forma perceptível, enquanto as propriedades mecânicas mostraram redução de rigidez e resistência à tração, mas aumento da ductilidade, especialmente com MC. Além disso, o revestimento proporcionou propriedades de selagem térmica, ausentes em PA.

Considerações Finais

A curcumina modificada, obtida em escala nanométrica, mostrou-se altamente eficiente em aumentar a solubilidade em água e a atividade antioxidante, conferindo potencial funcional importante para conservação de alimentos. Filmes de gelatina incorporando essa curcumina mantiveram suas propriedades físicas e mecânicas, mas apresentaram aumento expressivo da atividade antioxidante, contribuindo para reduzir o escurecimento e prolongar a vida útil de bananas. Já os papéis revestidos com biopolímeros, com ou sem curcumina modificada, apresentaram maior espessura, suavidade e flexibilidade, além de superfícies hidrofóbicas e oleofóbicas,

baixa permeabilidade a vapor de água e oxigênio, selabilidade e potencial antioxidante. A combinação de curcumina modificada, filmes de gelatina e papéis revestidos oferece uma alternativa sustentável e antioxidante atrativa para embalagens alimentícias, aliando conservação de alimentos, propriedades antioxidantes e desempenho estrutural adequado.

Palavras-chave: curcumina modificada; embalagens de alimentos; celulose; extensão de vida útil; precipitação antissolvente; papel.

ABSTRACT

Packaging is essential for preserving and protecting food, and recent advances have focused on the use of sustainable materials such as paper and biopolymers. Despite its barrier limitations, paper can be enhanced with biopolymeric coatings. Gelatin coatings and films, for example, are effective in protecting against oxidation, and can be enriched with bioactive compounds such as curcumin, which has antioxidant properties. However, the effectiveness of curcumin is limited by its low solubility and bioavailability, problems that can be solved by modifying its structure, enhancing its functional properties for food preservation. This study addresses the development of active gelatin films containing modified curcumin (MC) by antisolvent precipitation and their application to reduce banana browning; and investigated the development of bilayer paper packaging coated with gelatin and MC. Gelatin films with different MC concentrations (0.0, 0.1, 0.3, 0.5 and 1%) were produced by the casting method; and film-forming solutions containing 0 and 1% MC were used to coat kraft paper (85.47 g/m²) to produce bilayer packaging. MC showed higher water solubility and antioxidant activity compared to natural curcumin. Structural analyses (FTIR, Raman and XRD) of the films confirmed that MC was well incorporated without altering the thickness, permeability, wettability and mechanical properties of the film. Scanning electron microscopy (SEM) showed a uniform distribution of MC and the resulting films had improved antioxidant activity (DPPH and ABTS tests) as a function of MC concentration. Gelatin films containing 1% MC reduced banana browning during storage of this fruit at 25°C and 75% RH for 6 days. The coated papers (compared to the uncoated paper) showed greater sealability, thickness, weight and continuous surface structure, in addition to lower stiffness and greater flexibility, especially in the presence of MC. Water vapor permeability increased with the coating, while oxygen permeability was reduced, and the contact angle indicated higher affinity for water and lower oleophilicity. Color analysis revealed that the addition of MC intensified the red-orange hues of the material. These findings highlight the potential of the combination of MC, gelatin films, and coated papers offers an attractive, sustainable, antioxidant alternative for food packaging, while ensuring food preservation.

Keywords: modified curcumin; food packaging; cellulose; shelf life extension; antisolvent precipitation; paper.

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ABBREVIATIONS LIST

MC	Modified Curcumin
FTIR	Fourier Transform Infrared Spectroscopy
Raman	Raman Spectroscopy
XRD	X-ray Diffraction
DPPH	2,2-diphenyl-1-picrylhydrazyl (não incluído conforme solicitado)
ABTS	2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid)
UV-Vis	Ultraviolet-Visible Spectroscopy
HPMC	Hydroxypropyl Methylcellulose
Op	Opacity
WCA	Water Contact Angle
WVP	Water Vapor Permeability
RH	Relative Humidity
ΔP	Pressure Difference
ML	Mass Loss
m_0	Initial Mass
m_t	Mass at Time t (Massa no Tempo t)
L^*	CIE $L^*a^*b^*$ Color Coordinate (Brightness; 0 is black, 100 is white)
a^*	CIE $L^*a^*b^*$ Color Coordinate (Negative = green, positive = red)
b^*	CIE $L^*a^*b^*$ Color Coordinate (Negative = blue, positive = yellow)
Paper(board)	Paper and/or paperboard
PA	Uncoated paper
PG	Gelatin-coated paper
PC	Gelatin-coated paper with modified curcumin

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THESIS CONCEPTUAL SCHEME

What?

Development of gelatin and gelatin-paperboard materials incorporated with modified curcumin for food packaging applications.

Why?

- Although natural curcumin is a dye that has good antioxidant, antimicrobial and anticarcinogenic properties, it has low bioavailability and solubility, limiting its effectiveness in food applications.
- Food waste is a global problem that affects food security and exacerbates environmental problems. It is estimated that about a third of all food produced worldwide is lost or wasted.
- The excessive use of plastic packaging contributes greatly to environmental pollution. Replacing traditional plastics with biodegradable or reusable materials is urgent. Such changes are essential to promote sustainable consumption and protect the environment.

How?

- Characterizing curcumin modified by antisolvent precipitation.
- Producing, characterizing, and applying biodegradable gelatin films with modified curcumin in the preservation of bananas.
- Producing and characterizing bilayer packaging of paper coated with gelatin and modified curcumin.

What has already been researched?

- The use of bioactive compounds in food packaging has been widely studied lately.
- There are some works in the literature that use biopolymeric films bioactive compounds for application in food, one specifically, developed curcumin films with HPMC - There are only a few reports on the food application of bilayer packaging of paper coated with biopolymers to treat oily wastewater.
- There are no works, so far, on the studies investigating the combination of gelatin with MC in films or bilayer packaging from cellulose paper coated specifically with gelatin and modified curcumin; Thus, this is an unprecedented work.

Hypothesis

- It is possible to obtain modified curcumin at the nanometric scale, with enhanced properties compared to natural curcumin.
- Active films of gelatin and modified curcumin can be used in the preservation of bananas.
- It is possible to obtain bilayer packaging of cellulose paper coated with gelatin and modified curcumin.

CHAPTER 1
INTRODUCTION AND OBJECTIVES

1 INTRODUCTION

Packaging plays a crucial role in the preservation, safety and quality of food. It acts as a protective barrier against external factors such as moisture, light, oxygen and microbiological contamination, extending the shelf life of food and reducing waste. In addition, packaging ensures the integrity of products during transport and storage, facilitates handling and distribution, and provides essential information such as expiration date, ingredients and instructions for use. In a world increasingly aware of sustainability, packaging design is also essential to minimize environmental impact, encouraging the use of recyclable and biodegradable materials (BRIGHAM, 2018; HAN et al., 2018; MOHAMED; EL-SAKHAWY; EL-SAKHAWY, 2020).

Paper is a promising option among biodegradable materials because it is renewable, recyclable and economical. However, its porosity and low resistance to water, gases and oils limit its application as food packaging. To improve its barrier properties, paper can be coated with another class of biodegradable materials, the biopolymers (ALLOUI et al., 2011; BASAK; DANGATE; SAMY, 2024; LIMA et al., 2024). Biopolymers from renewable sources are sustainable alternatives to synthetic polymers. Gelatin coatings and films, in particular, stand out in food protection, preventing dehydration and degradation due to oxidation and light, thanks to their ability to form barriers against gases (BARANWAL et al., 2022; BRIGHAM, 2018; KRAŚNIEWSKA; GALUS; GNIEWOSZ, 2020; RUBINI et al., 2021).

Advances in the area of biopolymeric packaging include the incorporation of bioactive compounds into gelatin, providing additional functional properties. Among these compounds, curcumin, present in turmeric, has gained prominence for its antioxidant properties, beneficial both for human health and for food preservation. However, the bioactivity efficacy of curcumin is limited due to its low solubility and bioavailability, problems that can be mitigated by modifying its crystalline structure and particle size, which enhances its bioactive properties (RUBINI et al., 2021; SILVA et al., 2019; YEN et al., 2010).

Previous studies have examined the use of bilayer packaging composed of paper coated with biopolymers biopolymers (DOS SANTOS et al., 2022b; HEFFT, 2018; NAITZEL et al., 2023; ZHARKEVICH et al., 2023). Research has

also explored gelatin films incorporated with curcumin (BITENCOURT et al., 2014; ROY; RHIM, 2020; RUBINI et al., 2021), with Silva et al., (2019) specifically investigating the application of modified curcumin (MC) in HPMC films. However, to date, no studies have investigated the combination of gelatin with MC or explored the production of bilayer packaging from cellulose paper specifically coated with gelatin and MC.

1.1 OBJECTIVES

1.1.1 General Objective

This study aims to develop and characterize gelatin and gelatin-paperboard materials incorporated with modified curcumin for food packaging applications.

1.1.2 Specific Objectives

The specific objectives are presented below, separated by the chapters of the thesis.

Chapter 2

- To comprehensively analyze and classify the different biopolymers and methods used to coat paper(board), and to analyze the physicochemical properties and food applications of the resulting materials.

Chapter 3

- To produce MC and study its properties; to produce and evaluate gelatin films incorporating different MC concentrations; to assess the effectiveness of these antioxidant films for banana packaging.

Chapter 4

- To develop and evaluate bilayer materials based on paper and gelatin incorporated with modified curcumin for packaging application.

CHAPTER 2

LITERATURE REVIEW

2 LITERATURE REVIEW

This chapter presents the theoretical background and literature review for the thesis, with an emphasis on the use of cellulose-based packaging coated with biopolymers. The chapter forms part of a review article entitled “Progress in biopolymer coatings for achieving sustainable paper and paperboard packaging for food applications: A brief review.” published in the *Packaging Technology and Science* journal (DOI: 10.1002/pts.2861).

Author Contributions:

Kennya Thayres dos Santos Lima – manuscript writing.

Jéssica de Matos Fonseca – critical review of the manuscript.

Alcilene Rodrigues Monteiro – critical review of the manuscript.

Germán Ayala Valencia – conception of the idea, partial manuscript writing, and revision.

Abstract

Paper and paperboard are cellulosic materials used to manufacture food packaging. However, their low resistance to water and fat, as well as their poor barrier properties have limited their packaging applications as food contact materials. The current review analyzes the use of biopolymeric coatings as an alternative to improve surface and barrier properties in paper and paperboard with focus in food sector application. Advances in coated paper(board) can be classified into three groups: (1) coatings manufactured with a single biopolymer; (2) coatings manufactured with composite biopolymers; and (3) biopolymeric coatings incorporated with bioactive molecules or nanoparticles. The use of a single or composite biopolymer coating improved the mechanical, and barrier properties, and grease resistance of cellulosic materials/paper. In contrast, the use of compounds and nanoparticles has imparted active properties (antibacterial and ethylene scavenging) in the resulting coating paper(board). Furthermore, the water vapor transmission rate has been reduced with the use of biopolymeric coatings. Future studies should focus on applying coated paper(board) in foods with a special interest in food self-life, migration, and toxicity.

Keywords: Cellulose, coatings, food packaging, shelf-life extension.

2.1 INTRODUCTION

Paper and paperboard are materials based on cellulose whose difference is the grammage. While paper is an agglomerate of cellulose that forms a thin material (grammage < 250 g/m²), paperboard results from the aggregation of multiple layers to form thick materials (grammage > 250 g/m²) (DESHWAL; PANJAGARI; ALAM, 2019; TYAGI et al., 2021).

Approximately 55% of cellulose is used to manufacture paper(board) materials for packaging applications (HAGGITH et al., 2018). In particular, paper(board) is widely used for food or consumer products because of its structure, attractive shelf display, effective printable surface, versatility, low cost,

and easy recycling (ARCHAVIBOONYOBUL et al., 2014). Recently, this type of material has been appointed as a potential candidate to replace synthetic plastic in packaging applications (BAKKER et al., 2022a, 2022b).

Paper(board) has high porosity and hydrophilicity, making this material highly permeable to gasses (water vapor, oxygen, carbon dioxide, and nitrogen), permeable to oils, and extremely sensitive to water, thereby affecting its barrier and mechanical properties, and limiting its applications as food contact packaging (BASAK; DANGATE; SAMY, 2024; KHWALDIA; ARAB-TEHRANY; DESOBRY, 2010).

Paper(board) materials have an average biodegradation time of around 6 months. These materials are normally coated with synthetic polymers to reduce their sensitivity to water and oil. Unfortunately, synthetic polymers are not biodegradable (BASAK; DANGATE; SAMY, 2024; KUNAM et al., 2022; TYAGI et al., 2021). In this way, the use of biopolymers as coatings of paper(board) has been investigated in recent years (ARCHAVIBOONYOBUL et al., 2014; CHI et al., 2023; CHRISTOPHLIEMK et al., 2023; DOS SANTOS et al., 2022a, 2023a). The use of biopolymers has been appointed as an alternative to achieve the sustainable production of paper(board) food packaging considering the different Sustainable Development Goals of the United Nations (SDGs-UN) as “*Industry, Innovation and Infrastructure*” and “*Responsible Consumption and Production*” (DE SOUSA, 2021).

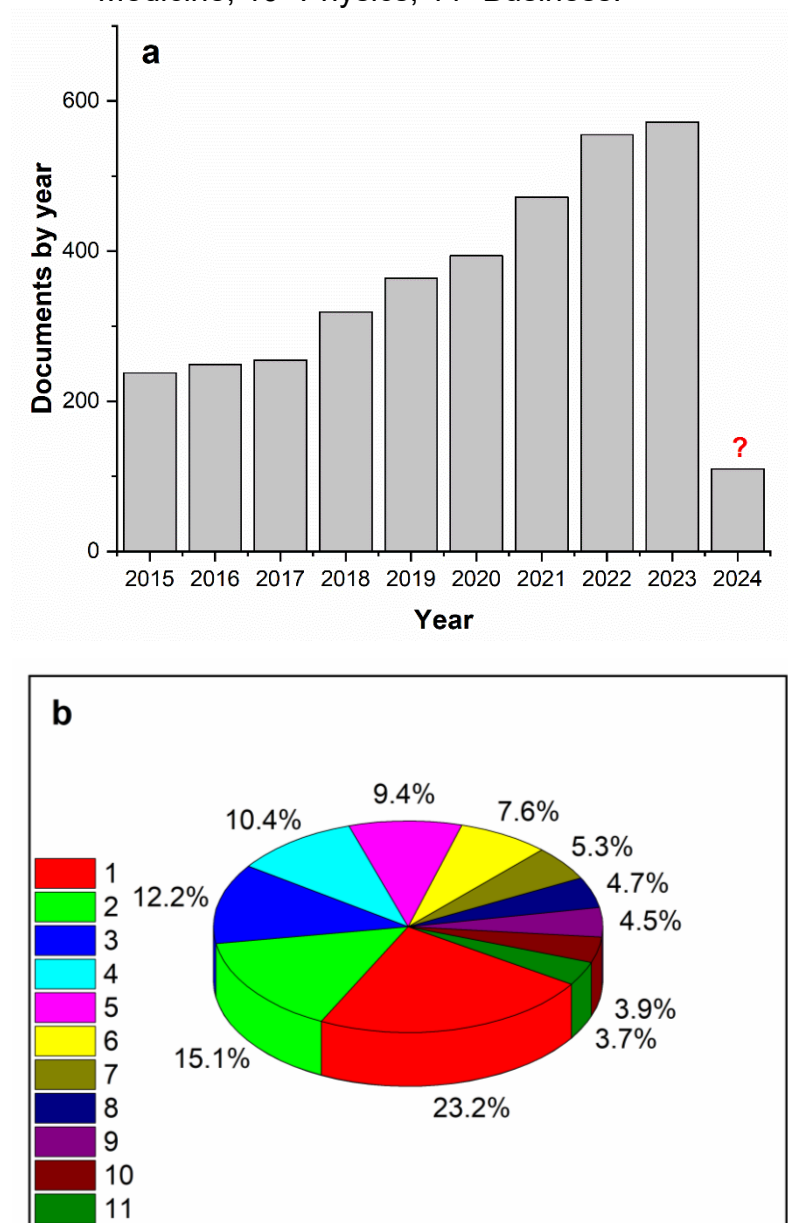
However, to the best of our current knowledge, no review paper has comprehensively analyzed and reviewed the use of biopolymers as coatings of paper(board) for food applications. Therefore, the objective of this review was to comprehensively analyze and classify the different biopolymers and methods used to coat paper(board), as well as to analyze the physicochemical properties and food applications of the resulting materials. The chosen database was Scopus for studies published in the last decade.

2.2 PAPERBOARD PRODUCTION AND PACKAGING APPLICATIONS

A search using the SCOPUS database revealed that the number of scientific manuscripts on “paper AND food AND packaging” has increased in the last ten years (Figure 1a). Agricultural and biological science followed by

engineering and materials science are the main areas where this type of manuscripts has been published (Figure 1b). These results confirm that research on cellulose materials (paper and paperboard) is an important topic, and most of these studies were published in areas related to food science.

Figure 1 – A search of the SCOPUS database for scientific production using the keywords “paper AND food AND packaging”: (a) Number of publications by year from 2015 to April 2024; (b) Document by area: 1- Others; 2- Agricultural and biological science; 3- Engineering; 4- Material science; 5- Chemistry; 6- Environmental sciences; 7- Chemical engineering; 8- Biochemistry; 9- Medicine; 10- Physics; 11- Business.



Source: prepared by the authors.

A new search using the same database and the keywords “biopolymer AND coating AND paper AND packaging” revealed 116 research papers in the last decade. Between them, only 20 manuscripts focused on developing biopolymeric coatings on paper(board) and some investigated their potential food applications. Therefore, these 20 manuscripts were chosen for the current review.

2.3 BIOPOLYMERIC COATINGS APPLIED ON PAPER(BOARD)

Biopolymers are natural polymers produced by living organisms and they can be classified into three categories: polymers from biomass, polymers synthesized through microbial routes, and polymers chemically synthesized using monomers from agro-resources (VIEIRA et al., 2011). In general, biopolymers can be transformed into water, carbon dioxide, methane, inorganic compounds, or biomass through the action of enzymes and microorganisms when these materials are disposed of in soils (POLMAN et al., 2021). Furthermore, some biopolymers have acceptable water, gas, oil, odor, and barrier (O_2 , CO_2 , H_2O , and UV-light) properties, ideal for coating paper and paperboard materials aiming at food contact applications (VALENCIA; SOBRAL, 2018; VIEIRA et al., 2011). Poly(lactic acid) (PLA), chitosan, starch and its derivatives, cellulose and its derivatives, and lignin have been the macromolecules most used to coat paper(board). The effect of each macromolecule on the resulting properties of the coated paper(board) will be discussed in this section. More detailed characteristics and/or properties of biopolymers can be found in the specialized literature (AWASTHI et al., 2022; POLMAN et al., 2021; VALENCIA; SOBRAL, 2018; VIEIRA et al., 2011).

2.3.1 Biopolymeric coatings

A single biopolymer can be applied in one or more layers as a coating on paper(board) to produce food packages with improved physical properties. Thus, Tanpichai et al. (2022) coated cellulose-based paper with up to 5 layers of chitosan-based coating and evaluated its wettability, mechanical properties, and

overall migration. The authors observed that chitosan filled the pores between the cellulose fibers and the addition of chitosan-based coating (1 – 5 layers) increased the grammage from 44 g/m² to 49 g/m² and did not alter the visual aspect or color of the resulting coated materials. Adding chitosan resulted in an improved hydrophobicity; hence, the water contact angle (WCA) increased from 66.7° (uncoated material) to 94.7° in coated materials. Furthermore, the presence of biopolymeric coatings reduced the migration of low-molecular-weight residuals from paper to food and enhanced the mechanical properties (tensile stress at break (TSB): 37.9 → 44.9 MPa and elongation at break (EB): 2.3 → 3.5%) of the paper. Unfortunately, coated materials had less thermal stability, and uncoated paper had a maximum decomposition temperature of 234 °C which decreased with the presence of chitosan to 225–229 °C. This behavior was due to the lower thermal stability of chitosan when compared to cellulose (TANPICHAI et al., 2022).

Table 1 summarizes the main physical properties of commercial synthetic plastics used in food packaging applications. Compared with the WCA results from Tanpichai et al. (2022), (TANPICHAI et al., 2022) it is possible to conclude that paperboard coated with chitosan has comparable WCA values to those obtained in commercial synthetic plastics.

Table 1 – Surface contact angle (WCA), water vapor transmission rate (WVTR), oxygen rate transmission (OTR), tensile strength at break (TSB), and elongation at break (EB) of commercial synthetic polymers used in food packaging.

Synthetic polymer	WCA (°)	WVTR (g/m ² ·day)	OTR (cm ³ /m ² ·day)	TSB (MPa)	EB (%)	Reference
LLDPE	92 – 96	1.2 – 5.9	70 – 550	26	553	(KURIHARA; HOKARI; TAKADA, 2021; LU; ZHENG, 2018; MÜLLER et al., 2022)
PP	101	1.5 – 5.9	93 – 300	33	150	(COLIN HINDLE, 2024; LU; ZHENG, 2018)
PET	74	0.5 – 6.0	2 – 20	55	600	(FADEL et al., 2020; MATWEB, 2024)

LLDPE: linear low-density polyethylene; PP: polypropylene, PET: polyethylene terephthalate. Source: prepared by the authors.

Dos Santos et al. (2022, 2023b) coated paperboard packaging material with a coat based on chitosan, palmitic acid, and activated carbon and characterized the mechanical properties, water vapor transmission, fat barrier, and biodegradation of these materials. The authors observed that the coating filled the pores of the cellulosic matrix and improved grease resistance (DOS SANTOS et al., 2022a, 2023b). Mechanical properties (TSB: 75.6 MPa and EB: 1.8 – 2.1%) remained constant and the water vapor transmission rate ($221 \rightarrow 160 \text{ g/m}^2\cdot\text{day}$) decreased with the incorporation of biopolymer coatings; however, the alteration of this property was not significant from a technological viewpoint (DOS SANTOS et al., 2022a, 2023b). Paperboard has higher TSB and lower EB values when compared with synthetic polymers (Table 1), suggesting that paperboard is more rigid than plastic materials.

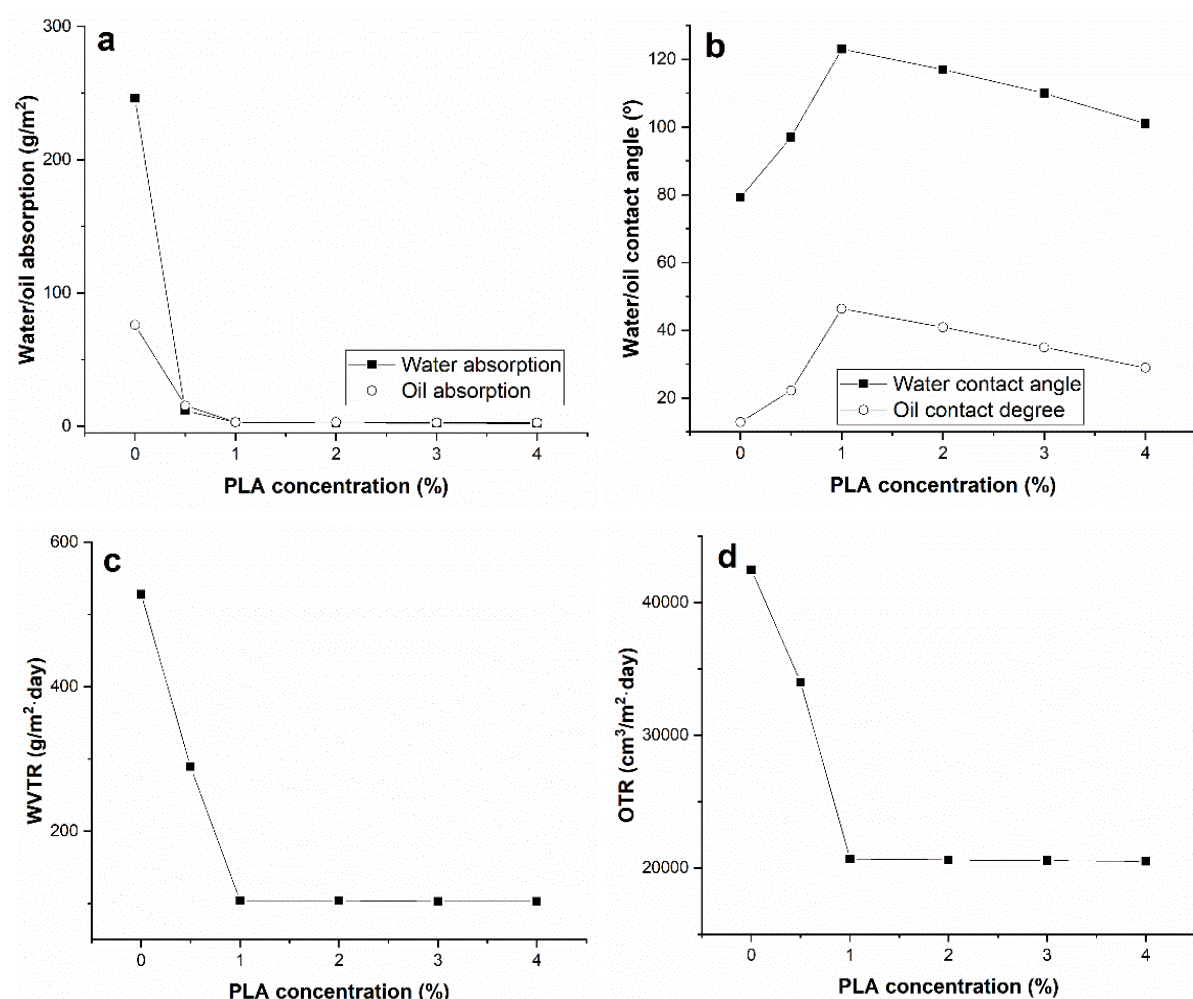
In another research, Jeong and Yoo (2020) applied a coating of whey protein concentrate-beeswax-sucrose on paperboard. It was observed that biopolymeric coatings provided good oil-barrier properties due to their polar characteristics, and beeswax provided good water-barrier properties (decreasing in water vapor permeability, WVP: $113 \rightarrow 46 \text{ g}\cdot\text{mm/m}^2\cdot\text{day}\cdot\text{kPa}$) due to their hydrophobic characteristics. The same authors concluded that the coating paperboard has the potential to replace uncoated paperboard in applications requiring water and oil contact (JEONG; YOO, 2020).

Currently, most of the research approaching coating biopolymers on paper or paperboard have been carried out at the laboratory-scale. Few works have been carried out on a larger scale. Mikriukova, Lahti and Kuusipalo (2022) compared the use of PLA, polybutylene succinate (PBS), and low-density polyethylene (LDPE) as paperboard coatings by extrusion on a pilot scale. Various natural oils were used to study the permeability of these polymers. The oils that showed the best properties as lubricants were cottonseed oil and castor oil. The utilized polymer coatings presented quite similar roughness $396 \pm 60 \text{ nm}$ for PLA, $198 \pm 60 \text{ nm}$ for PBS, and $425 \pm 60 \text{ nm}$ for LDPE (MIKRIUKOVA; LAHTI; KUUSIPALO, 2022).

With a similar approach, Archaviboonyobul et al. (2014) also performed the coating on paperboard with PLA. The authors used the electrospraying technique to reduce the amount of PLA in comparison with the conventional coating method. The authors applied five PLA concentrations (0.5, 1, 2, 3, and 4%) on paperboard and observed a reduction in water and oil absorption, as well as of water vapor and oxygen permeation, both with the increase of PLA concentration (Figure 2)

(ARCHAVIBOONYOBUL et al., 2014). WCA results are comparable to those found in commercial synthetic polymers used in food packaging (Table 1), however, permeability values (WVTR and OTR) of paperboard coated with PLA are higher when compared with the same synthetic materials (Table 1).

Figure 2 – **(a)** Water/oil absorption; **(b)** Water/oil contact angle; **(c)** Water vapor transmission rate (WVTR); and **(d)** Oxygen transmission rate (OTR) of paperboard coated with PLA. This graphic was constructed based on the results informed by Archaviboonyobul et al. (2014)



Source: prepared by the authors.

In another study, multilayered paper based on nano-silica and micro-fibrillated cellulose was developed by coating. The coated paper had a dry tensile strength and a remarkable wet tensile strength of 6542.5 N/m and 5875 N/m, increasing by 56% and 2277%, respectively. The authors concluded that multilayered paper with

acceptable mechanical properties and low permeability of air ($3.17 \times 10^{-3} \mu\text{m}/\text{Pa}\cdot\text{s}$), OTR ($0.97 \text{ cm}^3/\text{m}^2\cdot\text{day}$), and WVTR ($378.24 \text{ g}/\text{m}^2\cdot\text{day}$), respectively, as well as a superhydrophobic performance with a contact angle of $\sim 151.2^\circ$, was produced and has potential for food packaging applications (CHEN et al., 2022). At the same research, the OTR value was lower when compared with the same value informed in synthetic polymers (Table 1).

Based on these research studies, it is possible to conclude that the use of biopolymer coatings in paperboard principally improved WCA and OTR when compared with commercial plastics (Table 1). However, WVTR remains the main problem in the paper(board) materials.

2.3.2 Composite biopolymeric coatings

Composites coatings are composed of at least two materials, continuous and dispersed phases, that contribute to the final material properties. Thus, biopolymer-coated paper(board) exhibits properties of both materials. Naitzel et al. (2023) coated paperboard using the tape casting technique with different ratios of agar–agar/chitosan and different numbers of coating layers. The authors observed that paperboard coated with chitosan had higher grease resistance, which increased from 3 to 11 (kit number according to the TAPPI pm-96.27) after coating with composite coatings. Furthermore, WVP decreased from 7.2 to 3.1 $\text{g}\cdot\text{mm}/\text{m}^2\cdot\text{day}\cdot\text{kPa}$ with the presence of the coating material. In the same research, other properties such as water absorption capacity ($36\text{--}46 \text{ g}/\text{m}^2$) and mechanical properties (TSB: 22–25 MPa; EB: 3.0–3.5%) remained constant in coated paperboard. Regardless of the number of layers, the coatings showed homogeneity and improved the grease resistance of the material (NAITZEL et al., 2023).

Biodegradable thermoplastics (PLA, polybutylene adipate terephthalate and polybutylene succinate together) were used to coat nanocellulose-based multilayer-barrier paperboard (microfibrillated cellulose or carboxymethylated cellulose nanofibrils) by extrusion coating. In this research, it was observed that WVTR and OTR of the coated paperboard decreased from approximately 446 to 62 $\text{g}/\text{m}^2\cdot\text{day}$ and 0.81 to 0.31 $\text{cm}^3/\text{m}^2\cdot\text{day}$, respectively (KOPPOLU et al., 2023). WVTR and OTR are similar to those informed in synthetic polymers (Table 1).

A study examined improving the contaminant barrier properties of recycled paperboard by coating packaging materials with starch and sodium alginate mixtures. The substances that simulated contaminants tested were undecane, heptadecane, dodecane, triethyl citrate, dipropylphthalate, 4-methylbenzophenone, and cholestane. Films based on starch (90% of the total weight of polymers) and sodium alginate (10% of the total weight of polymers) exhibited good barriers against the penetration of test substances simulating the contaminants from paperboard and presented good barrier properties against the penetration of mineral oils (ZHARKEVICH et al., 2023).

Recently, Shorey and Mekonnen (2022) investigated paper coating with esterified lignin and a poly(butylene adipate-co-terephthalate) (PBAT) blend. For this purpose, pure and esterified lignin in different concentrations was incorporated into PBAT solutions and applied to paper substrates. Coated paper had good uniform surface morphology and improved mechanical (TSB: 5 → 20 MPa; EB: 3 → 4%), surface hydrophobicity (WCA: 80 → 130°), oxygen permeability (5.5 → 1.0 cm³·m/m²·day·Pa), and oil resistance (Kit number: 0 → 5) (SHOREY; MEKONNEN, 2022).

Industrial byproducts that are otherwise wasted can be used as a source of extraction compounds for application in food packaging. Poulouse et al. (2022) coated paperboard with potato fruit juice by rod coating and then taped it to a paper roll and then coated it with PLA or a blend of PLA and poly(butylene adipate terephthalate) using a pilot plant extrusion coater. The developed coated paperboard showed good creasing and reduced oxygen permeability (OTR: 98.69 → 2.96 cm³/m²·day·atm) and WVTR (266 → 11 g/m²·day) (POULOSE et al., 2022). WVTR and OTR are similar to those informed in synthetic polymers (Table 1).

Similar to paper(board) coated with a coating composed of a single biopolymer, the use of composite coatings also improved surface hydrophobicity and reduced the gas barrier in the resulting materials.

3.3. Biopolymeric coatings containing nanomaterials and active compounds

Nanomaterials and active compounds can be incorporated into biopolymeric coatings paper(board) to produce active food packaging, thereby preserving the quality

of food, and improving its shelf-life. A blend of anionic-starch-based nanocellulose/chitosan was used to coat offset paper and kraft paper. The results revealed that the biopolymer coating forms a dense film layer on the paper surface and fills the surface pores. The coating had good antibacterial properties (no mold at the end of the 60-day observation period) and significantly improved the air permeance ($12.20 \rightarrow 6.9 \times 10^{-3} \mu\text{m}/\text{Pa}\cdot\text{s}$) and oil resistance ($446.5 \rightarrow 7.1 \text{ g}/\text{m}^2$) of the paper. The tearing resistance and tensile strength of the coated kraft paper significantly improved, increasing 19.1% vertically and 10.9% horizontally, respectively (XUE et al., 2022).

Zhang et al. (2016) studied the recycling process of PLA-coated paper containing zinc oxide nanoparticles (ZnO) and characterized the material on a laboratory scale. The results obtained reveal that 99% of the solid material was recovered. The brightness value of processed materials decreased as a function of ZnO loading in the coating. The water absorption decreased ($24.2 \rightarrow 3.4 \text{ g}/\text{m}^2$) and the water contact angle increased ($70^\circ \rightarrow 96^\circ$) both with the incorporation of ZnO.

In another research, Wang et al., (2022) investigated the barrier properties and antibacterial properties of paperboard used for fast food packaging coated with multilayers of biopolymers (carboxymethyl chitosan/carboxymethyl cellulose sodium + PLA) and ZnO. Compared with a single coating layer, the composite coatings showed better oxygen, oil penetration, and water vapor barrier properties. The multilayer coating with 1.5% added of nano zinc oxide reduced the growth of bacteria (*Escherichia coli* and *Staphylococcus aureus*) by more than 99%.

Industrial byproducts can be used to extract active compounds for application in active food packaging. In this context, Saini et al. (2023) extracted nano-silica isolated from rice straw to study its ethylene gas scavenging properties and applied it to polyvinyl alcohol (PVA)-based coated on duplex paperboard made from softwood and rice straw. In this study, the size of the obtained silica particles was in the range of 60–80 nm. The nano-silica/PVA formulation showed ethylene scavenging activity and the volume of ethylene sorbed by the coated paperboard was higher than that of the uncoated paperboard in 12 days (ethylene scavenging: approximately 8 and 40 ppm in uncoated and coated materials, respectively) (SAINI et al., 2023). In another research, Winstrand et al. (2021) also developed a material for gas absorption; for this purpose, laccase-catalyzed oxygen-scavenging fractionated lignosulfonates for cardboard coating were prepared. The enzymatic reaction increased in the water

contact angle of the films from the range 25–49° to 56–81°. Fractionation of lignosulfonates had little effect on oxygen scavenging, but improved cross-linking, resulting in higher molecular weight and superior water resistance.

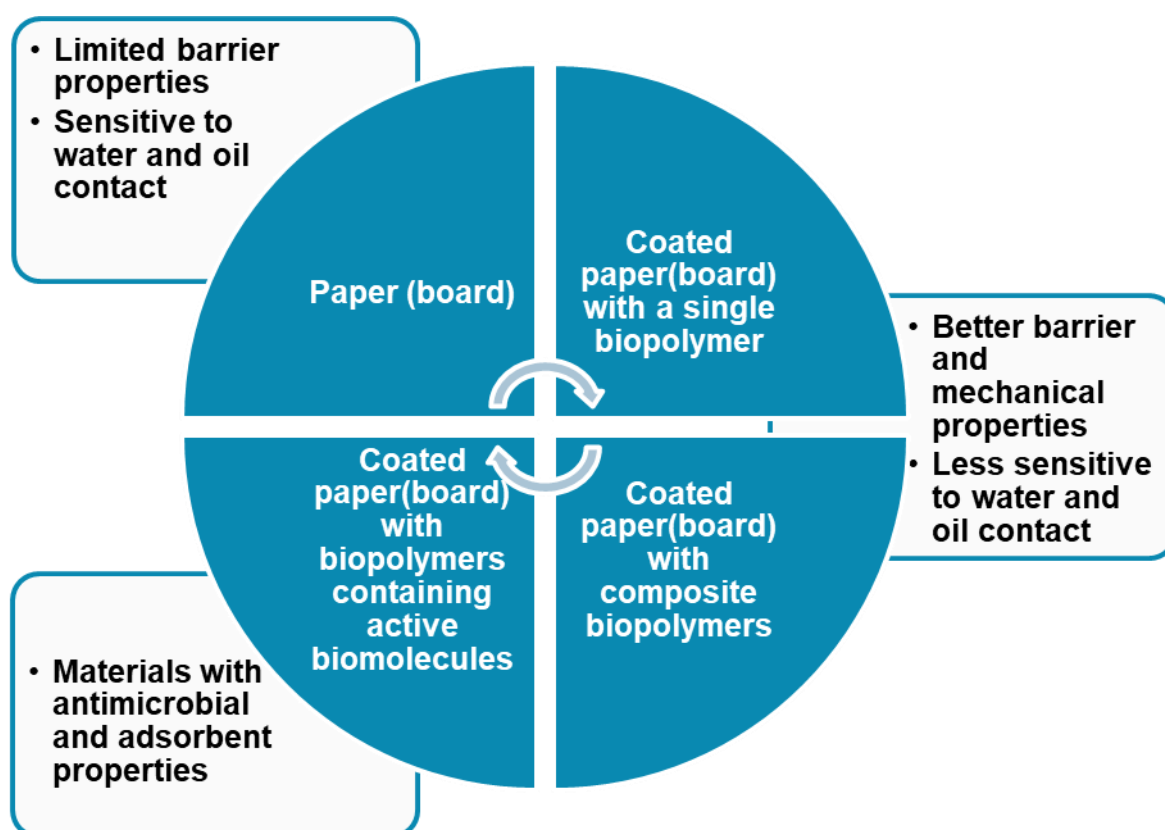
Cardboard coated with biopolymers and bioactive compounds can be used for food preservation. Lavoine et al. (2015) developed an antibacterial bio-packaging material using a chlorhexidine digluconate solution as a model of an antibacterial molecule, mixed it with microfibrillated cellulose, and used it as a coating on paperboard samples. For the application of this work, active boxes were made with the developed material, and similar pieces of pork liver were placed in contact with the coated side of the paperboard box. Packaging materials showed a lower proportion (20%) of chlorhexidine digluconate released when using microfibrillated cellulose, whereas constant release of low amounts of chlorhexidine digluconate over 40 h was observed. The samples remained active against *Bacillus subtilis*, with better results being obtained when microfibrillated cellulose was used. The coated paperboard boxes could better preserve the freshness of the pork (LAVOINE et al., 2015). Hefft (2018) developed 2% chitosan-coated paperboard trays and studied their effects on the botrytis cinerea formation and the sensory quality of bilberries (*Vaccinium myrtillus* L.) and redcurrants (*Ribes rubrum* L.). The coated material has a preventive effect on the growth of *Botrytis cinerea* and helped to extend the shelf life of bilberries by approximately 2 days and for redcurrants by up to 4 days. Sensory evaluation showed that the coated materials did not have a significant impact on the odor, berry appearance, aroma, or flavor.

2.4 FUTURE TRENDS

Based on the consulted literature, it is possible to conclude that the physical properties of paper(board) can be improved by coating the surface of these materials with biopolymers (Figure 3). Furthermore, advanced properties, such as antimicrobial and oxygen scavenging properties, can be incorporated in paper(board) by using biomolecules and nanoparticles (Figure 3). However, water vapor and oxygen permeability remain to be the main parameters limiting food packaging applications of coated paper(board) when compared with synthetic polymers used in food packaging applications (Table 1). Future research studies must be conducted to improve the permeability of coated paper(board). In this scenario, crosslinking and reactive

extrusion of biopolymers could be an alternative (ELLINGFORD et al., 2022; GARAVAND et al., 2017; HERNIOU--JULIEN; MENDIETA; GUTIÉRREZ, 2019), and the use of nanoparticles to increase the tortuosity of coating materials (PIRES et al., 2021; SOUZA et al., 2022). Biopolymer-coatings could improve heat sealing of paper(board) since these macromolecules can be melted at temperatures below 100 °C (VALENCIA; SOBRAL, 2018; VIEIRA et al., 2011). In this way, this property must be investigated in future research studies.

Figure 3 – Advances in the resulting paper(board) materials coated with biopolymers containing or not biomolecules and nanoparticles.



Source: prepared by the authors.

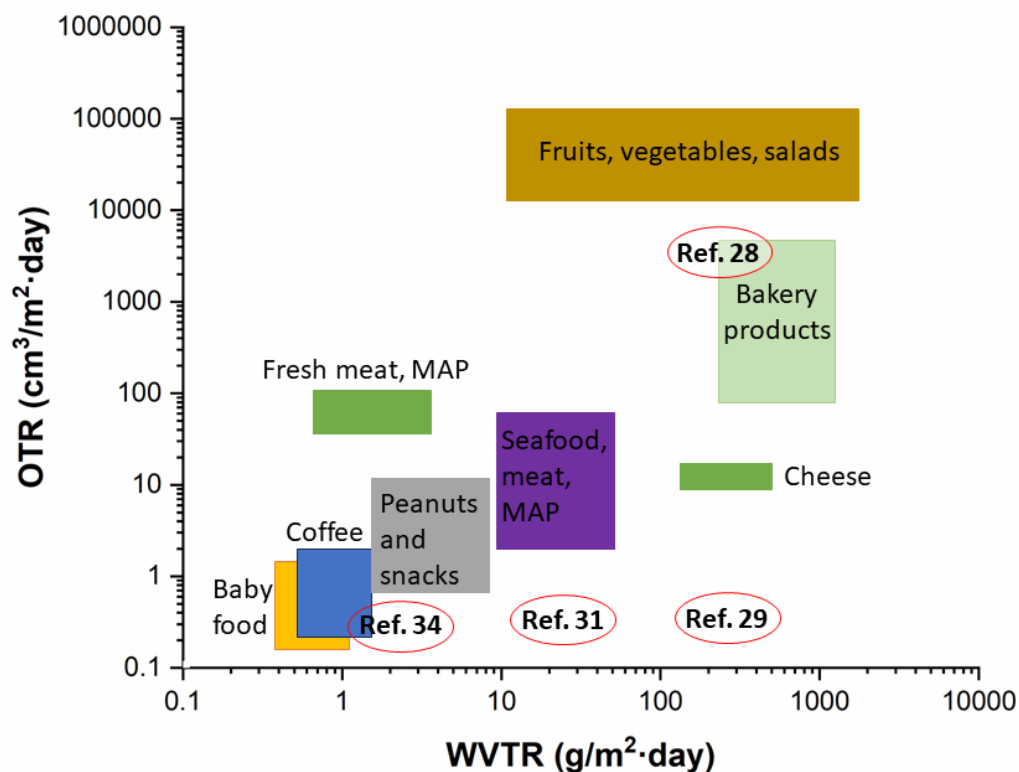
Similarly, the coating process can modify the biopolymer deposition and then the physicochemical properties of the resulting coating materials (RHEE; WHITE, 2022). Therefore, it is necessary to compare the coating production method (e.g. casting, spread casting, electrospinning, extrusion, among others) with the resulting properties of the coated paper(board) materials.

Concerning the production of paper(board) materials coated with a single or composite biopolymer coating, it is necessary to evaluate the coating properties of

each biopolymer systematically. In particular, the WVTR and OTR of coated paper(board) must be investigated in real atmospheric conditions for food packaging applications. A few papers have investigated both barrier properties (WVTR and OTR). In the current review, the WVTR and OTR values from the studies conducted by Archaviboonyobul et al. (2014), Chen et al. (2022), Koppolu et al (2023) and Poulouse et al. (2022) are plotted in Figure 4 (see red circles inserted in this figure) (TRINH; CHANG; MEKONNEN, 2023). The WVTR and OTR values informed by Archaviboonyobul et al. (2014) and Chen et al. (2022) are typical of paper(board) materials. In other words, the use of biopolymeric coatings did not improve these barrier properties from a technological viewpoint (TRINH; CHANG; MEKONNEN, 2023). However, a substantial reduction in WVTR was observed when PLA and polybutylene adipate terephthalate were used to coat paper(board) (KOPPOLU et al., 2023; POULOSE et al., 2022). This important reduction opens a window for promissory food packaging applications of coated paper(board) (Figure 4).

A limited number of research studies have applied coated paper(board) materials in food. Therefore, future research studies must apply these coated materials in food packaging to elucidate the potential application of these materials, as well as quantifying the toxicity and migration of chemical compounds from these materials in food simulants and food formulations.

Figure 4 – Water vapor transmission rate (WVTR) *versus* oxygen transmission rate (OTR). Requirements for foods using individual polymers.



Source: prepared by the authors, adapted from Trinh, Chang and Mekonnen (2023).

Finally, it is expected that the incorporation of biopolymer in paper(board) does not alter the biodegradability of the resulting coated material, therefore, it was not discussed in this literature review. However, the presence of other polymeric materials may make paper(board) recycling more difficult and expensive.

2.5 CONCLUSIONS

Paper(board) has limited packaging applications as a food contact material due to its poor barrier and surface properties, which make this material sensitive to water and fat. The incorporation of biopolymeric coatings on the paper(board) surface has increased surface hydrophobicity and grease resistance. Mechanical properties (TSB and EB) have also been improved with the incorporation of biopolymeric coatings. In general, biopolymers fill the pores of paper(board), improving the barrier and mechanical properties. Furthermore, the incorporation of chlorhexidine

digluconate and nanoparticles (ZnO and nanosilica) can impart antibacterial and ethylene scavenger properties in the coated paper(board). The properties of the developed coated paper(board) materials are promising for new packaging applications as food contact materials.

2.6 ACKNOWLEDGEMENTS

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CHAPTER 3

ACTIVE FILMS BASED ON GELATIN AND MODIFIED CURCUMIN: PHYSICOCHEMICAL PROPERTIES AND FOOD PACKAGING APPLICATION

3 ACTIVE FILMS BASED ON GELATIN AND MODIFIED CURCUMIN: PHYSICOCHEMICAL PROPERTIES AND FOOD PACKAGING APPLICATION

This chapter presents the results obtained from the characterization of MC, the characterization and application results of gelatin films incorporated with curcumin for banana preservation. The chapter forms part of a article entitled “Active films based on gelatin and modified curcumin: physicochemical properties and food packaging application” published in the Polymers for Advanced Technologies journal (DOI: 10.1002/pat.70123).

Author Contributions:

Kennya Thayres dos Santos Lima – contributed to nanoparticle production, films preparation, data processing and analysis, film application, and manuscript writing.

Vinicius Nogueira Bergamo –contributed to nanoparticle production, film preparation and film application.

Gabriel Coelho Lerandro – operated the SEM equipment for film analysis and assisted in producing three film units used in the food application.

Jéssica de Matos Fonseca – critical review of the manuscript.

Alcilene Rodrigues Monteiro – critical review of the manuscript.

Germán Ayala Valencia – conception of the idea, project supervision, and manuscript revision.

Abstract

This study investigated the development of active gelatin films containing curcumin nanoparticles and their application to reduce banana browning. Curcumin, a natural antioxidant with limited water solubility, was modified through antisolvent precipitation aiming at increased water solubility and higher antioxidant activity. Gelatin films with different MC concentrations (0.0, 0.1, 0.3, 0.5, and 1%) were produced by the casting method. The resulting films were tested for thickness, morphology, crystallinity, chemical bonds, water absorption, water solubility, optical properties, wettability, water vapor permeation, mechanical properties, and antioxidant activity. Furthermore, films were applied as banana packaging. Structural (FTIR, Raman, and XRD) and morphology (SEM) analysis confirmed that MC was well-incorporated with an uniform distribution, and no alteration on film's thickness, permeability, wettability, or mechanical properties was observed. The resulting films had improved antioxidant activity (DPPH and ABTS assays) as a function of MC concentration. Gelatin films containing 1% MC reduced banana browning during the storage of this fruit at 25 °C and 75% RH for 6 days. These findings highlight the potential of gelatin films containing MC as a sustainable bioactive packaging for maintaining the banana's visual aspect.

Keywords: antioxidant films, antisolvent precipitation, banana browning, nanocomposite films.

3.1 INTRODUCTION

Packaging is important for the food processing, preservation, supply and distribution chain. Its main function is to contain and protect food material, but it is also essential to ensure food quality and safety, while also extending shelf life and minimizing food loss and waste (ANUKIRUTHIKA et al., 2020; HAN et al., 2018). Traditionally, the industry has relied on synthetic polymers, such as polyethylene and polypropylene, due to their effectiveness in protecting food and extending its shelf life. However, growing concerns about environmental impact and sustainability have driven the search for more eco-friendly alternatives, leading to

the development of biopolymeric packaging (BHUNIA et al., 2013; LEI et al., 2014; MOHAMED; EL-SAKHAWY; EL-SAKHAWY, 2020; SIRACUSA et al., 2008).

Aiming to reduce environmental problems, biopolymers, derived from renewable resources, have shown promise as substitutes for synthetic polymers, offering a more sustainable and biodegradable solution. Among various biopolymers, gelatin films have been applied to protect foods from drying and reduce food degradation by oxidation and light because this macromolecule can form effective barriers against gases. Gelatin films have the advantage of being naturally biodegradable and highly compatible with food matrices (ANUKIRUTHIKA et al., 2020; CHACON et al., 2024; ELSABEE; ABDOU, 2013; TRINETTA, 2016).

In parallel, innovation in biopolymeric packaging has led to the enhancement of gelatin films through the incorporation of bioactive compounds, which can provide additional functional properties (RAWDKUEN et al., 2020; SILVA E SILVA et al., 2018). Among these bioactive compounds, curcumin found in turmeric has gained attention due to its antioxidant properties, which are beneficial both for human health and for food preservation. However, its bioactivity efficacy is often limited due to its low solubility and bioavailability. An effective strategy to address this limitation is to modify curcumin's crystalline structure and particle size, which enhances its solubility and bioavailability, also improving its bioactive properties (CHOPRA et al., 2021; SILVA et al., 2019; YEN et al., 2010).

Some studies in the literature have explored the production and use of gelatin films containing curcumin (BITENCOURT et al., 2014; ROY; RHIM, 2020; RUBINI et al., 2021); while Silva et al. (2019) investigated the application of MC in HPMC films. However, there have been no studies investigating the combination of gelatin (gelatin films are less hygroscopic compared to HPMC films) with MC.

Therefore, this research aimed to produce MC and study its properties, as well as to produce and evaluate gelatin films incorporating different MC concentrations, as well as to elucidate potential food packaging applications. The research aims not only to develop a more efficient packaging solution but also to contribute to the advancement of knowledge in the fields of biopolymeric packaging and bioactive compounds.

3.2 MATERIALS AND METHODS

3.2.1 Materials

Curcumin (Neon, Brazil) was used as the antioxidant ingredient, type A gelatin (260 Bloom and 30 mesh) (Gelnex, Brazil) and glycerol P. A. (Neon, Brazil) were used as the biopolymer matrix and plasticizer, respectively. Distilled water was used as the solvent of films, whereas ethanol ($\geq 99.5\%$, Neon, Brazil) was used as solvent to produce MC.

3.2.2 Production of Modified Curcumin

MC was obtained after the precipitation of curcumin using ethanol and water (antisolvent precipitation method). Unmodified curcumin (50 mg) was dissolved in 25 mL of absolute ethanol and dripped into 75 mL of distilled water under agitation of 7000 rpm, using a high-speed homogenizer (Ultra-Turrax T25, IKA) at room temperature for 15 min. The resulting solution was dried in a hood at room temperature and then on desiccators containing silica gel to completely remove water. The obtained MC powder was stored at 10°C in the absence of light (YEN et al., 2010).

3.2.3 Characterization of curcumin and its polymorphs

3.2.3.1 *Morphology and particle size*

Microstructure of powders (curcumin and MC) was analyzed using a scanning electron microscope (SEM, JSM-6390LV, JEOL®, Japan) with an accelerating voltage of 10 kV at different magnifications. Samples were fixed on aluminum stubs using a double face tape and coated with a thin gold layer before SEM analyses (CAPELLO et al., 2019).

Powder dispersions (0.03 % w/v) were determined with a Malvern Zetasizer Nano ZS (Malvern Instruments Ltd., GB) by the Dynamic light scattering (DLS). The dispersions were previously sonicated for 30 minutes in an ultrasonic bath, using Milli-Q water as a solvent, in order to avoid particle agglomeration (LIMA et al., 2021).

3.2.3.2 *Crystalline structure and chemical bonds*

Powders were analyzed using an X-ray diffractometer (Rigaku MiniFlex 600 DRX, Japan) equipped with Cu-K α radiation ($\lambda = 0.154056$ nm) in the scattering range of $3^\circ < 2\theta < 40^\circ$ with 0.05 of step size and scan speed of $10^\circ \text{ min}^{-1}$ (CHACON et al., 2024).

Chemical bonds of powders were analyzed using a Fourier-Transform Infrared spectrometer (FTIR, Cary 600, Agilent, U. S) in the infrared region between 4000 and 400 cm^{-1} , using 4 cm^{-1} resolution with 32 scans and KBr pellets. Furthermore, Raman spectroscopy measurements of powders were performed using Anton Paar Cora 5200 Raman (Austria) with an excitation wavelength of 785 nm employing laser power of 100 mW from 100 to 2250 cm^{-1} (CHACON et al., 2024).

3.2.3.3 *Solubility in water and water activity*

To determine the solubility (S) of powders in water, 0.008 g (W_o) of the sample was dissolved in 10 mL of distilled water and stirred for 24 h at 250 rpm at room temperature. Subsequently, the samples were filtered using a $0.45 \mu\text{m}$ filter membrane and dried in oven at 40°C for 48 h before being weighed (W_r) (CHACON et al., 2021). S was determined using Equation (1):

$$S (\%) = \frac{W_o}{W_r} \times 100 \quad (1)$$

Water activity (a_w) of powders were determined at room temperature using a water activity meter (Aqualab Model Series 3 instrument, Decagon Devices Inc., Pullman, WA, USA) (CHACON et al., 2021).

3.2.3.4 *Color*

Powder color characterization was carried out using a colorimeter (Delta Vista 450G, Delta Color, Brazil) based on the CIELab coordinates, namely, L^* (lightness

index), a^* (color tonalities from red “+” to green “-”), and b^* (color tonalities from yellow “+” to blue “-”). The color difference (ΔE) was calculated using Equation (2) and the color coordinates of untreated curcumin as the standard (CHACON et al., 2024).

$$\Delta E = \sqrt{(L_{sample}^* - L_{standard}^*)^2 + (a_{sample}^* - a_{standard}^*)^2 + (b_{sample}^* - b_{standard}^*)^2} \quad (2)$$

3.2.3.5 Antioxidant activity

The antioxidant activity of powders was evaluated using DPPH (2,2-diphenyl-1-picrylhydrazyl) free radical scavenging assay. In summary, 0.1 mL of sample (400 µg/mL) was mixed with 3.9 mL of 0.1 mM methanolic solution of DPPH. Absorbance was measured at 517 nm after 30 min in the dark at ambient temperature. Standard curve was obtained using ascorbic acid as the probe element. The results were expressed in µg/mL of ascorbic acid equivalent (ALVES et al., 2024). The percentage of DPPH free radical quenching activity was calculated using Equation (3):

$$DPPH (\%) = \left(\frac{AA_{DPPH} - S_{DPPH}}{AA_{DPPH}} \right) \times 100 \quad (3)$$

where AA_{DPPH} is the equivalent amount of ascorbic acid (µg/mL) and S_{DPPH} is the amount of sample (µg/mL) used.

The assay based on the capture of ABTS (2,2'-azino-bis(3-ethylbenzothiazoline-6-sulfonic acid) by antioxidants was also performed on the samples. Briefly, for the preparation of the ABTS radical 5 mL of solution of 7 mM ABTS in water was mixed with 88 µL of 140 mM potassium persulfate solution and kept in the dark at room temperature for 16 h. Subsequently, the mixture was diluted with ethanol to an absorbance value of $0.70 \pm 0,05$ at 734 nm. 30 µL of sample was added to 3 mL of the ABTS radical solution and the blend was incubated for 5min in dark at room temperature. Afterwards, the absorbance reading was performed at 734 nm. Standard curve was obtained using ascorbic acid as the probe element (ALVES et al., 2024). The results were expressed in µg/mL of ascorbic acid equivalent using Equation (4):

$$ABTS (\%) = \left(\frac{AA_{ABTS} - S_{ABTS}}{AA_{ABTS}} \right) \times 100 \quad (4)$$

where AA_{ABTS} is the equivalent amount of ascorbic acid ($\mu\text{g/mL}$) and S_{ABTS} is the amount of sample ($\mu\text{g/mL}$) used.

3.2.4 Production of composite films

Gelatin films containing 0, 0.1, 0.3, 0.5, and 1 % (g /100 g of gelatin) of MC were produced by the casting method. Initially, gelatin (4 g of biopolymer/100 g of solution) was hydrated for 30 min in 50 mL of distilled water and then the hydrated gelatin was gelatinized at 70 °C for 15 min under constant agitation (300 rpm) in a water bath. In parallel, glycerol (30 g /100 g of gelatin) together with the MC was mixed with water for 15 min in a magnetic stirrer (200 rpm). Then, the solution containing glycerol and MC was added to the gelatin solution, and the resulting film-forming solution (FFS) was mixed (300 rpm) at 70 °C for 15 min, followed by stirring with an ultra-turrax at 7000 rpm for 10 min and finally sonicated for 1 h at 40 KHz and 35 °C in a sonicator bath. At last, it was obtained homogenous FFS with different MC concentrations. Approximately 10 g of FFS were poured into Petri dishes (9 cm diameter) and dried in a convective oven at 25 °C for 48 h. The resulting films were conditioned into desiccators with controlled relative humidity (RH = 58 %) at 25 °C before characterization (CHACON et al., 2024).

3.2.5 Characterization of composite films

3.2.5.1 *Thickness and morphology*

Film thickness was measured using a micrometer (Mitutoyo N°293-561-30, Japan) and informed as the average of five different sampling points in each film done in triplicate (CHACON et al., 2024).

The surface and transversal fracture microstructures (liquid nitrogen) of the films were analyzed using Scanning Electron Microscopy (SEM, JSM-6390LV, JEOL®, Japan) with an accelerating voltage of 10 kV. Films were fixed on aluminum stubs by carbon tape and then coated with a thin gold layer before SEM analyses (CAPELLO et al., 2021).

3.2.5.2 *Crystalline structure and chemical bonds*

Film crystalline structure (XRD) and chemical bonds (Raman spectroscopy) were analyzed using the same equipment and protocols as in Section 3.2.3.2. FTIR spectra of films was obtained using a Fourier-transform infrared spectrometer operating with a diamond single reflection attenuated total reflectance device (ATR-FTIR, Bruker, Alpha, UK) in the infrared region between 4000 and 400 cm^{-1} and resolution of 4 cm^{-1} (CAPELLO et al., 2021).

3.2.5.3 *Water absorption and solubility in water*

Water absorption (W_a) and water solubility (W_s) of the films were measured according to Chacon et al. (2024). Films were cut into circular pieces, weighed (W_i), and dehydrated into desiccators containing silica for 24 h. Dried films were immersed in 50 mL of distilled water at 25 °C and stirred using a shaker at 120 rpm for 24 h. Afterwards, films were removed from the water with tweezers, the excess of water was drained off by placing them on a sieve, and the wet weight (W_f) was recorded. The W_a was calculated using Equation (5):

$$W_a (\%) = \frac{W_f - W_i}{W_i} \times 100 \quad (5)$$

After immersion, the swollen films were dried at 110 °C for 24 h, transferred to a desiccator with silica, cooled to 25 °C and weighed (W_d). The W_s was calculated using Equation (6):

$$W_s (\%) = \frac{W_i - W_d}{W_i} \times 100 \quad (6)$$

3.2.5.4 *Optical properties*

Film color characterization was carried out using the same equipment and protocol as in Section 3.2.3.4. The ΔE was calculated using Equation (2) and the color coordinates of gelatin films without MC as the standard.

Film light barrier properties were analyzed using UV-Vis spectrophotometer (HITACHI U-1900, Japan). Films were fixed using a quartz cuvette in such a way that the light beam passed through the film and measurements at different wavelengths (200 – 800 nm). Film transmittance to visible light was measured at 470 nm (T_{470nm}) (CHACON et al., 2024). Furthermore, film opacity (Op) was calculated measuring film absorbance at 600 nm and its thickness, as follows:

$$Op = \frac{Abs(600\text{ nm})}{Thickness} \quad (7)$$

3.2.5.5 *Water contact angle*

Water contact angle (WCA) was measured as proposed by Chacon et al. (2024) and using an optical tensiometer (RameHart 250, US) operating with a DROPimage Advanced v2.2 software. Films were fixed with double face tape in microscope slide and $\approx 5\ \mu\text{L}$ drop of distilled water was released onto the air surface of films. Contact angle measurements were made by analyzing the shape of each sessile drop after it had been placed over the films for 10 s.

3.2.5.6 *Water vapor permeability*

Water vapor permeability (WVP) of films was determined through gravimetric analysis at 25 °C (CHACON et al., 2024). Films were conditioned into desiccators containing silica gel for 24 hours before use and their thickness was measured. Aluminum capsules with an area of 31 cm² were utilized, each containing silica gel (RH $\approx 0\%$). These capsules were then sealed with the films and subsequently, they were placed into a desiccator containing distilled water (RH $\approx 100\%$). The capsules were periodically weighed during 120 h and WVP was calculated using Equation (8).

$$WVP = \frac{\Delta g}{\Delta t} \left(\frac{d}{A \times \Delta P} \right) \quad (8)$$

where $\frac{\Delta g}{\Delta t}$ represents the increase in weight of the capsule during the testing period, d is the films thickness (mm), A denotes the area available for water vapor permeation

(m²), and ΔP indicates the difference in partial pressure across the film (3.169 kPa at 25 °C).

3.2.5.7 *Mechanical properties*

Mechanical properties of films were determined using a TA.HD.plus texturometer (Texture Analyser, Stable Micro Systems, UK) at 25 °C. For conducting the tensile test, the initial distance between the grips was adjusted to 20 mm, and the film samples (1.5 × 4 cm) were analyzed at a velocity of 1 mm s⁻¹. The elastic modulus (EM), tensile strength (TS), and elongation at break (EB) were determined from mechanical curves, with a minimum of 20 samples per film. The EM values were derived from the slope of the mechanical curves within the elastic region, whereas TS and EB values were calculated using Equation (9) and Equation (10), respectively:

$$TS = \frac{T_{max}}{A} \quad (9)$$

$$EB (\%) = \frac{L_f - L_0}{L_0} \times 100 \quad (10)$$

where T_{max} (N) is the maximum load required to fracture the films, A (m²) is the cross-sectional area of the film, L_0 (mm) is the initial length of the film, and L_f (mm) is its length at breakage (CHACON et al., 2024).

3.2.5.8 *Antioxidant activity*

The antioxidant activity (DPPH and ABTS) of the films was evaluated using the same protocols as in Section 3.2.3.5, using film samples with 1 x 1 cm. The percentage of DPPH free radical quenching activity and ABTS scavenging activity of films was calculated using Equation (11) and Equation (12), respectively.

$$DPPH (\%) = \left(\frac{F0\%_{DPPH} - F_{DPPH}}{F0\%_{DPPH}} \right) \times 100 \quad (11)$$

$$ABTS (\%) = \left(\frac{F0\%_{ABTS} - F_{ABTS}}{F0\%_{ABTS}} \right) \times 100 \quad (12)$$

where $F0\%_{DPPH}$ is the equivalent amount of ascorbic acid ($\mu\text{g/mL}$) in the gelatin films without MC, and F_{DPPH} is the equivalent amount of ascorbic acid ($\mu\text{g/mL}$) in the samples containing MC. $F0\%_{ABTS}$ is the equivalent amount of ascorbic acid ($\mu\text{g/mL}$) in the gelatin films without MC and F_{ABTS} is the equivalent amount of ascorbic acid ($\mu\text{g/mL}$) in the films containing MC.

3.2.6 Packaging application

The model food application will be carried out with the formulation that presents the best antioxidant activity results with the 0% MC film. For this study, approximately 50 fresh bananas of uniform size, undamaged, and with a slightly greenish color were selected. The bananas were divided into three groups: control-1 (fruits without film), control-2 (fruits packed inside gelatin films without MC), and sample-1 (fruits packed inside gelatin films that had the best antioxidant activity). Each group, with three replicates, was stored at 25°C and 75% RH for 6 days. Visual inspection and color, texture, and mass loss of bananas were monitored on days 0, 1, 4, and 6 (ZHU et al., 2024).

Fruit mass loss (ML) was calculated as a percentage (%) of the initial weight using the following equation:

$$ML = \frac{m_0 - m_t}{m_t} \times 100 \% \quad (13)$$

where m_0 and m_t are the initial mass (g) and the mass (g) evaluated as functions of time, respectively.

Texture (hardness) was assessed using the same texture analyzer informed in Section 3.2.5.7. A single compression test using a 40 mm diameter probe and a speed of 30 mm/min was performed for each fruit without skin. Samples were compressed to 50 % of their original height and hardness was determined from the compression cycle (LI et al., 2020).

Films containing bananas were sealed using a sealer Selovac 200B (Brazil) in the heating mode 8 (180 °C) by 1.5 s.

3.2.7 Statistical analyses

All results were expressed as means $\pm$ standard deviation ($n \geq 3$). Analysis of variance (ANOVA) and Tukey test with a 5% significance level were performed using TIBCO Statistica software, version 13.5.0.17, which is licensed by UFSC for users connected to the redeUFSC network. Fitting of the models to data were performed with the software OriginPro version 8.5 (OriginLab, EUA).

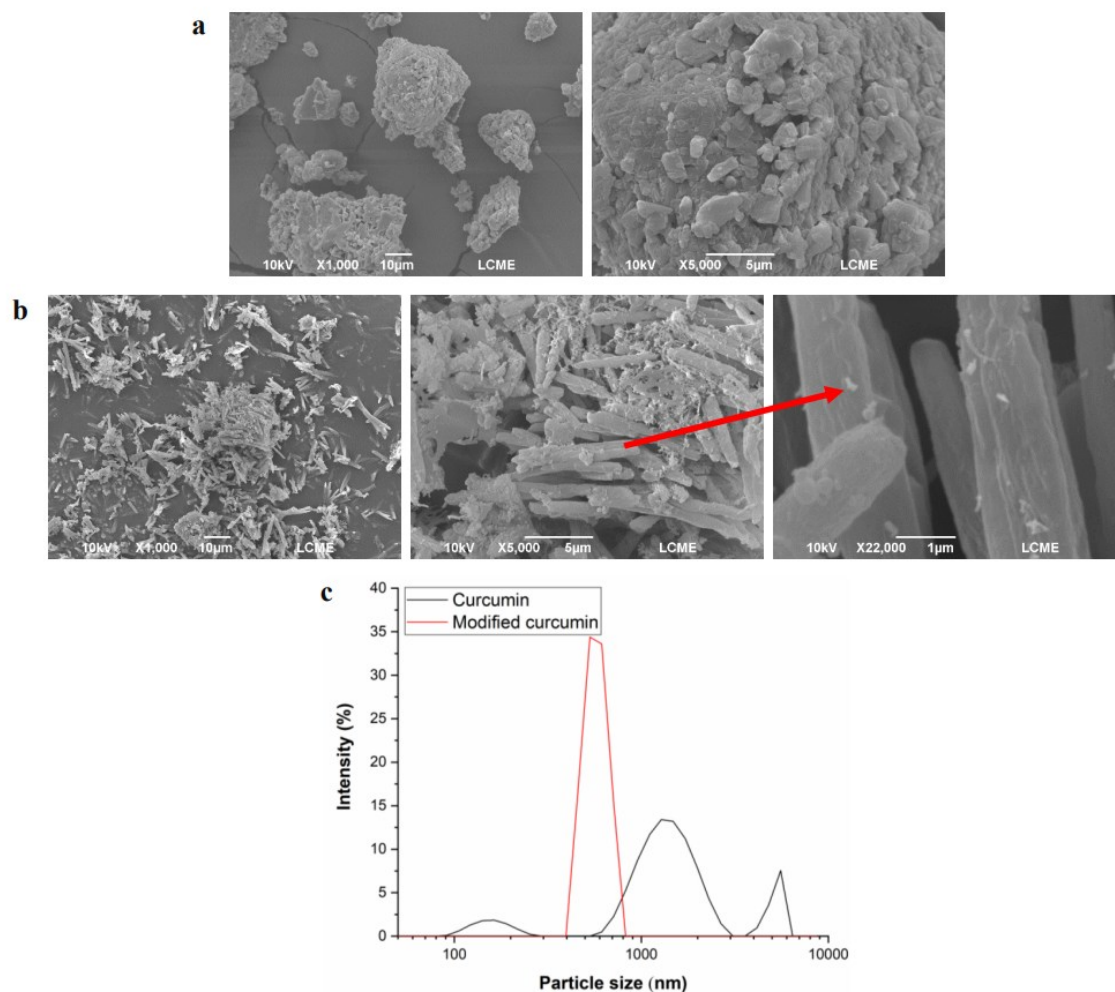
3.3 RESULTS AND DISCUSSIONS

3.3.1 Characterization of curcumin and its polymorphs

3.3.1.1 *Morphology and particle size*

Scanning electron microscopy (SEM) images are important for analyzing material morphology. SEM images revealed that the antisolvent precipitation method altered the particle size and shape of curcumin. In this way, SEM micrographs of curcumin showed that this material is an aggregate of particles with irregular shapes at the microscale (Figure 5a). In contrast, MC exhibited the presence of uniform particles with a dendritic needle format with a thickness of circa $1.0 \pm 0.3 \mu\text{m}$ (Figure 5b). These results are in concordance with those reported in the literature for curcumin modified by the antisolvent precipitation method (SILVA et al., 2019; THORAT; DALVI, 2014).

Figure 5 – Scanning electron micrographs of curcumin (a) and modified curcumin (b). Particle size distribution (DLS) of curcumin and modified curcumin (c).



Source: prepared by the authors.

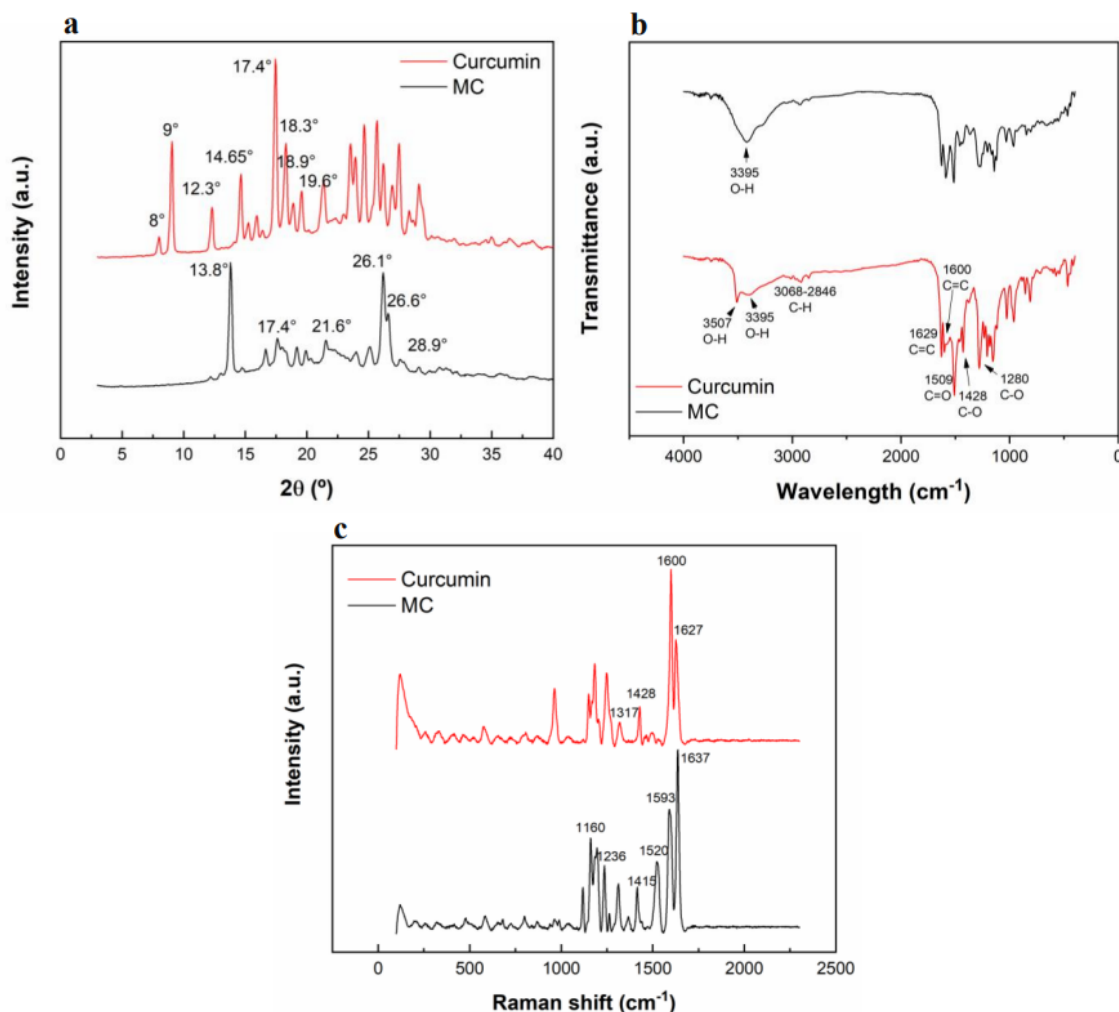
Particle size analysis is used to identify the range and frequency of particle sizes in a material. Curcumin revealed a threemodal particle size distribution with particle sizes between 80 and 7000 nm (Figure 5c). The first particle size distribution (80 – 400 nm) could be associated with curcumin particles dispersed in water. Particle size higher than 400 nm could be correlated with the presence of aggregates of curcumin particles (Figure 5c). MC exhibited a monomodal distribution with particle size between 400 and 800 nm (Figure 5c). The antisolvent precipitation method reduced particle size of curcumin, as shown in Figure 5b. However, the resulting modified particles aggregated producing particles at the nanoscale. The aggregation of the resulting particles by antisolvent precipitation has been appointed as a problem in this methodology to produce modified materials (CHACON et al., 2023). Based on SEM images and particle size results, it is possible to conclude that the antisolvent

precipitation method is an efficient approach to modify curcumin crystallinity and its particle size.

3.3.1.2 *Crystalline structure and chemical bonds*

X-ray analysis is used to determine the material crystallinity. X-ray diffractograms (XRD) revealed that curcumin and MC had a crystalline structure (Figure 6a). However, MC exhibited a reduction in crystallinity due to the antisolvent precipitation process (SILVA et al., 2019; THORAT; DALVI, 2014). Curcumin displayed a diffraction pattern typical of native curcumin in form 1 with two main diffraction peaks at angles of $2\theta = 8^\circ$ and 17.4° . Other peaks between $2\theta = 9^\circ$ and 29° also have been reported in curcumin (SILVA et al., 2019; THORAT; DALVI, 2014). In contrast, MC showed a diffraction pattern typical of curcumin polymorphs, differing in the relative orientation of keto-enol interactions between molecules (SILVA et al., 2019; THORAT; DALVI, 2014). The polymorph in form 2 exhibited a characteristic diffraction peak at $2\theta = 13.8^\circ$, with overlapping peaks at angles of $2\theta = 17.4^\circ$ and 17.8° , along with an intensified peak at $2\theta = 21.6^\circ$. Furthermore, there was a shift from $2\theta = 28.9^\circ$ in XRD of MC to $2\theta = 26.1^\circ$. Additionally, a second peak at $2\theta = 26.6^\circ$ in MC suggested the presence of another curcumin polymorph in form 3 (SILVA et al., 2019).

Figure 6 – X-ray diffractograms (a), Fourier-transform infrared (b), and Raman (c) spectra of curcumin and modified curcumin (MC).



Source: prepared by the authors.

Chemical bonds of materials can be identified by using FTIR and Raman spectroscopies. The FTIR spectra of curcumin revealed the presence of three bands centered at 3241 , 3446 , and 3345 cm^{-1} , indicating interactions between curcumin and solvent molecules, and associated with the stretching of O–H groups (Figure 6b). In contrast, MC displayed a broad band and peaks at 3241 and 3507 cm^{-1} correlated with the axial stretching of O–H groups in intra-molecular hydrogen bonds and phenol groups, respectively (Figure 6b). The intense band of MC at 3241 cm^{-1} could be due to the reduction in particle size and increase in surface area in this material. Furthermore, the intensity of the C–O stretching of phenol groups (1415 cm^{-1}) decreased in MC when compared with curcumin, while the

intensity of the ketone groups ($1602\text{--}1502\text{ cm}^{-1}$) increased in MC (Figure 6b). These changes were correlated with different interactions between keto and enol groups in the polymorphs (SILVA et al., 2019). Finally, curcumin displayed two bands for the C=C stretching in the aromatic ring at 1602 and 1596 cm^{-1} , which shift to 1581 and 1623 cm^{-1} in MC (LIU et al., 2015).

Raman spectra of curcumin revealed bands typical of C=C stretching (aromatic ring) at 1598 cm^{-1} , C=O stretching at 1627 cm^{-1} , CH₃ bending at 1431 cm^{-1} , and C–OH (enol group) or C–C–H (aromatic ring) bending at 1315 cm^{-1} (Figure 6c). The lack of bands in the carbonyl region ($1800\text{--}1650\text{ cm}^{-1}$) suggests that curcumin exists in the keto-enol tautomeric form. For MC, band shifts were observed, with C=O stretching at 1638 cm^{-1} , C=C stretching (aromatic ring) at 1593 cm^{-1} , and CH₃ bending at 1416 cm^{-1} (Figure 6b). Vibrations at 1233 cm^{-1} were associated with C–OH (enol group), and the band centered at 1158 cm^{-1} was associated with C–OH (phenol), C–C–H bending and CH₃ bending (LIU et al., 2015).

3.3.1.3 *Solubility in water and water activity*

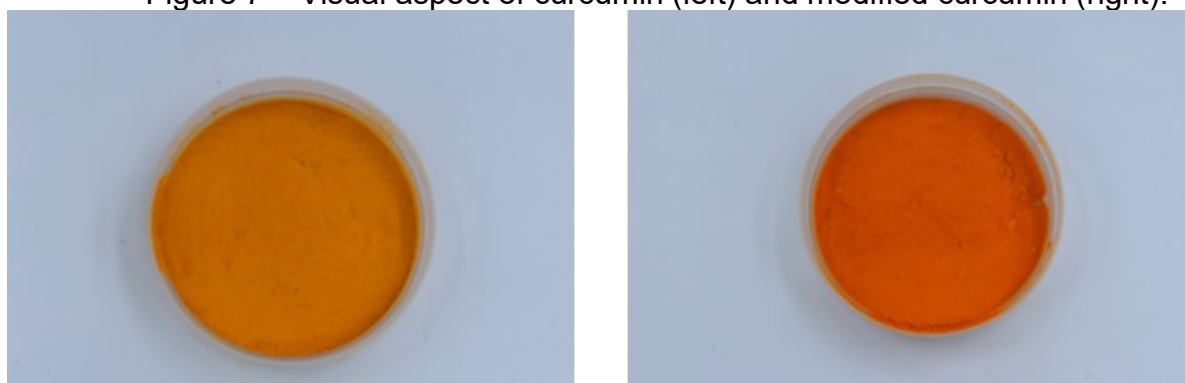
Solubility in water is a test used to quantify the amount of a substance from a sample that dissolves in water at a specific temperature. Water activity is related to the amount of water in a system that is available for chemical or biological reactions. Antisolvent precipitation significantly increased solubility and decreased water activity, respectively. In this way, the solubility in water of curcumin was 76.11 ± 6.26 and increased after modification of curcumin to 97.35 ± 2.13 ($p < 0.05$). Furthermore, water activity reduced from 0.95 ± 0.01 in curcumin to 0.55 ± 0.01 in MC ($p < 0.05$). Modifications of solubility in water and water activity could be due to crystalline and particle size modifications of curcumin after the antisolvent precipitation process (BHAWANA et al., 2011). These results suggest that MC has higher solubility in water, a desired property to enable its incorporation into biopolymer-based materials.

3.3.1.4 *Color*

Color analysis is a method for identifying material color parameters under standardized conditions. Curcumin shows the color parameters $L^* = 62.0 \pm 0.6$, $a^* =$

32.0 ± 0.3 , and $b^* = 61.8 \pm 0.0$, typical of ingredients with medium luminosity and orange color. After modified (MC), L^* value was comparable ($L^* = 59.06 \pm 0.43$, $p > 0.05$); however, higher redness ($a^* = 43.54 \pm 0.41$, $p < 0.05$) and yellowness ($b^* = 76.16 \pm 0.05$, $p < 0.05$) values were recorded. According to Andretta et al. (2019), $\Delta E > 3$ can be considered visually perceptible to the human eye. In the current research, the ΔE value between curcumin and MC was 18.64, indicating that the color difference is clearly perceptible to the naked eye for most people under normal lighting conditions. Although there were no significant differences in luminosity, MC exhibited significantly higher a^* and b^* values compared to curcumin, suggesting that MC had a more intense red hue, resulting in a more saturated and vibrant orange color (Figure 7).

Figure 7 – Visual aspect of curcumin (left) and modified curcumin (right).



Source: prepared by the authors.

3.3.1.5 *Antioxidant activity*

Analysis of antioxidant properties of packaging materials is desired to quantify and identify the chemical compounds associated with the antioxidant activity. Antioxidant compounds can impart new active properties in packaging materials. Antioxidant activity of curcumin was enhanced after its modification. Curcumin had ABTS and DPPH scavenging values of $259.06 \pm 6.72 \mu\text{g/mL}$ and $164.11 \pm 0.54 \mu\text{g/mL}$, respectively, while MC had $308.15 \pm 2.03 \mu\text{g/mL}$ and $218.86 \pm 2.12 \mu\text{g/mL}$ ($p < 0.05$). The high antioxidant properties of curcumin have been correlated with H-atom donation from the phenolic groups (BARZEGAR, 2012). After modification, MC could have a higher surface area, increasing the exposure of phenolic groups and consequently improving its antioxidant properties. The increase in antioxidant

properties in MC is desired for its application as a packaging additive since a small amount of MC can be sufficient to impart antioxidant properties in the resulting materials.

3.3.2 Characterization of composite films

3.3.2.1 *Thickness and morphology*

Film thickness is used to identify possible thickness alteration after the incorporation of a new film ingredient. The films exhibited thicknesses ranging between 70 and 80 μm , with an average of $77.60 \pm 3.48 \mu\text{m}$, which remained constant with the incorporation of MC ($p > 0.05$). Thickness results suggest that the incorporation of MC did not alter film density.

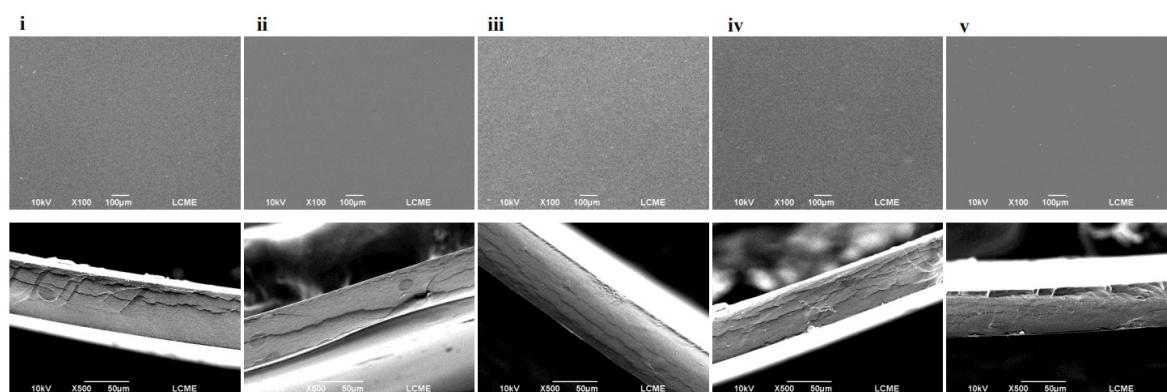
The films had a homogeneous visual aspect, which increased with MC incorporation (Figure 8a). SEM micrographs revealed that MC was uniformly dispersed in the gelatin matrix, indicating good compatibility, intermolecular binding, and affinity between MC and the polymer matrix (Figure 8b). The cross-section of films showed small cracks, without the presence of pores. Furthermore, the incorporation of MC did not reveal substantial differences between the samples, which is consistent with the X-ray diffraction patterns obtained (see Section 3.2.3.2). Similar results were obtained by Silva et al. (2019) in hydroxypropyl methylcellulose films with curcumin polymorphs.

Figure 8 – Visual aspect and microscopy of the films. (a) Gelatin films containing 0% (i), 0.1% (ii), 0.3% (iii), 0.5% (iv), and 1% (v) of modified curcumin. (b) Surface and cross-section micrographs of gelatin films containing 0% (i), 0.1% (ii), 0.3% (iii), 0.5% (iv), and 1% (v) of modified curcumin.

a



b

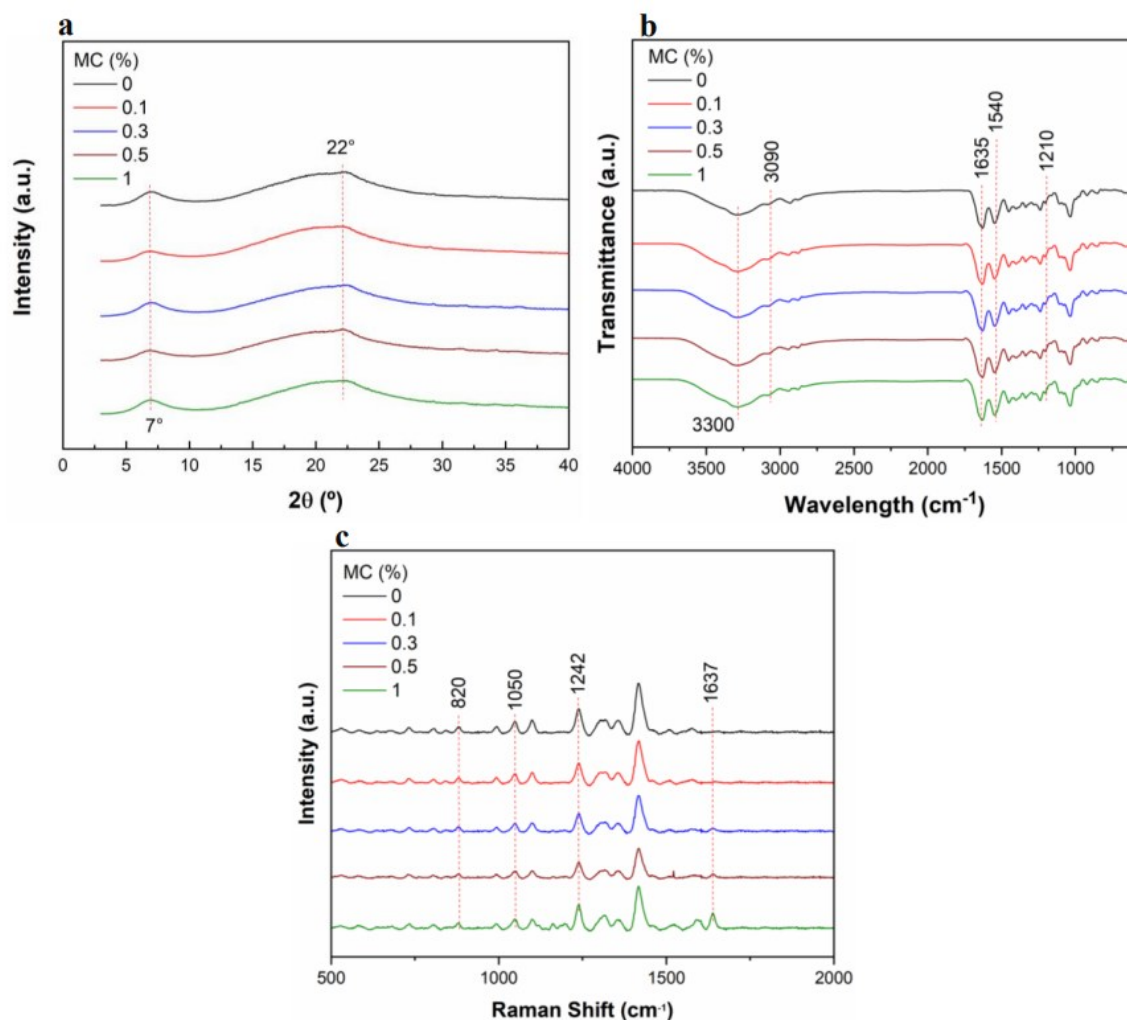


Source: prepared by the authors.

3.3.2.2 Crystalline structure and chemical bonds

Films had XRD typical of partially crystalline materials with a well-defined peak at $2\theta = 7.1^\circ$ attributed to crystalline triple helix structure of gelatin and a broad peak centered around $2\theta = 22^\circ$, characteristic of the amorphous phase (Figure 9a) (VALENCIA et al., 2016). Furthermore, diffraction peaks typical of MC (Figure 9a) were not observed in films, indicating that this material was well-dispersed in the polymeric matrix, without alter the crystalline structure of gelatin (Figure 9a).

Figure 9 – X-ray diffractograms (a), Fourier-transform infrared (b), and Raman (c) spectra of gelatin films incorporated with different concentrations of modified curcumin (MC).



Source: prepared by the authors.

The films exhibited characteristic FTIR spectra of gelatin structure (Figure 9b), featuring multiple bands, primarily in the amide I region at 3300 and 3090 cm^{-1} , triple helix at 1635 cm^{-1} , and amide II at 1540 and 1210 cm^{-1} (VALENCIA et al., 2016). The Raman spectra of all films exhibited typical peaks of gelatin structure (Figure 9c). Peaks corresponding to proline/hydroxyproline C—C bonds, sulfated glycosaminoglycan chondroitin sulfate, and amide III vibrations were identified at 820 – 880 cm^{-1} , 1050 cm^{-1} , and 1242 cm^{-1} , respectively (ESMONDE-WHITE, 2014). A peak centered at 1637 cm^{-1} typical of C=O stretching was observed in films containing MC, this peak increased with MC concentration (Figure 9c). No alteration of FTIR and Raman spectra suggest that the incorporation of MC did not alter the molecular structure of gelatin in the films, but introduced a distinct contribution in the C=O region.

Gelatin films have good packaging properties (CHACON et al., 2024; TRINETTA, 2016), and no alteration of crystalline structure and chemical bonds in gelatin films after MC incorporation suggests that these materials preserve their desired packaging properties.

3.3.2.3 *Water absorption and water solubility*

Gelatin is a biopolymer with hydrophilic and hydrophobic groups and the resulting materials from this biopolymer have high W_a and W_s values (RAWDKUEN; SAI-UT; BENJAKUL, 2010). In the current research, W_a and W_s remained constant with MC incorporation between 3007.42 – 3480.18 % and 41.0 – 55.9 %, respectively ($p > 0.05$), indicating that the strongly hygroscopic character of gelatin and glycerol controlled both parameters. Comparable W_a and W_s values have been informed in the literature in gelatin films (CHACON et al., 2024; VALENCIA et al., 2016).

3.3.2.4 *Optical properties*

The films exhibited a uniform coloration, with the MC distributed evenly (Figure 7a). While control films (0 % MC) remained colorless, those containing MC displayed hues ranging from yellow to orange, with the intensity of the orange increasing with higher concentrations of MC in the films (Table 2). The color difference was clearly perceptible to the naked eye, with a ΔE greater than 3, indicating that the variation in

color was evident to most people under normal lighting conditions (ANDRETTA et al., 2019). However, regarding the a^* parameter, only the film with 1 % MC showed a significantly higher value, indicating a more pronounced red hue compared to the other films (Table 2). Concerning the b^* parameter, all films had values that leaned towards yellow, with the film containing 1 % MC being significantly more yellow than the others (Table 2). Film luminosity decreased with MC concentration (Table 2) and tended to the observed value in MC (see Section 3.4.4).

Table 2 – Color parameters (L^* , a^* , b^* and ΔE), transmittance at 470 nm ($T_{470\text{nm}}$), and opacity (Op) of gelatin films incorporated with modified curcumin (MC).

MC (%)	L^*	a^*	b^*	ΔE	$T_{470\text{nm}}$ (%)	Op (a.u./mm)
0	88.00 ± 0.08^a	-1.56 ± 0.00^b	-0.55 ± 0.11^c	0.00	87.08 ± 0.41^a	0.59 ± 0.02^d
0.1	74.14 ± 1.22^{bc}	1.21 ± 1.48^b	28.97 ± 3.97^b	32.73	79.10 ± 2.81^b	0.52 ± 0.08^d
0.3	78.93 ± 1.30^b	2.26 ± 1.09^b	33.18 ± 4.74^b	35.15	51.33 ± 2.71^c	1.87 ± 0.15^c
0.5	76.57 ± 1.32^b	4.87 ± 1.34^b	40.98 ± 4.83^b	43.98	47.64 ± 2.32^c	2.62 ± 0.18^b
1	69.53 ± 4.17^c	16.19 ± 5.08^a	60.60 ± 10.57^a	63.30	23.11 ± 1.65^d	5.64 ± 0.04^a

All values were expressed as mean $\pm$ standard error ($n = 3$).

Means in the same column followed by different lowercase are significantly different ($p < 0.05$).

Biopolymeric films having light barrier properties could be used to protect the quality of packaged foods against photochemical damage caused by visible light (EZATI et al., 2023). Normally, gelatin films have transmittance to visible light (T_{470nm}) around 90 %, not desired for light barrier applications (CHACON et al., 2024; SAID; SARBON, 2022). In the current research, film transmittance to visible light (T_{470nm}) decreased gradually with MC incorporation (Table 2), indicating that MC had significant light adsorption in the visible region. In this way, the transmittance of gelatin films to visible light decreased by 74 % in films containing 1 % MC (Table 2). Finally, film Op increased with MC concentration due to the colorimetric properties of this ingredient (Table 2). Based on the literature, films can be considered as transparent when $Op < 5$ a.u./mm (GUZMAN-PUYOL; BENÍTEZ; HEREDIA-GUERRERO, 2022). In this way, gelatin films containing until 0.5 % MC can be used as transparent materials to pack foods where the visual appeal is important (Table 2). Although gelatin films with 1 % MC had values slightly higher than 5 a.u./mm, this film also could be used as a transparent material (Figure 8a).

3.3.2.5 *Water contact angle and water vapor permeability*

The WCA indicates whether a material is hydrophilic or hydrophobic. When the WCA is less than 90° , the material is classified as hydrophilic, meaning water spreads more easily on the surface. In contrast, when the WCA $> 90^\circ$, the material can be considered as hydrophobic, causing water to form more rounded, less spread-out droplets (BIALOPIOTROWICZ; JAŃCZUK, 2002). This property is crucial for film applications such as food packaging. In this study, all films presented a hydrophobic surface with an average WCA of 108.32 ± 2.48 , without the effect of MC ($p > 0.05$). The high WCA values in the films possibly result from the orientation of amino acid groups in gelatin during gelation. Hydrophilic amino acids stay in the aqueous matrix, while hydrophobic ones migrate to the air interface, creating a very hydrophobic surface (PULLA-HUILLCA et al., 2021).

WVP of gelatin films remained around 0.27 ± 0.01 g·mm/m²·h·kPa ($p > 0.05$) with no effect of MC. This result could be also credited to the strongly hygroscopic character of gelatin (hydrophilic amino acids) and glycerol (PULLA-HUILLCA et al.,

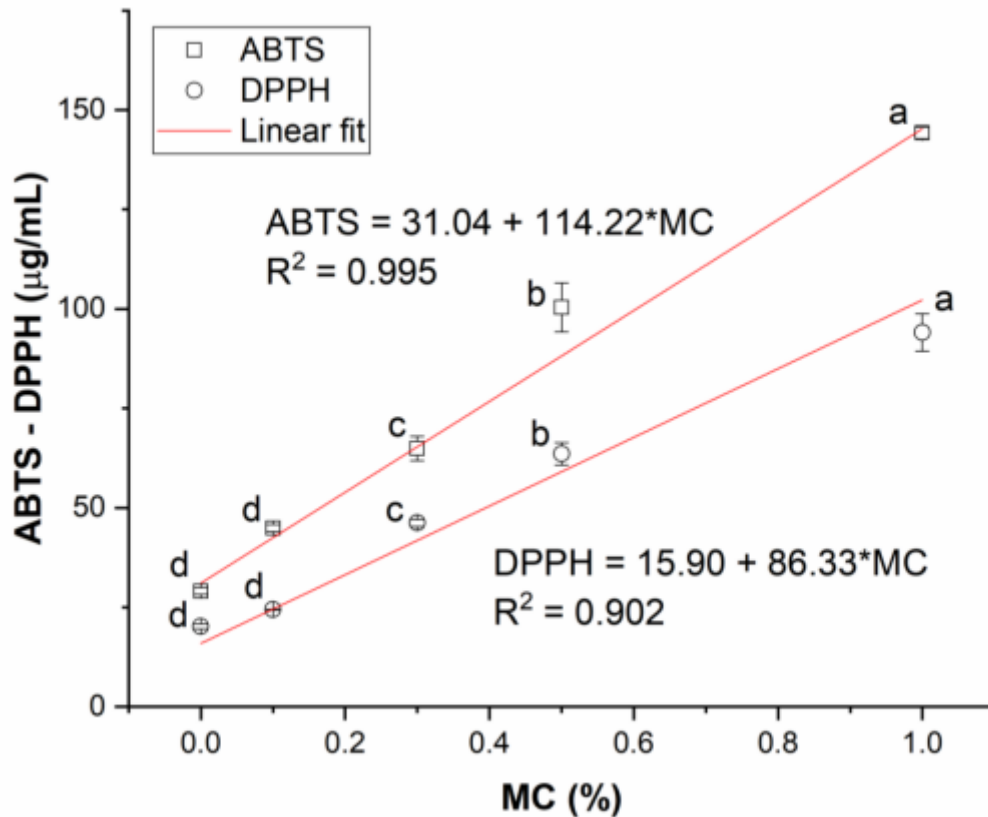
2021). In the current research, WVP values are in concordance to those informed in gelatin films (CHACON et al., 2024; SAID; SARBON, 2022; VALENCIA et al., 2016).

3.3.2.6 *Mechanical properties and antioxidant properties*

The tensile strength test is important to determine the material's resistance to external stress. Mechanical properties remained constant with no effect of MC ($p > 0.05$), suggesting that these properties were modulated by the biopolymeric matrix. In this way, EM, TS, and EB oscillated between 455 – 595 MPa, 15 – 23 MPa, and 24 – 31 %, respectively. Gelatin films have acceptable mechanical properties to be used as packaging materials (SAID; SARBON, 2022). Based on mechanical results, it is possible to conclude that the incorporation of MC did not deteriorate the desired mechanical properties of gelatin films.

Gelatin films had antioxidant properties due to the presence of phenolic rings and methoxyl groups, which allow the donation of electrons and the neutralization of free radicals (ROY; RHIM, 2020; YEN et al., 2010). The antioxidant activity of films increased linearly with MC incorporation (Figure 10), suggesting that the incorporation of MC improved the antioxidant properties of these materials as previously discussed in Section 3.3.5. Films containing 1 % MC had an improved of ABTS and DPPH values of approximately 491 % and 453 %, respectively when compared with gelatin films (Figure 10). ABTS and DPPH scavenging capacities of films with 1 % MC was approximately half the values informed in MC (see Section 3.5.5), indicating that small amounts of MC can improve antioxidant properties in the resulting material. Films with antioxidant properties are known as active films, and they could be used to reduce the oxidation process and extend food shelf life (RAWDKUEN et al., 2020; SILVA E SILVA et al., 2018).

Figure 10 – Antioxidant activity (ABTS and DPPH) as a function of modified curcumin (MC) concentration. Means in the same characterization (ABTS or DPPH) followed by different lowercase are significantly different ($p < 0.05$).



Source: prepared by the authors.

3.3.3 Packaging Application

Films with 0 % and 1 % MC were applied to packed fresh bananas (Figure 11). ML increased during the storage time without effect of the treatments (control-1: fruits without film: control-2: fruits packed inside gelatin films without MC, and sample-1: fruits packed inside gelatin films containing 1 % MC). In this way, ML oscillated between 1.77 – 1.97 %, 6.57 – 7.57 %, and 9.48 – 10.83 % for 1, 4, and 6 days of storage, respectively. These results were expected since gelatin films had high WVP values and they were not impacted with the incorporation of MC (see Section 3.4.5). Similarly, banana hardness decreased during the storage time with not effect of the presence of films ($p > 0.05$). Bananas had an initial hardness of 26.20 ± 2.04 N which decreased to 16.99 ± 1.88 N, 14.96 ± 2.49 N, and 12.80 ± 1.43 N for 1, 4, and 6 days of storage, respectively. Reduction of ML and hardness in bananas was expected

because of its physiology where fruit respiration provoked transpiration and the protopectin was continuously transformed into pectic acid and then hydrolyzed to sugar, while the fruit cells were destroyed, resulting in softening and deterioration of bananas during storage (LI et al., 2024).

Figure 11 – Visual aspect of packed bananas with gelatin films containing 0 % (top) and 1 % (down) of modified curcumin.



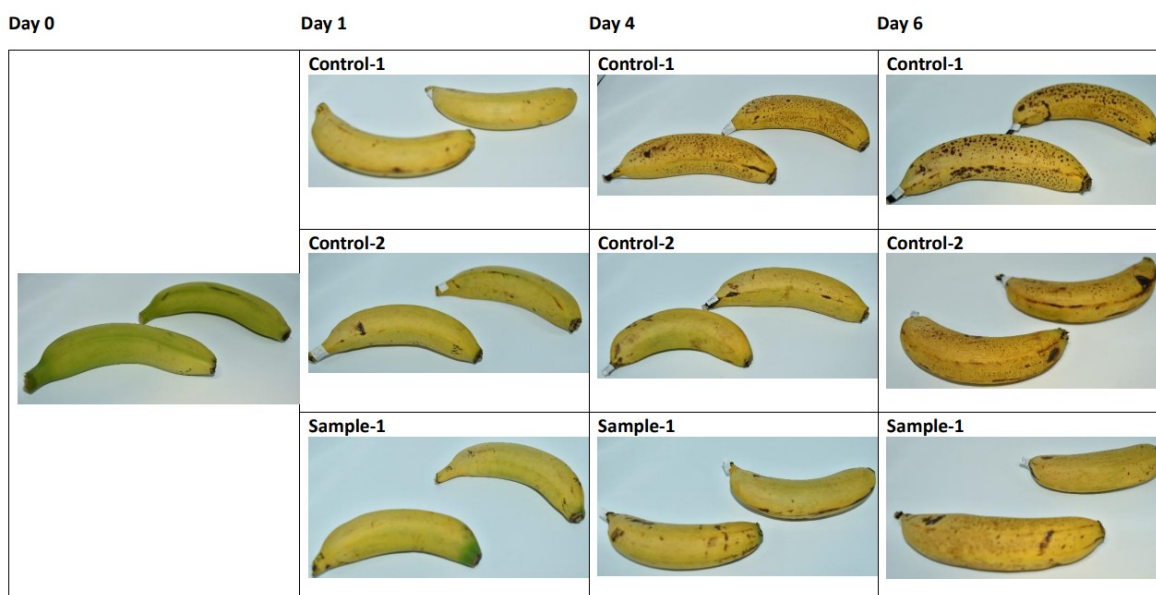
Source: prepared by the authors.

Interestingly, the presence of package films reduced browning of banana peels during their storage (Figure 12). Banana peels had a better visual aspect when packed inside gelatin films with 1% MC, followed by those packed in gelatin films without MC and unpacked fruits. Unpacked (control-1) and packed (control 2 and sample 1) bananas were classified as having a ripening stage of 7 and 6 (RINGER; BLANKE, 2021), respectively, after 4 days of storage (Figure 12). A ripening stage of 7 was

achieved in bananas from control 2 and sample 1 after 6 days of storage; however, bananas from sample 1 had fewer browning points when compared with bananas from the other treatments (Figure 12). These results evidence the importance of using packaging materials to preserve banana quality and the use of MC to reduce the browning of banana peels during storage. The antioxidant properties of both films could be responsible for the visual preservation of bananas during storage.

In summary, the incorporation of MC did not alter the desired gelatin film properties. However, MC imparted antioxidant activity in gelatin films, opening new applications for these materials as active food packaging.

Figure 12 – Change in color of bananas during storage period.



Source: prepared by the authors.

3.4 CONCLUSIONS

In this study, modified curcumin (MC) was successfully obtained on the nanometric scale (400–800 nm) using the antisolvent precipitation technique. MC was incorporated into gelatin films that were applied for banana preservation. Antisolvent precipitation significantly increased the water solubility ($\approx 97\%$) and the antioxidant activity of curcumin (ABTS $\approx 308 \mu\text{g/mL}$ and DPPH $\approx 219 \mu\text{g/mL}$). The presence of MC did not affect the thickness, water vapor permeability, wettability, and mechanical properties of gelatin films. However, this nanomaterial drastically

increased the antioxidant properties of the resulting films by 491% (1% MC). Gelatin films containing MC reduced banana browning due to their antioxidant properties. In this way, bananas packed with gelatin films containing MC showed fewer browning points when compared with unpacked bananas and packed bananas with gelatin films without MC. Gelatin films incorporated with MC can be an excellent alternative to biodegradable materials with antioxidant properties for food packaging applications.

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CHAPTER 4

DEVELOPMENT OF BILAYER MATERIALS BASED ON PAPER AND GELATIN
INCORPORATED WITH MODIFIED CURCUMIN FOR PACKAGING APPLICATION

4 DEVELOPMENT OF BILAYER MATERIALS BASED ON PAPER AND GELATIN INCORPORATED WITH MODIFIED CURCUMIN FOR PACKAGING APPLICATION

This chapter presents the results obtained from the development and characterization of bilayer packaging Kraft paper coated with gelatin incorporated with MC.

4.1 INTRODUCTION

The use of synthetic polymers dominates the food packaging sector, especially due to their low cost and versatility. However, these materials are derived from non-renewable sources, present environmental problems due to their slow degradation and accumulation in ecosystems, generating waste that remains in the environment for centuries. This context has driven the search for sustainable alternatives, such as biodegradable packaging, which has a lower environmental impact and can even contribute to food safety and quality (BARANWAL et al., 2022; BRIGHAM, 2018; GEORGE et al., 2020).

Paper emerges as a promising option among biodegradable materials, since it is renewable, recyclable, low cost and has good aesthetics. However, the high porosity and hydrophobicity of paper make it permeable to gases and oils and sensitive to water, which limits its use as food packaging. Another class of biodegradable materials is biopolymers, which have some characteristics such as non-toxicity, non-polluting, effective carrier of food additives, low cost, recyclability and good aesthetic appearance (BARANWAL et al., 2022; BASAK; DANGATE; SAMY, 2024; LIMA et al., 2024).

Bilayer packaging formed by paper and biopolymer combine the characteristics of both in a single package. Bilayer packaging can be produced by applying a layer of biopolymer to the surface of cellulose paper, improving the barrier properties of the paper and, at the same time, allowing the incorporation of bioactive compounds that help preserve the food (ALLOUI et al., 2011; LIMA et al., 2024). For those applications, curcumin, a polyphenol present in turmeric, stands out for its antioxidant and antimicrobial properties. These properties have been explored in food preservation, as curcumin can slow oxidation processes and inhibit microbial growth, factors that are fundamental for extending the shelf life of perishable products (SILVA et al., 2019; YEN et al., 2010).

Previous studies have already addressed the use of bilayer packaging made of paper coated with biopolymers (DOS SANTOS et al., 2022b; HEFFT, 2018; NAITZEL et al., 2023; ZHARKEVICH et al., 2023). Silva et al., (2019) investigated the application of modified curcumin (MC) in HPMC films. Recently, our research group

developed a packaging film based on gelatin and modified curcumin based on antisolvent precipitation. These films had antioxidant properties and reduced the browning of banana peels during the storage of this fruit (Lima et al., 2025a). To date, no study has explored the production of bilayer packaging from cellulose paper coated specifically with gelatin and modified curcumin. In this context, the objective of this work was to develop an active bilayer material based on paper coated with gelatin and modified curcumin for packaging applications.

4.2 MATERIALS AND METHODS

4.2.1 Materials

Curcumin (Neon, Brazil) was used as the antioxidant ingredient, type A gelatin biopolymer (Gelnex, Brazil) with 260 Bloom and 30 Mesh and glycerol P. A. (Neon, Brazil) were used as the macromolecule and plasticizer, respectively. Distilled water was used as the solvent of films, whereas ethanol ($\geq 99.5\%$, Neon, Brazil) was used as solvent to produce MC. Kraft paper with a grammage of 85.47 g/m^2 .

4.2.2 Production of bilayer packaging

Bilayer paper packaging with gelatin and curcumin were produced by coating paper with film-forming solutions of gelatin without and with modified curcumin (1%). The film-forming solution was placed on the surface of the paper (0.43 mL/cm^2) and then dried at $25\text{ }^\circ\text{C}$. Film-forming solutions with 0% and 1% of modified curcumin were prepared as previously informed by Lima et al., 2025a.

4.2.3 Characterization of bilayer packaging

Uncoated paper (PA) and coated paper with gelatin film-forming solution (PG) and gelatin film-forming solution with modified curcumin (PC) were analyzed.

4.2.3.1 *Morphology, thickness and grammage*

The surface and the internal microstructure after immersion in liquid nitrogen of packaging were analyzed using Scanning Electron Microscopy (SEM, JSM-6390LV, JEOL®, Japan) with an accelerating voltage of 10 kV . Samples were fixed on

aluminum stubs by carbon tape and then coated with a thin gold layer before SEM analyses (Capello et al., 2021; Lima et al., 2025a).

Thickness was measured using a micrometer (Mitutoyo N°293-561-30, Japan) with an accuracy of 1 μm . Sample thickness was informed as the average of five different sampling points in each film done in triplicate (Chacon et al., 2024b; Lima et al., 2025a).

To calculate the grammage, the mass of samples (6.5 cm-diameter) was measured using an analytical balance (Marte Científica ATX224, Brazil) and the results were expressed as g/m^2 .

4.2.3.2 *Contact angle*

Water contact angle (WCA) and oil contact angle (OCA) were measured as using an optical tensiometer (RameHart 250, US) operating with a DROPimage Advanced v2.2 software (LIMA et al., 2025a). Samples were fixed with double face tape in microscope slide and $\approx 5 \mu\text{L}$ drop of distilled water or soja oil was released onto the sample surface and then analyzing the shape of each sessile drop for 10 s.

4.2.3.3 *Barrier properties*

Water vapor transmission rate (WVTR) of samples was determined through gravimetric analysis at 25 °C (CHACON et al., 2024b; LIMA et al., 2025a). Samples were conditioned into desiccators containing silica gel for 24 hours before use and their thickness was measured. Aluminum capsules with an area of 31 cm^2 were utilized, each containing silica gel ($\text{RH} \approx 0 \%$). These capsules were then sealed with the films and subsequently, they were placed into a desiccator containing distilled water ($\text{RH} \approx 100 \%$). The capsules with the sample PA were weighed during 24 h, whereas capsules with samples PG and PC were periodically weighed during 120 h. WVTR was calculated using Eq. (1).

$$WVTR = \frac{\Delta g}{A \times \Delta t} \quad (1)$$

where $\frac{\Delta g}{\Delta t}$ represents the increase in weight of the capsule during the testing period and A is the area available for water vapor permeation (m^2).

The oxygen transmission rate (OTR) was measured using a gas permeability tester as informed by Tosati et al., 2015). The permeability coefficient was determined by applying a mass balance for O₂ in the chamber with lower pressure, as measured by the pressure transducer, and using Eq. (2):

$$P_{2(n+1)} = P_1 - \frac{(P_1 - P_2) \exp(-(A \times R \times T \times t \times P))}{V \times l} \quad (2)$$

where A denotes the permeable area (m²), V is the chamber volume (m³), t represents time (s), R is the gas constant (m³·Pa/K·mol), T is the gas temperature inside the chamber (K), l is the sample thickness (μm), P is the permeability (mol·μm/m²·s·Pa), $P_{2(n+1)}$ is the pressure at a given time (Pa), P_1 is the pressure for constant flow (Pa), and P_{2i} is the pressure in the lower-pressure chamber (Pa). Gas permeability was calculated using data to be obtained from the pressure variation as a function of time, utilizing the Solver supplement (Microsoft Office Excel®) (Tosati et al., 2015).

4.2.3.4 Color

Color characterization was carried out using a colorimeter (Delta Vista 450G, Delta Color, Brazil) based on the CIELab coordinates, namely, L^* (lightness index), a^* (color tonalities from red “+” to green “-”), and b^* (color tonalities from yellow “+” to blue “-”). The color difference (ΔE) was calculated using Eq. (3) and the color coordinates of PA (Chacon et al., 2024b).

$$\Delta E = \sqrt{(L_{sample}^* - L_{standard}^*)^2 + (a_{sample}^* - a_{standard}^*)^2 + (b_{sample}^* - b_{standard}^*)^2} \quad (3)$$

4.2.3.5 Mechanical properties

Mechanical properties were determined using a TA.HD.plus texturometer (Texture Analyser, Stable Micro Systems, UK) at 25 °C. For conducting the tensile test, the initial distance between the grips was adjusted to 20 mm, and the samples (1.5 × 4 cm) were analyzed at a velocity of 1 mm/s. The elastic modulus (EM), tensile strength (TS), and elongation at break (EB) were determined from mechanical curves, with a minimum of 20 samples per film. The EM values were derived from the slope of the

mechanical curves within the elastic region, whereas TS and EB values were calculated using Eq. (4) and (5), respectively:

$$TS = \frac{T_{max}}{A} \quad (4)$$

$$EB (\%) = \frac{L_f - L_0}{L_0} \times 100 \quad (5)$$

where T_{max} (N) is the maximum load required to fracture the samples, A (m²) is the cross-sectional area of the sample, L_0 (mm) is the initial length of the sample, and L_f (mm) is its length at breakage (Chacon et al., 2024b).

4.2.3.6 *Antioxidant properties*

The antioxidant activity of gelatin without and with modified curcumin was evaluated the DPPH (2,2-diphenyl-1-picrylhydrazyl) free radical scavenging and the capture of ABTS (2,2'-azino-bis(3-ethylbenzo-thiazoline-6-sulfonic acid) by antioxidants (Lima et al., 2025a). Gelatin films (1 x 1 cm) were removed from paper and the dissolved in methanol. In sequence, 0.1 mL of the ethanolic extract (400 µg/mL) was mixed with 3.9 mL of 0.1 mM methanolic solution of DPPH. Absorbance was measured at 517 nm after 30 min in the dark at ambient temperature. For ABTS characterization, 30 µL of ethanolic extract was added to 3 mL of the ABTS radical solution and the blend was incubated for 5min in dark at room temperature. Afterwards, the absorbance reading was performed at 734 nm. DPPH and ABTS results were expressed as µg/mL (ALVES et al., 2024).

4.2.3.7 *Sealing properties*

The sealing properties of each material was studied using a commercial pedal sealing machine (Sulpack SP400 TE G2). PA, PG, and PC were compressed by 10 s using the temperature level of 6.

4.2.3.8 *Statistical analyses*

All results were expressed as means ± standard deviation ($n \geq 3$). Analysis of variance (ANOVA) and Tukey test with a 5% significance level were performed using

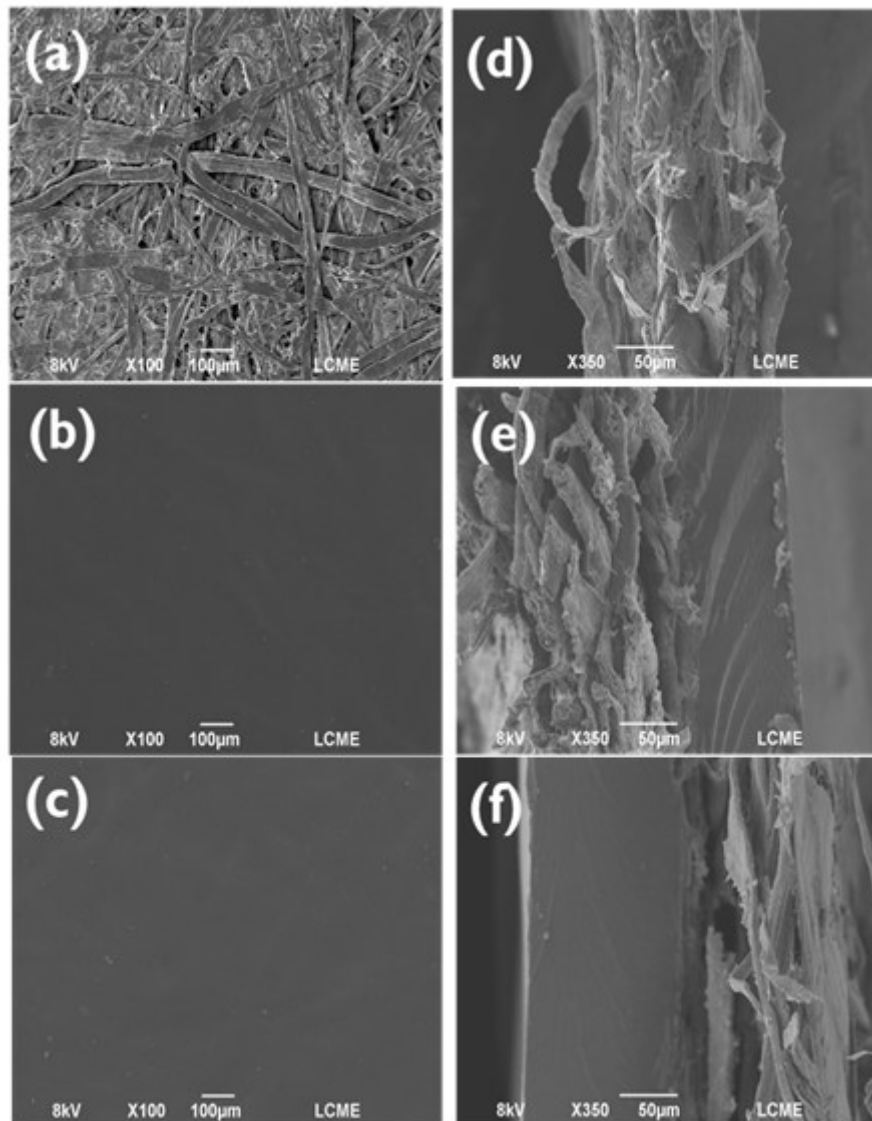
TIBCO Statistica software, version 13.5.0.17 which is licensed by UFSC for users connected to the redeUFSC network. Fitting of the models to data were performed with dedicated software (OriginPro, version 8.5, OriginLab, US).

4.3 RESULTS AND DISCUSSIONS

4.3.1 Morphology, thickness, and grammage

Micrographs of the surface of PA revealed the presence of cellulose fibers that form the paper, as well as several pores between the fibers on the PA surface (Figure 13), typical of paper materials (COSTA et al., 2022). In contrast, after coating, all cellulose fibers were completely covered, resulting in a continuous and smooth surface, without pores (Figure 13c and 13e). Regarding the micrographs of the cross-section, it was possible to observe the paper fibers in the PA samples (Figure 18b). In the coated papers, the formation of a well-defined gelatin film onto the paper surface was observed (Figure 13d and 1f), suggesting that gelatin adhered to the paper surface and created a bilayer material. Similar results were observed by Jeong et al. (JEONG; YOO, 2020) in coated paper with whey protein concentrate-beeswax-sucrose suspensions.

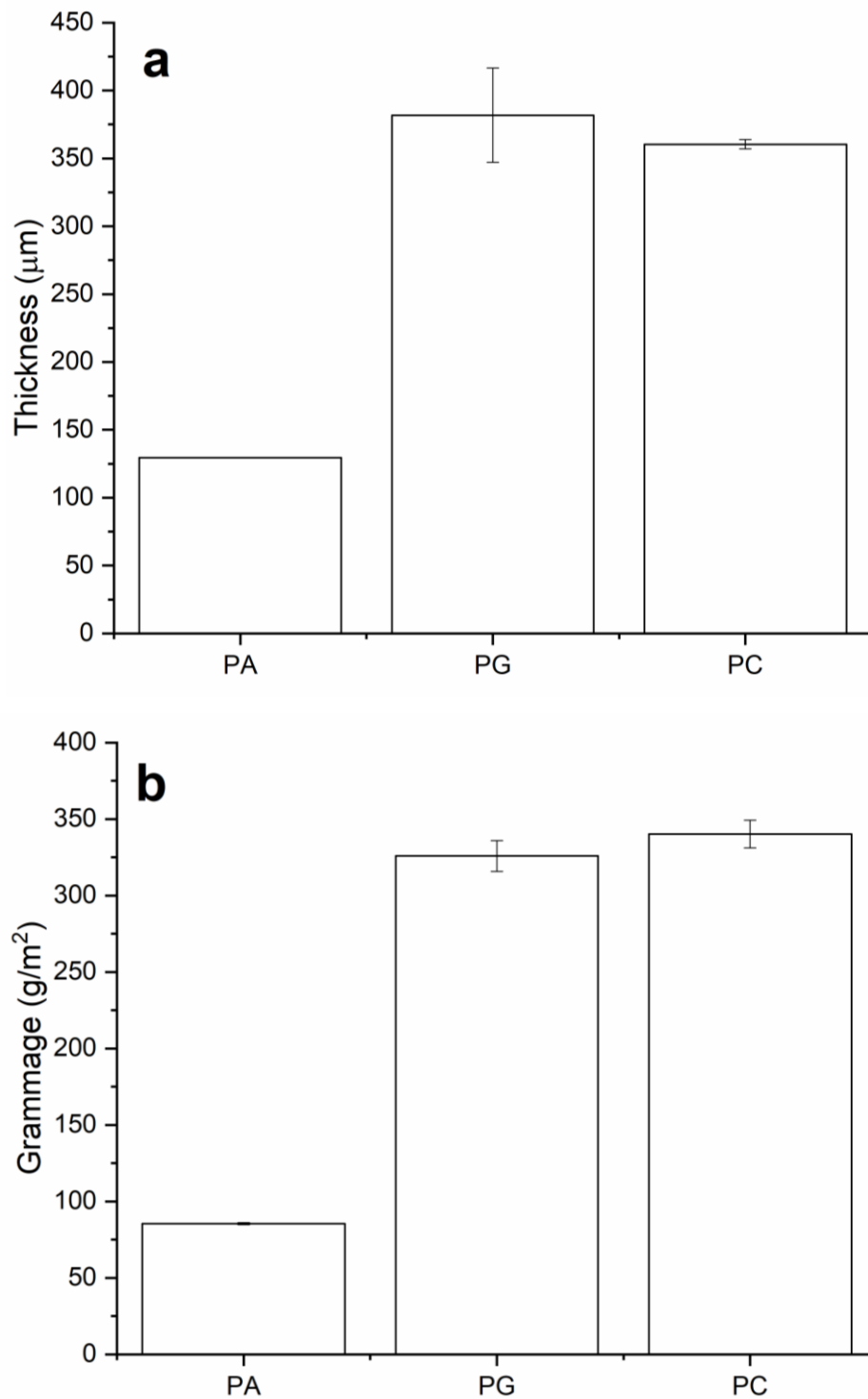
Figure 13 – Surface and cross-section micrographs of: paper (**a** and **d**); coated paper with gelatin containing 0% (**b** and **e**) and 1% (**c** and **f**) of modified curcumin.



Source: prepared by the authors.

Thickness and grammage increased after paper coating, however, it was not observed statistical differences with the incorporation of modified curcumin (Figure 14) ($p > 0.05$). PA had significantly lower thickness and grammage than PG and PC (Figure 14). PA had a grammage lower than 250 g/m², classifying it as paper, while PG and PC, with grammages exceeding 250 g/m², are classified as paperboard (LIMA et al., 2024).

Figure 14 – Thickness (**a**) and grammage (**b**) of uncoated paper (PA) and coated paper with gelatin without (PG) and with (PC) modified curcumin.



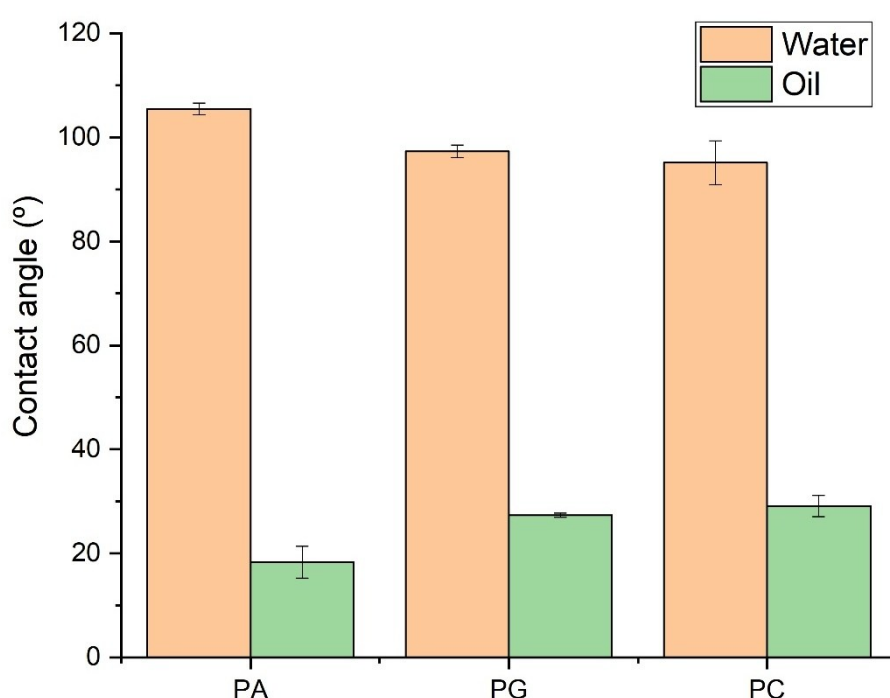
Source: prepared by the authors.

4.3.2 Contact angle

The WCA indicates the wettability of the packaging surface, determining whether a material is hydrophilic ($\text{WCA} < 90^\circ$), where water spreads easily, or

hydrophobic ($WCA > 90^\circ$), where water forms rounded droplets (BIALOPIOTROWICZ; JAŃCZUK, 2002). PA had the higher WCA, typical of highly hydrophobic surface, while PG and PC had significantly smaller WCA values ($p < 0.05$) (Figure 15), evidencing an increase in the affinity of the surface with water due to the hydrophilic nature of the applied biopolymer (CHACON et al., 2024a). However, all coated papers were classified as having hydrophobic surfaces (BIALOPIOTROWICZ; JAŃCZUK, 2002).

Figure 15 – Water and oil contact angle of uncoated paper (PA) and coated paper with gelatin without (PG) and with (PC) modified curcumin.



Source: prepared by the authors.

Regarding the OCA, the opposite behavior was observed. PA had the lowest OCA value when compared with coated papers (PG and PC), indicating lower oleophilicity of the coated surfaces and correlated with the hydrophobic nature of gelatin (Pulla-Huillca et al., 2021a). The addition of modified curcumin did not significantly interfere with the contact angle (WCA or OCA). These results suggest that gelatin-based coatings, with or without modified curcumin, reduced the interaction of the packaging surface with oil, whereas the hydrophobic surface properties are preserved in the bilayer material. Coated papers with gelatin may be advantageous for applications that require a barrier to grease or resistance to oil absorption.

4.3.3 Barriers properties

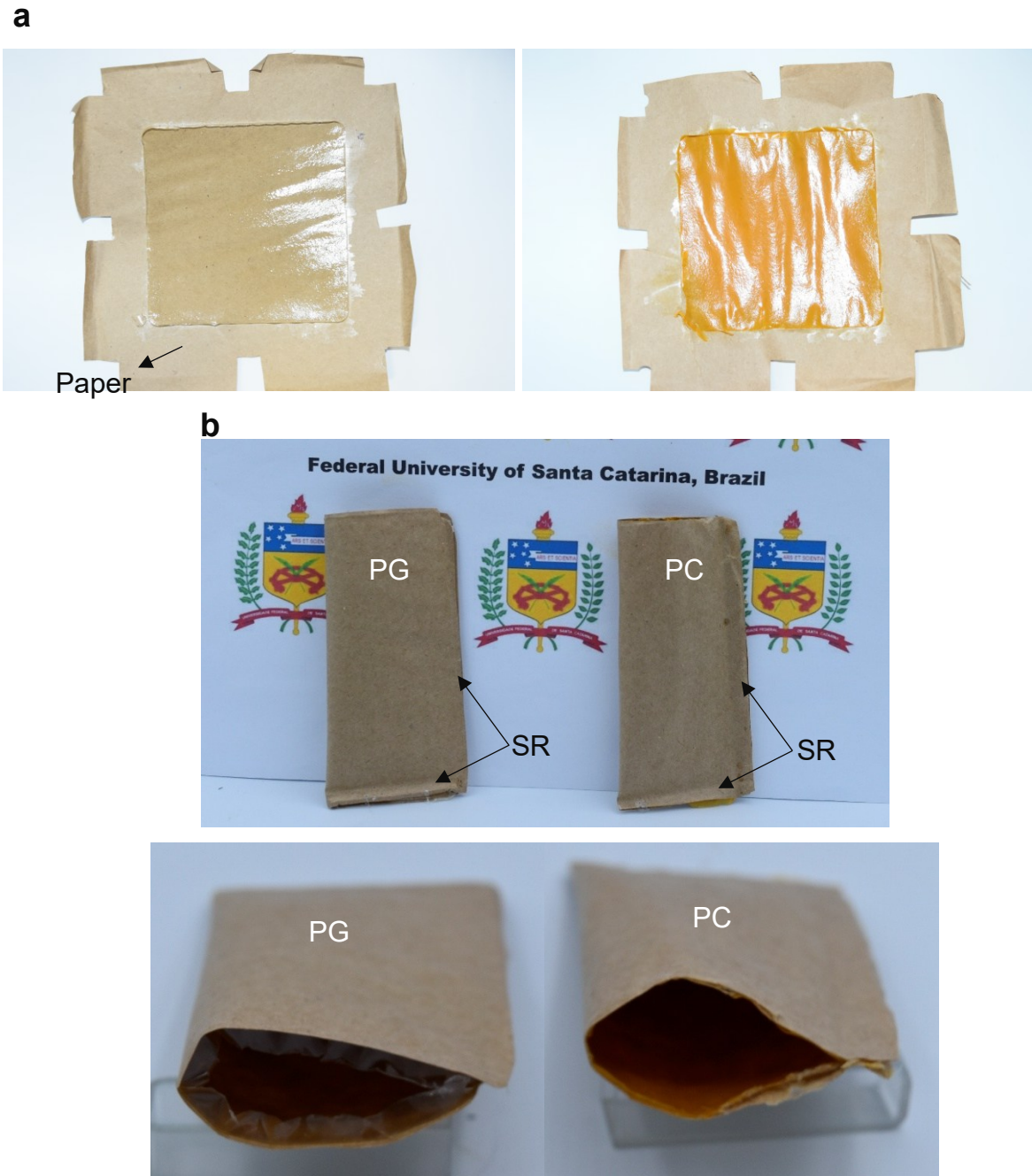
Uncoated paper (PA) had a WVTR of $0.11 \pm 0.00 \text{ g/h}\cdot\text{m}^2$, which decreased to $0.05 \pm 0.00 \text{ g/h}\cdot\text{m}^2$ in coated papers (PG and PC). The reduction of WVTR in coated papers was due to the presence of gelatin, which has low WVTR when compared with paper (VALENCIA et al., 2016b). Oxygen permeability is a crucial parameter for packaging materials, as it directly influences the preservation and oxidative stability of food (SHAH et al., 2024). PA showed structural fragility, breaking during the oxygen permeability test, which made it impossible to obtain results. This behavior reinforces the limitation of the use of paper alone as packaging, due to its high oxygen permeability associated with its porous structure. However, after its coating with gelatin, PG ($2.16 \times 10^{-8} \pm 1.10 \times 10^{-9} \text{ mol}\cdot\mu\text{m/m}^2\cdot\text{Pa}\cdot\text{s}$) and PC ($2.40 \times 10^{-8} \pm 1.73 \times 10^{-9} \text{ mol}\cdot\mu\text{m/m}^2\cdot\text{Pa}\cdot\text{s}$) had oxygen barrier properties. The addition of modified curcumin did not significantly interfere with oxygen permeability, indicating that its presence did not contribute significantly to the oxygen barrier of the evaluated packages. These results highlight the potential of gelatin coating as a strategy to improve the functionality of cellulosic substrates, making them promising alternatives in packaging with improved barrier properties (LIMA et al., 2024; MACNAMARA et al., 2024). In the current research, the coated papers have promising applications as packaging of peanuts, snacks, and coffee (TRINH; CHANG; MEKONNEN, 2023b).

4.3.4 Color

Color change of the coated packaging may affect the application of the packaging and its acceptability by the consumer. Analysis of the color coordinates in the CIELAB system of the packaging shows significant changes in the appearance of the samples depending on the coatings applied (Figure 16a). The parameter L^* (luminosity) decreased significantly with the gelatin coating (52.94 ± 0.33) and even more with the gelatin and modified curcumin coating (51.18 ± 0.27), compared to PA (59.58 ± 0.09), indicating darkening of the surface. The a^* parameter, which represents the intensity of the red color, increased progressively from 8.01 ± 0.02 in PA to 10.08 ± 0.16 in PG and to 27.47 ± 0.27 in PC. The b^* parameter, associated with the yellow hue, also increased significantly with the coatings, changing from 20.27 ± 0.00 in PA, to 28.31 ± 0.31 in PG and reaching 66.56 ± 0.53 in PC, suggesting a strong color shift towards orange tones in PC, due to the strong coloring power of curcumin (LIMA et al.,

2025a). The total color difference index (ΔE^*) reinforces these visually perceptible changes, with a moderate value of 10.6 for PG and a very sharp difference of 50.9 for PC, in relation to the PA control.

Figure 16 – (a) Visual aspect of paper coated with gelatin without (PG, left) and with (PC, right) modified curcumin. (b) Visual aspect of heat-sealed PG and PC. SR denotes the sealing region.

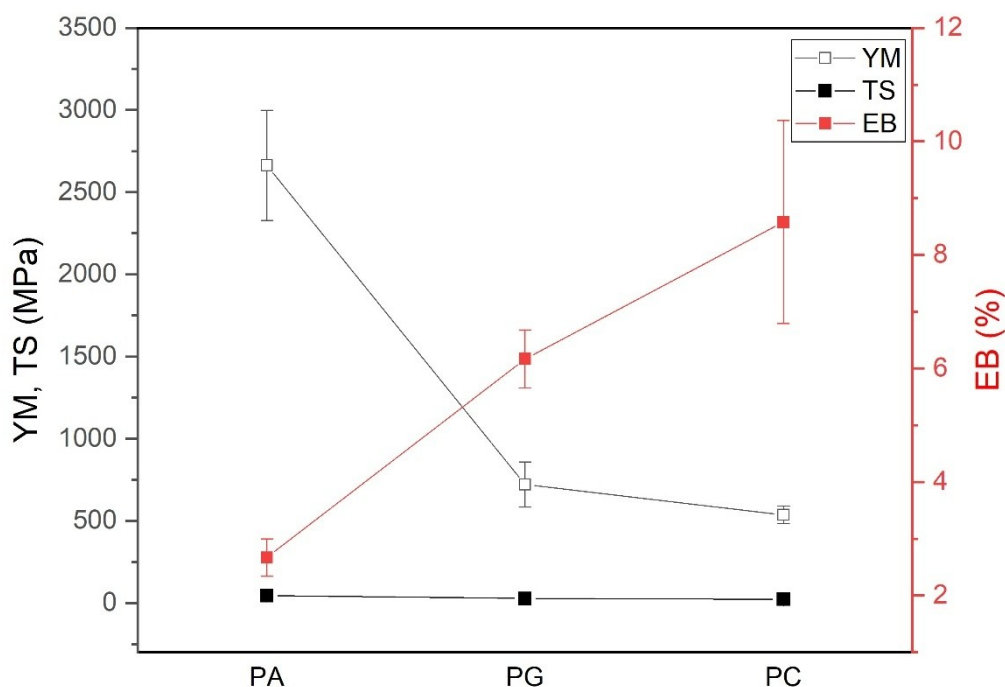


Source: prepared by the authors.

4.3.5 Mechanical properties

Mechanical properties are essential for evaluating the structural performance of packaging materials and for ensuring packaging stability during transportation, handling, and storage. Young's modulus (YM) indicates the rigidity of the material, reflecting its resistance to elastic deformation, while tensile strength (TS) represents the maximum force that the material can withstand before breaking. Finally, strain at break (DR) expresses the material's ability to stretch before breaking, and is related to its flexibility. These parameters allow to understand the balance between strength and malleability in the formulations evaluated (ALLOUI et al., 2011b; GEORGE et al., 2023). The mechanical properties of the samples revealed significant changes after the application of the coatings. PA presented the highest values of YM (2662.96 ± 334.89 MPa) and TS (44.97 ± 4.15 MPa), being significantly different from the values observed for PG (YM = 721.54 ± 137.01 MPa and TS = 27.19 ± 5.17) and PC (YM = 536.63 ± 53.97 MPa and TS = 22.67 ± 4.22 MPa), which did not differ from each other (Figure 17). These results indicate that the coatings reduced the stiffness and strength of the paper. Regarding the EB, all treatments presented significant differences from each other, with increasing values of $2.67 \pm 0.33\%$ (PA), $6.17 \pm 0.51\%$ (PG) and $8.58 \pm 1.79\%$ (PC) (Figure 17). This demonstrates that the coatings progressively increased the flexibility of the samples, especially with the presence of modified curcumin. These results suggest that although coatings reduce the mechanical strength of the paper, they promote greater ductility, which may be advantageous in applications requiring more flexible and less brittle materials.

Figure 17 – Young modulus (YM), tensile strength (TS), and elongation at break (EB) of uncoated paper (PA) and coated paper with gelatin without (PG) and with (PC) modified curcumin



3.6. Antioxidant and heat-sealing properties

Gelatin film removed from PG had antioxidant properties ($\text{ABTS} = 25.62 \pm 1.2 \mu\text{g/mL}$ and $\text{DPPH} = 21.23 \pm 0.54 \mu\text{g/mL}$) due to the presence of phenolic rings and methoxyl groups (ROY; RHIM, 2020; YEN et al., 2010). Furthermore, the incorporation of modified curcumin increased the antioxidant properties of gelatin films ($\text{ABTS} = 132.12 \pm 8.5 \mu\text{g/mL}$ and $\text{DPPH} = 88.04 \pm 5.23 \mu\text{g/mL}$). These results agree with those informed by Lima et al. (LIMA et al., 2025a) in gelatin films incorporated with the same modified curcumin (0% and 1%). The same authors observed that 1% of modified curcumin was sufficient to reduce food oxidation. Based on antioxidant results is possible to conclude that the modified curcumin increased the antioxidant properties of gelatin in PC and this material could be used as an active film to reduce oxidation process and extent food shelf life (RAWDKUEN et al., 2020; SILVA E SILVA et al., 2018).

With respect to heat seal ability, PA did not have sealing properties due to the thermal stability of cellulose (SHARMA E VARMA, 2014). However, when gelatin was coated on the paper surface, it was observed that the resulting materials (PG and PC) had sealing properties, allowing the assembly of a package (Figura 16b). The sealing

properties of gelatin have been broadly explored to manufacture packaging films with heat seal ability (NILSUWAN; BENJAKUL; PRODPRAN, 2017; TONGNUANCHAN et al., 2016).

4.4 CONCLUSIONS

In this study, biopolymer coatings based on gelatin and gelatin incorporated with modified curcumin were successfully applied to paper. Coating significantly altered morphological and structural properties of the paper, resulting in thicker materials with smoother and continuous surfaces. Despite the coatings reducing the rigidity and tensile strength of the substrates, they substantially improved flexibility, particularly in the presence of modified curcumin. Contact angle analysis revealed hydrophobic surfaces and an increase in oleophobicity after coating. Furthermore, coated papers exhibited low barrier properties (water vapor and oxygen permeability) than uncoated paper due to structural changes caused by the film-forming process. The presence of modified curcumin increased paper elasticity and imparted antioxidant activity. Coated papers had heat seal ability allowing the assembly of the package. In general, these findings highlight the feasibility of using gelatin-based coatings, with or without modified curcumin, to produce functional cellulosic packaging materials with enhanced flexibility, barrier performance, and visual appeal, with potential antioxidant activity, offering sustainable alternatives for food packaging applications.

5 CONCLUDING REMARKS

This thesis achieved its objective by developing and characterizing gelatin- and gelatin–paperboard-based materials with modified curcumin (MC) for food packaging applications. Biopolymeric coatings improved the barrier, mechanical, and functional properties of paper(board), while MC was successfully produced at the nanoscale, enhancing its solubility and antioxidant activity. Gelatin films incorporated with MC maintained their mechanical and barrier properties and effectively reduced browning in bananas, demonstrating their potential as active biodegradable packaging. Furthermore, bilayer systems combining gelatin and paper showed improved flexibility, hydrophobicity, sealability, and antioxidant functionality. Together, these results highlight the feasibility of gelatin- and MC-based films and coated paper as

sustainable, functional, and biodegradable alternatives for next-generation food packaging.

6 SUGGESTIONS FOR FUTURE WORK

Although the development of gelatin and MC films and gelatin- and CM-coated paper-based bilayer packaging for food application has shown encouraging results, there are still several opportunities for refinement and further investigation. Thus, this section outlines a number of promising directions for future research and development:

- Evaluate the effectiveness of gelatin films with MC and bilayer packaging in preserving other perishable foods, such as fruits, vegetables, meat, and seafood.
- Investigate the specific antimicrobial activity of the films and packaging against foodborne pathogens, such as *E. coli*, *Salmonella*, and *Listeria*.
- Study controlled-release mechanisms of antioxidants from gelatin films with MC and their impact on food preservation over time.
- Conduct cost-benefit and economic feasibility studies for large-scale production of these materials.
- Perform a visual homogeneity analysis of coated papers with the aim of verifying whether the coating applied to the paper presents a uniform and continuous distribution over the surface, without visible flaws such as stains, bubbles or uncovered areas.
- Investigate the migration of coated papers into food, with the aim of assessing the total amount of compounds that can be transferred from the packaging material — in this case paper — to the food with which it comes into contact.
- To investigate the water absorption capacity of packaging with the aim of evaluating its resistance to direct contact with water, an essential property for packaging intended for foods with high water activity, such as fruits and vegetables.
- Apply and investigate the efficiency in the conservation of bilayer packaging in food.

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