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Improving the Mechanical and Thermal Performance of PVA with Arbutus andrachne leaf and Fruit Extract

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Thesis Approval

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**Jerusalem- Palestine
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Dedication

This thesis is dedicated to my beloved parents, whose endless love, prayers, and sacrifices have been my greatest source of strength.

To my family, who supported me with patience and encouragement throughout this journey.

And to all those who believed in me and inspired me to never give up — this achievement is as much yours as it is mine.

Declaration

I, the author of this thesis, hereby declare that it has been submitted to *Al-Quds University* in partial fulfillment of the requirements for the degree of *Master's*, and that it represents my own original research work, except where references have been duly cited. I further confirm that this study, or any part of it, has not been submitted previously for the award of any degree or diploma at any other university or institution.

Signed: 

Mohammad Jalal Mohammad Shihadeh Jabari

Date: 12/1/2026

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Abstract

This study aims to enhance the mechanical, thermal, and functional performance of polyvinyl alcohol (PVA) films through the incorporation of methanolic extracts derived from the leaves and fruits of *Arbutus andrachne* L., a Mediterranean plant rich in phenolic and flavonoid compounds. Despite its biodegradability and biocompatibility, neat PVA exhibits low mechanical strength, limited thermal stability, and poor ultraviolet (UV) and antimicrobial resistance. To overcome these limitations, *A. andrachne* extracts were incorporated into the PVA matrix at varying volumes (0–30 mL) using the solvent-casting method to investigate the volume-dependent effects on PVA composite properties.

Fourier Transform Infrared Spectroscopy (FTIR), tensile testing, UV–Visible spectroscopy, and agar disc diffusion assays—was employed to evaluate the physicochemical, mechanical, thermal, optical, and antibacterial performance of the films. DSC analysis showed an increase in the glass transition temperature from ~50 °C for neat PVA to ~57–58 °C at higher extract loadings, accompanied by suppression of the melting peak and a reduction in melting enthalpy by ~25–40%, indicating inhibited crystallization and the formation of a more amorphous, hydrogen-bonded network. FTIR spectra confirmed strong molecular interactions, evidenced by the shift of the O–H stretching band from ~3398 cm⁻¹ to ~3375 cm⁻¹, demonstrating effective hydrogen bonding between PVA hydroxyl groups and phenolic constituents of the extract.

Mechanical testing revealed a progressive increase in tensile strength from 3.62 ± 0.28 MPa (pure PVA) to 4.42 ± 0.39 MPa at 30 mL extract, and in Young's modulus from 94.8 ± 3.8 MPa to 116.2 ± 8.6 MPa, while elongation at break decreased from 41.8% to 27.9%, reflecting enhanced film rigidity. UV–Vis analysis indicated excellent UV-blocking performance (~98–99% in both UVA and UVB regions) without significantly compromising visible light transparency. Antibacterial tests showed strong inhibition against Gram-positive *Staphylococcus aureus* and *Streptococcus* spp., and moderate inhibition against Gram-negative *Escherichia coli*, likely due to phenolic compounds disrupting bacterial cell walls.

Overall, the findings confirm that *Arbutus andrachne* extracts act as multifunctional natural reinforcements, simultaneously improving the structural, thermal, optical, and antimicrobial performance of PVA-based composite films without compromising biodegradability. While mechanical properties continued to improve up to 30 mL, the 15–20 mL range provided the most balanced combination of strength, stability, and functional performance. These eco-friendly PVA–extract composites show strong potential for biomedical applications, food packaging, and UV-protective coatings, offering a sustainable pathway toward high-performance bio-based materials.

Keywords: Polyvinyl alcohol, Composite, *Arbutus andrachne*, Mechanical properties, Thermal properties, UV-blocking, Antibacterial activity.

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Abbreviations, symbols and terminology

Abbreviation	Description
PVA	Polyvinyl Alcohol
PVAc	Polyvinyl Acetate
A. andrachne	<i>Arbutus andrachne</i> Linnaeus A. andrachne
DSC	Differential Scanning Calorimetry
FTIR	Fourier Transform Infrared Spectroscopy
UV–Vis	Ultraviolet–Visible Spectroscopy
UTM	Universal Testing Machine
GC–MS	Gas Chromatography–Mass Spectrometry
HPLC	High-Performance Liquid Chromatography
DPPH	2,2-Diphenyl-1-picrylhydrazyl
T _m	Melting Temperature
T _g	Glass Transition Temperature
T _d	Decomposition Temperature
ΔH _m	Melting Enthalpy
ΔH _d	Decomposition Enthalpy
TS	Tensile Strength
YM	Young’s Modulus
YS	Yield Strength
EB	Elongation at Break
UTS	Ultimate Tensile Strength
PTG	Precision Thickness Gauge
RE	Rotary Evaporator
LS	Laboratory Shaker
ADDA	Agar Disc Diffusion Apparatus
UVA	Ultraviolet A
UVB	Ultraviolet B

Chapter One :

Introduction

1.1 Polymers: Fundamentals and Significance

Polymers are macromolecules formed by the repetitive linkage of structural units called monomers, connected via covalent bonds. The configuration and nature of these monomers dictate the polymer's physical, chemical, and mechanical characteristics. Due to their molecular diversity, polymers can be designed to display a broad spectrum of properties, including flexibility, elasticity, stiffness, and high strength (Callister & Rethwisch, 2018). The diversity of polymers elucidates their status as a pivotal class of materials in contemporary science and technology, underpinning numerous industrial and biomedical applications.

Polymers are categorized into two primary types: natural and synthetic. Natural polymers, including cellulose, starch, proteins, and natural rubber, exist in biological systems and have been utilized for centuries in textiles, food, and medicine (Freire et al., 2021). Synthetic polymers, such as polyethylene, polystyrene, and polyvinyl alcohol (PVA), are produced using regulated chemical processes and prevail in contemporary industrial uses (Nagarkar & Patel, 2019). This classification underscores the adaptability of polymers, as they may be customized for particular functions based on their origin and structure.

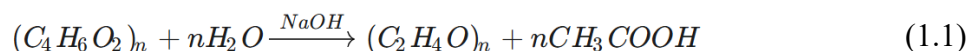
The significance of polymers in contemporary industry arises from their adjustable qualities. They are extensively utilized in packaging, textiles, automobile parts, agriculture, and biomedical engineering. Polymers yield lightweight and durable films for packaging, robust and flexible fibers for textiles, impact-resistant components for autos, soil-protective films for agriculture, and biocompatible materials for medication delivery systems and

prosthetics (Masti et al., 2022). Their versatility renders them essential to technical advancement and daily existence.

The extensive utilization of synthetic polymers, especially petroleum-derived plastics, has generated considerable environmental apprehensions. Conventional plastics exhibit resistance to disintegration, resulting in their accumulation in landfills and oceans (Tenuta et al., 2019). This endurance has amplified global research on biodegradable polymers, which decompose into innocuous byproducts under natural settings. Biodegradable polymers provide a sustainable option that diminishes environmental pollution while preserving functional efficacy (Özgen et al., 2009). Recent advancements in polymer research indicate a transition towards sustainable materials sourced from renewable resources, functional composites enhanced with natural additives, and biomedical polymers designed for biocompatibility and regulated degradation (WHO, 2013; Tlili et al., 2013).

1.2 Polyvinyl Alcohol: Synthesis and Properties

Polyvinyl alcohol (PVA) is a synthetic polymer that has gained considerable attention for its distinctive combination of properties. Unlike many synthetic polymers, PVA is not directly synthesized from vinyl alcohol due to the chemical instability of vinyl alcohol, which readily undergoes tautomerization to acetaldehyde. Instead, PVA is produced through the hydrolysis of polyvinyl acetate (PVAc), a process that replaces acetate groups with hydroxyl groups, as shown in Equation (1.1) (Nagarkar & Patel, 2019; BenchChem, 2025):



This technique facilitates exact regulation of hydrolysis extent and molecular weight, permitting the synthesis of materials with diverse solubility, crystallinity, and mechanical properties (Nagarkar & Patel, 2019; Chiellini et al., 2003). The polymer's hydroxyl-rich structure is crucial, as it enables extensive hydrogen bonding, enhancing its film-forming capability, water solubility, and compatibility with natural and synthetic additives (Peppas & Merrill, 1977).

PVA possesses numerous beneficial qualities from a physicochemical perspective, rendering it exceptionally versatile. It is water-soluble, biodegradable, and biocompatible—attributes that render it particularly appropriate for biomedical and environmental applications (Bhardwaj & Kundu, 2010). PVA exhibits excellent film-forming properties, producing translucent and flexible films with substantial mechanical strength. Its chemical resistance to oils, solvents, and different chemicals, along with superior oxygen barrier qualities, enhances its usefulness in sectors like as packaging and healthcare (Haider et al., 2015).

PVA's versatility has resulted in its adoption across numerous sectors. In biomedical applications, its hydrogels are used in wound care, contact lenses, and drug delivery systems where biocompatibility and moisture retention are key requirements (Peppas & Merrill, 1977; Bhardwaj & Kundu, 2010). In food packaging, PVA films function as effective oxygen barriers, thereby extending product shelf life and protecting against oxidation (Chiellini et al., 2003). The textile industry appreciates PVA fibers for their strength and flexibility, while its film-forming and adhesive qualities are vital in surface coatings and adhesives (Haider et al., 2015). Furthermore, in pharmaceuticals, PVA is utilized in controlled-release drug formulations and tablet coatings due to its solubility and safety profile (Nagarkar & Patel, 2019).

Compared to many synthetic polymers, PVA offers notable advantages. Its biodegradability means it poses minimal environmental impact, unlike polymers such as polyethylene or polystyrene that can linger in ecosystems for decades (Chiellini et al., 2003). Its biocompatibility also makes it preferable for biomedical applications over other synthetic counterparts that might lead to toxicity or inflammatory reactions (Bhardwaj & Kundu, 2010). These attributes position PVA as an eco-friendly material with promising potential in sustainable technologies.

Nonetheless, pure PVA is not without limitations. It exhibits relatively low mechanical strength, modest thermal stability, high hydrophilicity, and poor ultraviolet (UV) resistance—factors that significantly restrict its application in more demanding contexts. To address these shortcomings, ongoing research increasingly focuses on enhancing PVA properties by incorporating natural additives (Haider et al., 2015).

Among these, *Arbutus andrachne* L.—a Mediterranean plant rich in phenolic compounds, flavonoids, and antioxidants—has shown strong potential as a bioactive modifier. Its extracts can establish intermolecular interactions, particularly hydrogen bonding, with the hydroxyl groups of PVA, leading to improved mechanical performance, enhanced thermal stability, and added functional properties such as antimicrobial and antioxidant activity (Gulati et al., 2019).

1.3 Phytochemistry of *Arbutus andrachne*

***A recent Master's thesis by Amal Fawzi Arar (2024) at Hebron University presented a detailed phytochemical and bioactivity profile of methanolic leaf and fruit extracts of *Arbutus andrachne* (*A. andrachne*). The study identified a diverse array of phenolic acids, including gallic acid, caffeic acid, ferulic acid, p-coumaric acid, syringic acid, vanillic acid, chlorogenic acid, sinapic acid, 3,4-dihydroxybenzoic acid, 3,4-dihydroxyphenylacetic acid, 4-hydroxyphenylacetic acid, and trans-cinnamic acid, alongside flavonoids such as rutin (found in leaves), quercetin, kaempferol, and verbascoside. Other phytochemicals comprised anthocyanins, tannins, saponins, phytosterols, and terpenoids. Gas Chromatography–Mass Spectrometry (GC–MS) analysis identified hydroquinone in leaves

and penten-1-ol in fruits, whereas High-Performance Liquid Chromatography (HPLC) validated the presence of rutin in leaves and gallic acid in fruits.

The compounds demonstrated notable bioactivity: leaves displayed a high total phenolic content (391.25 mg gallic acid equivalent per gram), robust antioxidant capacity (>90% scavenging in the 2,2-diphenyl-1-picrylhydrazyl free-radical assay), and antibacterial efficacy against *Staphylococcus aureus* (17 mm inhibition zone), while fruits contained gallic acid as a predominant compound and exhibited antibacterial activity against *Escherichia coli* (10 mm inhibition zone). This study explores the integration of *Arbutus andrachne* leaf and fruit extracts into PVA films, with the objective of creating sustainable, high-performance materials that exhibit improved mechanical, thermal, optical, and antimicrobial properties for biomedical and food packaging applications."**

1.4 The Role of Medicinal Plants in Polymer Enhancement

Medicinal plants have increasingly attracted attention not only for their therapeutic applications but also for their potential in material science. The World Health Organization (WHO, 2013) has underscored the importance of traditional medicine and the exploration of medicinal plants as valuable sources of bioactive compounds. These plants contain diverse phytochemicals—such as phenolic compounds (Thaipong et al., 2006), flavonoids (Özgen et al., 2009), anthocyanins, triterpenes, and fatty acids—that are well documented for their pharmacological activities against infectious, cardiovascular, and inflammatory diseases (Kunduhoglu et al., 2011; Saklani & Kutty, 2008; Tlili et al., 2013; Tenuta et al., 2019). Beyond their biomedical functions, these metabolites offer considerable potential as natural reinforcements for polymer matrices. Their chemical structures and multifunctionality can improve the physical, mechanical, and thermal behavior of polymers while also imparting bioactive features such as antioxidant and antimicrobial resistance. Within this framework, the integration of *Arbutus andrachne* extracts into polyvinyl alcohol (PVA) presents a novel strategy to produce sustainable polymer composites with enhanced durability and added biological functionalities.

1.5 Palestinian Herbal Medicine and the Case of *Arbutus andrachne*

Palestine's ethnopharmacological tradition is enriched by a wide diversity of native plants, shaped by the region's varied topography, soils, and climatic conditions. Several species—including *Asphodeline lutea* (Liliaceae), *Clematis cirrhosa* (Ranunculaceae), and *Salvia fruticosa* (Lamiaceae)—have long been utilized in folk medicine (Ali-Shtayeh et al., 1998). Among these, *Arbutus andrachne* Linnaeus. (Eastern Strawberry Tree), locally known as "Kotlob," holds particular significance due to the strong antioxidant potential of its leaves and fruits.

Arbutus andrachne is widely available and exhibits notable ecological and genetic diversity, providing a rich source of bioactive compounds. One promising avenue lies in

extending its application beyond traditional medicine into high-value material science. Specifically, incorporating its extracts into PVA-based matrices enables the development of environmentally friendly materials with enhanced mechanical, thermal, UV-protective, and antibacterial properties. This approach highlights the cultural, ecological, and scientific significance of *A. andrachne*, demonstrating its potential to contribute to sustainable innovation in the Palestinian context.

1.6 Problem Statement

Despite its merits as a biodegradable and biocompatible polymer, the widespread application of polyvinyl alcohol (PVA) is hindered by its inherent suboptimal thermal stability and mechanical strength. Neat PVA films are characterized by relatively low tensile strength and poor thermal resistance, which render them inadequate for demanding applications in biomedical devices, advanced packaging, or flexible electronics. Conventional approaches to mitigate these weaknesses have relied on synthetic fillers and plasticizers. While often effective at enhancing performance, these additives frequently compromise the material's biodegradability and eco-compatibility, thereby negating a primary advantage of PVA.

In contrast, renewable, plant-derived additives offer a promising alternative by aligning functionality with sustainability. Nevertheless, many natural reinforcements are plagued by challenges such as poor dispersion, weak interfacial adhesion with the polymer matrix, and low environmental stability. Consequently, a critical research gap persists in the development of plant-based additives that can effectively overcome the mechanical and thermal limitations of PVA without sacrificing—and potentially even enhancing—its green credentials.

This study aims to bridge this gap by introducing a novel, sustainable reinforcement for PVA: extracts from the leaves and fruits of *Arbutus andrachne* L. Leveraging the plant's rich Volume of phenolic and antioxidant compounds, we hypothesize that these extracts will facilitate strong interfacial bonding within the PVA matrix. The research systematically investigates the potential of these extracts to produce composite films that simultaneously exhibit superior mechanical strength, enhanced thermal stability, and maintained biodegradability, addressing a key need in the development of high-performance eco-friendly materials.

Despite its favorable characteristics, the broader application of polyvinyl alcohol (PVA) in high-performance and critical sectors is restricted by several inherent limitations. These drawbacks include:

- **Inadequate Mechanical Strength:** Neat PVA films exhibit low tensile strength and elasticity, rendering them unsuitable for demanding applications such as durable packaging or load-bearing biomedical devices.

- **Poor Thermal Stability:** PVA undergoes degradation at relatively low temperatures, which limits its utility in environments where thermal resistance is essential.
- **Susceptibility to UV Degradation:** The polymer's sensitivity to ultraviolet radiation accelerates its breakdown, reducing the functional shelf life of products intended for outdoor or food packaging applications.
- **Lack of Inherent Antimicrobial Activity:** Pure PVA does not possess intrinsic antimicrobial properties, a critical requirement for applications in wound dressings, medical coatings, and active food packaging to prevent microbial growth.

These limitations present a significant challenge for industries seeking to adopt PVA as a fully functional and reliable biodegradable alternative to conventional plastics. Consequently, there is a pressing need for innovative modification strategies that can enhance these physicochemical properties without compromising PVA's core advantages of biodegradability and environmental compatibility.

Natural additives, particularly plant-based extracts rich in polyphenols, flavonoids, and tannins, offer a promising sustainable solution to these challenges. However, despite the general promise of phytochemicals, there remains a critical gap in the understanding of the specific interactions between *Arbutus andrachne* extracts and the PVA matrix. The extent to which these interactions can concurrently improve the mechanical, thermal, and functional properties of composite films is largely unexplored. This study directly addresses this gap by systematically investigating the role of *A. andrachne* leaf and fruit extracts in developing advanced, multifunctional PVA composites.

1.7 Objectives

This study aims to address the existing research gap by achieving the following objectives:

- **Introduction of an innovative sustainable reinforcement:** To systematically incorporate leaf and fruit extracts of *Arbutus andrachne* L., which are rich in phenolic and antioxidant compounds, as natural additives in polyvinyl alcohol (PVA) films for the development of environmentally friendly composite materials.
- **Evaluation of multifunctional performance:** To evaluate the effects of *A. andrachne* extracts on the mechanical and thermal properties, ultraviolet (UV) shielding capability, and antimicrobial activity of PVA-based composite films.
- **Analysis of interfacial interactions at the molecular level:** To analyze the nature and mechanisms of interfacial interactions between *A. andrachne* phytochemicals and the PVA matrix, with particular emphasis on physical and chemical bonding phenomena and their role in enhancing the microstructural integrity and overall material performance.

1.8 Research Questions

To achieve the objectives of this investigation and provide a systematic framework for analysis, the study seeks to address the following research questions:

1. **Phytochemical composition:** What are the dominant bioactive compounds—particularly phenolics and flavonoids—present in the methanolic extracts of *Arbutus andrachne* leaves and fruits, and how effective is the chosen extraction method in isolating them?
2. **Mechanical performance:** To what extent does the incorporation of *Arbutus andrachne* extract influence the mechanical behavior of PVA composite films, specifically tensile strength, Young's modulus, and elongation at break?
3. **Thermal stability:** How does the addition of the extract affect the thermal stability and decomposition kinetics of the PVA matrix, as reflected in its transition temperatures and degradation profiles?
4. **Functional properties:** What is the impact of the extract on the functional characteristics of the composites, namely ultraviolet (UV) shielding efficiency and antimicrobial activity against representative Gram-positive and Gram-negative bacteria?
5. **Intermolecular interactions:** What are the primary chemical interactions (e.g., hydrogen bonding, covalent cross-linking) between the phytochemical constituents of the extract and the PVA polymer chains, and how do these interactions explain the observed improvements in material properties?

By addressing these questions, the study aims to establish a clear understanding of the structure–property relationships in PVA bio-composites reinforced with *Arbutus andrachne* extracts. The findings are expected to advance the field of sustainable material design by demonstrating a viable strategy for enhancing synthetic polymers through the integration of multifunctional natural additives.

1.9 Hypothesis

This study is guided by the following **central hypothesis**:

The incorporation of bioactive extracts from *Arbutus andrachne* L. leaves and fruits into a polyvinyl alcohol (PVA) matrix will significantly enhance the polymer's mechanical strength and thermal stability, while simultaneously imparting multifunctional properties—such as ultraviolet (UV) resistance and antimicrobial activity—without compromising its biodegradability or eco-friendly nature.

To validate this overarching proposition, the following **sub-hypotheses** are formulated:

1. Mechanical Enhancement Hypothesis

The hydroxyl and other functional groups within the extract's phytochemical profile (e.g., phenolics, flavonoids) will establish strong intermolecular interactions, particularly hydrogen bonding, with the hydroxyl groups of the PVA chains. This molecular cross-linking is hypothesized to yield concurrent improvements in tensile strength, Young's modulus, and elongation at break.

2. Thermal Stability Hypothesis

The incorporation of phytochemical-rich extracts is expected to act as thermal stabilizers, leading to measurable increases in the glass transition temperature (T_g) and melting temperature (T_m), as determined by DSC analysis. This enhancement is attributed to restricted polymer chain mobility and improved crystallinity resulting from the extract's nucleating effect.

3. Functional Activity Hypothesis

The intrinsic bioactivity of the phytochemical constituents will impart additional functional attributes to the composite films. Specifically, the reinforced PVA is expected to demonstrate superior UV-shielding capacity and significant antimicrobial activity against representative Gram-positive and Gram-negative bacteria, extending its suitability for active food packaging and biomedical applications.

4. Amount-Dependency Hypothesis

The enhancement of material properties will follow an amount-dependent trend. An optimal extract amount is anticipated that maximizes mechanical, thermal, and functional improvements; however, beyond this threshold, negative effects such as agglomeration, phase separation, or plasticization may emerge, resulting in diminished performance.

Chapter Two :

Literature review

(Nagarkar & Patel, 2019) Conducted an extensive analysis of the inherent structural elements that dictate the physicochemical characteristics of polyvinyl alcohol (PVA). Their research highlighted the essential influence of hydrolysis degree and molecular weight on the polymer's mechanical strength, solubility, and durability. They specifically revealed that PVA with a high degree of hydrolysis displays significant intermolecular hydrogen bonding among hydroxyl groups, leading to the creation of tight and stable polymeric networks. This molecular configuration markedly improves tensile strength and durability while concurrently decreasing water solubility. These enhancements are especially beneficial in the creation of biodegradable films, where mechanical strength and regulated solubility are crucial for functional efficacy and ecological compatibility. These findings highlight the necessity of customizing PVA's structural properties as a crucial step in enhancing its application potential. Researchers may efficiently optimize strength, flexibility, and water resistance of PVA by adjusting the degree of hydrolysis and molecular weight, consequently improving its applicability in advanced fields such as biomedical materials, packaging, and controlled-release systems.

(Freire et al., 2021; Masti et al., 2022) Elaborated on these discoveries by emphasizing the constraints of tidy PVA, including its limited mechanical strength, inadequate thermal stability, and poor ultraviolet (UV) resistance. PVA is esteemed for its water solubility, biodegradability, and film-forming ability; yet, its direct use in sophisticated sectors like food packaging and biomedicine necessitates substantial property improvement. Consequently, reinforcement schemes have emerged as a primary focus of research.

(Nan, 2016) investigated the incorporation of wood-derived biochar particles into PVA matrices, reporting notable improvements in tensile modulus and thermal stability. However, a critical limitation was identified: biochar contributed little to no bioactivity,

such as antimicrobial or antioxidant properties, which are increasingly indispensable in multifunctional polymer systems designed for biomedical or active packaging applications.

Arar (2024) conducted a comprehensive phytochemical analysis of *Arbutus andrachne*, a medicinal plant native to the Mediterranean region. The study demonstrated that the leaves contain significantly higher concentrations of phenolic compounds and antioxidants than the fruits, with flavonoids and tannins exhibiting strong radical scavenging activity. These findings validate the traditional medicinal use of *A. andrachne* and highlight its potential as a bioactive enhancer for polymer-based materials.

Building on these results, the present study utilized the detailed phytochemical profiles to guide the selection and application of *A. andrachne* extracts in PVA films. The identification of key phenolic and antioxidant compounds informed the choice of leaf and fruit extracts to maximize bioactive content, anticipate interfacial interactions with the PVA matrix, and enhance the films' mechanical, thermal, UV-protective, and antimicrobial performance. In this way, Arar's work provided a foundational framework that shaped the experimental design and strategic use of natural extracts in the current study.

(Gulati et al., 2019) Complemented this perspective by demonstrating that incorporating natural fibers, such as *Sterculia alata*, into PVA films enhanced both mechanical strength and biodegradability. This aligns with the broader trend in material science toward plant-based reinforcements that deliver dual benefits: structural improvement and ecological sustainability.

(Barbălată-Mândru et al., 2022) Further advanced this line of inquiry by showing that plant extracts rich in phenolic and flavonoid compounds can interact with polymer matrices through mechanisms such as hydrogen bonding and π - π stacking. These interactions lead to improved tensile and thermal properties while also imparting antioxidant and antimicrobial functionalities. (Hiremani et al., 2022) Reinforced these findings by demonstrating that incorporating natural polyphenols into PVA improved its antimicrobial activity and delayed UV-induced degradation, thereby extending the functional lifespan of the films.

(Shaheen et al., 2024) Showed that essential oils and methanolic fruit extracts of *A. andrachne* contain more than 35 distinct compounds, with pentadecanoic acid, 14-methyl-, methyl ester identified as a major constituent. Importantly, these extracts displayed moderate antiproliferative activity without significant cytotoxic effects, supporting the plant's safety and efficacy as a source of functional bioactive agents suitable for incorporation into therapeutic and material applications.

(Abdullah & Dong, 2019) Highlighted the extensive potential of natural additives in polymer modification, demonstrating that the integration of plant-based reinforcements into biodegradable polymers markedly enhanced their physicochemical properties. This

collection of work collectively underscores the potential of natural phytochemicals—specifically phenolics and flavonoids—in improving polymer matrices.

Despite these advancements, numerous knowledge deficiencies persist. The optimal loading volumes of *A. andrachne* extracts remain undetermined, resulting in ambiguity regarding the exact concentrations necessary to enhance mechanical strength, thermal stability, and functional performance without causing adverse effects such as phase separation or plasticization. Comparative evaluations of properties between pristine PVA films and PVA composites augmented with *A. andrachne* extracts are scarce, obstructing the determination of the genuine effectiveness of bioactive reinforcing. Moreover, the long-term biodegradation kinetics and ecological safety of these bio-hybrid films have not been adequately examined, despite their potential significance for sustainable packaging and biomedical applications.

To address these gaps, the present study focuses on the incorporation of methanolic extracts of *A. andrachne* leaves and fruits into PVA films, evaluating their effects on mechanical, thermal, and functional properties. Table 1 summarizes and compares previous studies, highlighting their contributions, limitations, and the rationale for using bioactive plant extracts in polymer enhancement.

Table 2:1 Comparison of Previous Studies on the Modification and Enhancement of Polyvinyl Alcohol (PVA) Using Natural and Synthetic Additives

Study	Type of Additive/Compound	Improved PVA Properties	Notes and Limitations
Nagarkar & Patel, 2019	PVA structural modification (degree of hydrolysis and molecular weight)	Increased mechanical strength, enhanced durability, reduced water solubility	Did not include external additives; focused on intrinsic polymer structure control
Freire et al., 2021; Masti et al., 2022	None (review of PVA limitations)	Highlighted low mechanical strength, thermal instability, and poor UV resistance	Emphasized the need for property enhancement for advanced applications
Gulati et al., 2019	Natural fibers (<i>Sterculia alata</i>)	Enhanced mechanical strength and biodegradability	Achieved structural and environmental advantages

Table 2:1 Comparison of Previous Studies on the Modification and Enhancement of Polyvinyl Alcohol (PVA) Using Natural and Synthetic Additives

Study	Type of Additive/Compound	Improved PVA Properties	Notes and Limitations
Nan, 2016	Arbutus andrachne leaf and fruit extracts	High content of phenolic compounds and antioxidants	Extracts not yet incorporated into PVA; study provided a foundation for natural additive selection
Arar, 2024	Arbutus andrachne leaf and fruit extracts	High content of phenolic compounds and antioxidants	Extracts not yet incorporated into PVA; study provided a foundation for natural additive selection
Barbălată-Mândru et al., 2022	Plant extracts rich in phenolics and flavonoids	Improved tensile and thermal properties, antioxidant and antimicrobial activity	Hydrogen bonding and π - π stacking interactions crucial for performance enhancement
Hiremani et al., 2022	Natural polyphenols	Increased antimicrobial activity and delayed UV-induced degradation	Extended functional lifespan of films without compromising biodegradability
Shaheen et al., 2024	Essential oils and methanolic fruit extracts of A. andrachne	Over 35 compounds identified, moderate antiproliferative activity, non-cytotoxic	Supports safe use as functional bioactive agents for polymer applications

Chapter Three :

Experimental work

3.1 Materials

3.1.1 Chemicals and Reagents

Polyvinyl alcohol (PVA) was employed as the primary polymer matrix in this study. The PVA powder was obtained from Sigma-Aldrich (USA) with a hydrolysis degree of 98–99% and an average molecular weight of approximately 31,000 g/mol. In addition to the polymer, methanol of analytical grade ($\geq 99.8\%$) supplied by Merck (Germany) and distilled water were used as solvents during the extraction and film preparation processes. The natural additives consisted of methanolic extracts prepared from the leaves and fruits of *Arbutus andrachne* L., collected from Wadi Al-Quff, Palestine.

3.1.2 Instruments and Equipment

The preparation and characterization of the films required several instruments. Film thickness was precisely measured using a calibrated digital micrometer (Precision Thickness Gauge, PTG, Germany). Plant material was ground using an electric grinder (Morphy Richards, MR9100, UK), and concentrated extracts were obtained with a rotary evaporator (Büchi Rotavapor R 210, RE, Switzerland). Mechanical properties were measured using a Universal Testing Machine (UTM, ASTM D882, Shimadzu, Japan). Structural analysis was performed with a Fourier Transform Infrared spectrometer (FTIR, Tensor 27, Bruker, Germany), while thermal transitions were examined using a Differential Scanning Calorimeter (DSC 4000, DSC, PerkinElmer, USA). Optical properties were evaluated using an Ultraviolet–Visible spectrophotometer (UV–Vis, UVS 2700, LaboMed, USA). Antibacterial activity was assessed using a standard agar disc diffusion apparatus (ADDA). Additionally, a laboratory shaker (Cheker, LS, Germany) was employed to ensure homogeneous mixing during sample preparation.

3.2 Methods

3.2.1 Experimental Site and Sampling

Fresh leaves and fully ripened fruits of *Arbutus andrachne* L. were collected from Mahmiyat Wadi Al-Quff, a natural reserve situated near Tarqumiya in the Hebron District, Palestine. Sampling was carried out during the autumn season (late September to late October 2024), coinciding with the species' natural fruiting period, which is considered the optimal stage for obtaining high Volumes of phenolic and flavonoid compounds (Freire et al., 2021; Masti et al., 2022).

The geographical location of Wadi Al-Quff is characterized by a typical Mediterranean climate, with mild wet winters and hot dry summers, providing favorable ecological conditions for the growth of *A. andrachne* and enhancing its phytochemical profile (Callister & Rethwisch, 2018).

Botanical identification and taxonomic authentication of the plant materials were conducted by specialists from the Department of Biology at Al-Quds University, ensuring accurate species verification prior to further experimental procedures. Similar approaches to plant material authentication have been emphasized in previous phytochemical and polymer-based composite studies to guarantee reliability and reproducibility of results (Prabawati et al., 2020; Ayvaci, 2013).

3.2.2 Sample Preparation

The preparation of *Arbutus andrachne* L. samples was performed following the standardized methodology described by Arar (2024), with slight modifications to ensure reproducibility. Freshly collected leaves and fruits were initially washed 2–3 times with tap water to eliminate dust, soil particles, and other surface contaminants, followed by a final rinse with distilled water to reduce microbial load. This step is critical for preventing external impurities from interfering with subsequent extraction processes (Okmen, 2015). The slight modifications refer to minor adjustments in certain steps—such as drying time, sample grinding size, solvent volume, or extraction duration—aimed at ensuring that the results are consistent and reproducible when the experiment is repeated.

The washed samples were then dried under shade at ambient room temperature (≈ 25 °C) for a period of two to three weeks. Shade-drying, as opposed to direct sunlight exposure, was employed to minimize the degradation of thermolabile bioactive constituents such as polyphenols and flavonoids, which are known to be sensitive to high temperature and ultraviolet radiation (Masti et al., 2022; Freire et al., 2021). Once thoroughly dried, both the leaves and fruits were ground separately into fine homogeneous powders using an electric grinder (Morphy Richards, MR9100, UK).

The resulting powdered materials were transferred into pre-cleaned, airtight glass jars, labeled appropriately, and stored under controlled room conditions in a dark and dry environment to prevent oxidative degradation and moisture uptake. This storage approach has been widely reported in phytochemical studies to preserve the stability of bioactive compounds until further experimental analysis (Çinar, 2016; Chatwirakom, 2018).

3.2.3 Extraction of *Arbutus andrachne* Fruit and Leaf Compounds

3.2.3.1 Solvent Preparation and Extraction Process

The extraction of bioactive compounds from *Arbutus andrachne* fruits and leaves was carried out following the procedure of (Arar, 2024), with slight modifications to enhance recovery efficiency. A hydroalcoholic solvent system was prepared using methanol and distilled water in a ratio of 80:20 (v/v) — equivalent to 80 mL of methanol and 20 mL of distilled water per 100 mL of solvent mixture. For each extraction batch, 10 g of the dried plant powder (either fruit or leaf) was mixed with 100 mL of the solvent to ensure optimal solute-to-solvent contact. Following extraction, 7.5 g of solid residue remained, while 2.5 g were extracted into the solvent, indicating the effective extraction yield from the initial biomass.

This binary solvent system has been widely recognized as one of the most efficient media for extracting phenolic and flavonoid compounds from plant matrices, owing to its ability to dissolve both polar and moderately non-polar phytoconstituents (Masti et al., 2022; Prabawati et al., 2020).

Precisely weighed powdered samples of dried fruits and leaves were immersed in the solvent mixture and subjected to continuous agitation in a shaking incubator (Cheker, Germany) at room temperature (~25 °C) for 24 hours. The constant shaking enhanced solvent penetration into plant tissues and facilitated the diffusion of secondary metabolites, thereby improving the yield of extracted compounds (Freire et al., 2021).

Following incubation, the mixtures were filtered through Whatman No. 1 filter paper to remove plant residues. The filtrates were subsequently concentrated under reduced pressure using a rotary evaporator (Büchi Rotavapor R-210, Switzerland) at 40 °C, allowing for the removal of methanol without compromising the integrity of thermolabile compounds. The resulting aqueous extracts were then transferred into amber-colored glass vials to protect them from light-induced degradation and stored at 4 °C until further utilization in polyvinyl alcohol (PVA) film formulation and subsequent physicochemical and antimicrobial analyses (Çinar, 2016; Chatwirakom, 2018).

3.2.3.2 Control Sample Preparation

A solvent control was prepared using the same hydroalcoholic system of methanol and distilled water (80:20 v/v) but without the addition of *Arbutus andrachne* fruit or leaf

powders. This blank extract was subjected to identical extraction, filtration, and Volume steps as those applied to the experimental samples, ensuring consistency across all procedures, as shown in figure (3.1) The inclusion of such a control is essential to differentiate between the intrinsic effects of the solvent system and the bioactive contributions of *A. andrachne* phytochemicals when incorporated into polyvinyl alcohol (PVA) films (Hiremani et al., 2022; Dexter et al., 2011).

By employing this methodological control, potential solvent-related interferences—such as background absorption or residual solvent interactions—could be accounted for, thereby strengthening the reliability of subsequent analyses and ensuring that observed improvements in film performance were attributable exclusively to the plant-derived compounds (Ayvaci, 2013; Prabawati et al., 2020).

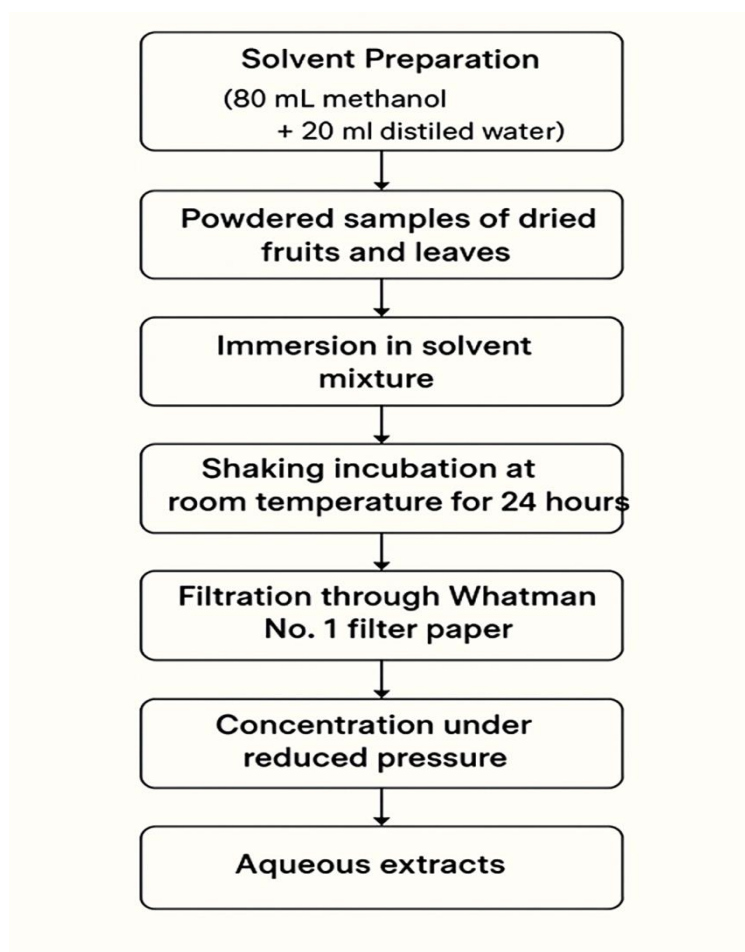


Figure (3.1): Flowchart illustrating the hydroalcoholic extraction process of *Arbutus andrachne* leaves and fruits.

3.2.4 Preparation of PVA Films

3.2.4.1 Film Formation

Polyvinyl alcohol (PVA) films were fabricated using the solvent-casting method. A 10% (w/v) PVA solution was prepared by dissolving 10 g of PVA powder in 100 mL of distilled water under constant magnetic stirring at 80 °C for 1 hour, until a clear and homogeneous solution was obtained. The elevated temperature was necessary to overcome the strong intermolecular hydrogen bonding in PVA and to ensure complete dissolution of the polymer chains (Hiremani et al., 2022).

After cooling the PVA solution to approximately 40 °C, predetermined volumes of *Arbutus andrachne* aqueous extract— 5 mL, 10 mL, 15 mL, 20 mL, 25 mL, and 30 mL — were added to **20 mL of the PVA solution** under continuous magnetic stirring. This process was carefully performed to ensure homogeneous dispersion of the bioactive compounds within the PVA matrix, resulting in a systematic series of composite films with increasing extract loadings. Such variation in extract volume was designed to evaluate the Volume-dependent influence on the mechanical, thermal, and antimicrobial properties of the fabricated PVA-based films (Barbălată-Mândru et al., 2022; Prabawati et al., 2020).

3.2.4.2 Casting and Drying

The prepared PVA–*Arbutus andrachne* aqueous extract mixtures were poured into pre-cleaned glass Petri dishes for controlled film casting. To ensure uniform thickness, the same volume of each mixture was carefully measured and poured into each dish, and the dishes were placed on a perfectly level surface. This approach allowed the solution to spread evenly across the surface, resulting in homogeneous films. Additionally, care was taken to avoid vibrations or disturbances during the drying process, which could cause uneven thickness. In some protocols, a gentle leveling tool can be used to further distribute the solution evenly before drying.

The films were dried in a hot-air oven at 40 °C for 24 hours. This temperature was chosen to promote gradual solvent evaporation while minimizing thermal degradation of thermolabile phytochemicals, preserving their bioactive properties (Freire et al., 2021). The 24-hour duration was sufficient to allow complete drying of the films at this thickness, ensuring that the solvent fully evaporated and the films became solid, transparent, and flexible. The combination of controlled volume, level surfaces, and careful temperature selection resulted in films with consistent thickness, smooth surface morphology, and reproducible physical properties suitable for further characterization.

After oven-drying, the films were carefully peeled from the Petri dishes to avoid any tearing or deformation. They were then subjected to a second drying stage at ambient laboratory conditions (~25 °C) for 24 hours. This additional air-drying step served multiple purposes. First, it ensured the complete removal of any residual moisture that might remain

after oven drying, which is crucial for preventing microbial growth and ensuring long-term film stability. Second, it enhanced the mechanical integrity of the films by allowing slow relaxation of internal stresses that could have developed during oven drying, thereby reducing the risk of cracking or warping. Finally, this step prevented structural deformation during subsequent handling and characterization, ensuring that the films maintained their intended shape, smooth surface, and consistent thickness (Masti et al., 2022; Barbălată-Mândru et al., 2022). This combination of oven and ambient drying produced homogeneous, transparent films with reproducible physical and mechanical properties suitable for further physicochemical, thermal, optical, and antibacterial analyses.

3.2.4.3 Storage

The dried PVA–*Arbutus andrachne* composite films were stored under controlled ambient laboratory conditions (25 ± 2 °C, ~50% relative humidity) in sealed polyethylene bags until further characterization. The use of sealed packaging minimized exposure to atmospheric oxygen and fluctuations in humidity, thereby preventing premature degradation, moisture uptake, or microbial contamination. Maintaining these storage conditions was critical to preserve the mechanical integrity, dimensional stability, and bioactivity of the films prior to subsequent analyses (Barbălată-Mândru et al., 2022; Masti et al., 2022).

3.2.5 Characterization of PVA Films

3.2.5.1 Mechanical Properties

The mechanical behavior of the prepared PVA–*Arbutus andrachne* composite films was systematically evaluated using a universal testing machine (Shimadzu, Japan) (Figure 3.2).

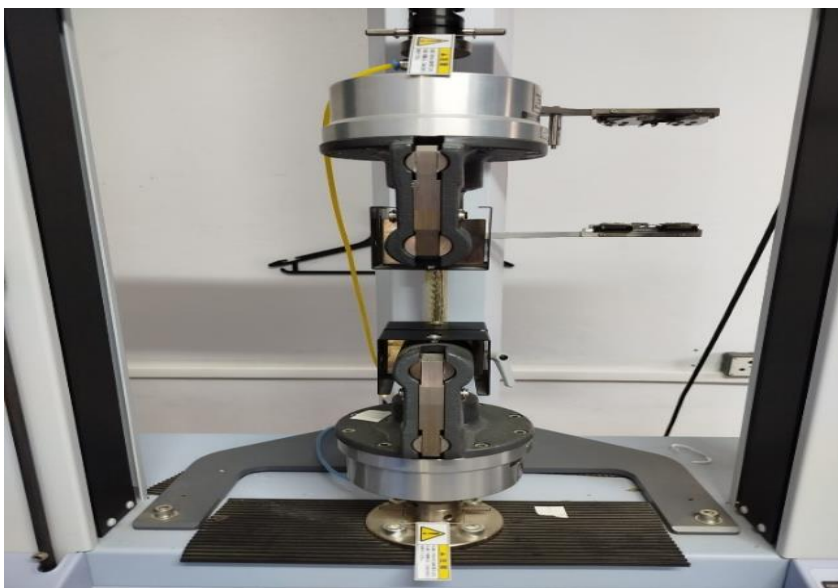


Figure (3. 2): Mechanical Testing of PVA–*Arbutus andrachne* Composite Films.

The analysis focused on determining tensile strength (TS), elongation at break (EL%), Young's modulus (E), elongation at fracture strength (EFS%), and fracture strength (FS) to comprehensively assess the films' load-bearing and deformation capacities.

Prior to mechanical testing, the thickness of each film was accurately measured at three different locations using a digital micrometer with an accuracy of ± 0.001 mm, and the average value was used in subsequent calculations to ensure dimensional uniformity. Rectangular tensile specimens were carefully cut from each film with standardized dimensions of 60 mm in length and 20 mm in width in order to maintain consistent geometry and minimize edge-related defects. During tensile testing, a constant gauge length of 30 mm was employed between the grips of the testing machine, in accordance with standard procedures for polymer film characterization.

All measurements were performed in triplicate for each film type, and the mean $\pm$ standard deviation values were recorded to ensure statistical reliability and reproducibility (Huang et al., 2023; Hiremani et al., 2022). Based on the collected data, the prepared films exhibited an average thickness of 0.400 ± 0.060 mm, representing the overall range across all samples. Thickness values correspond to the mean of three independent measurements taken from different regions of each film using a calibrated digital micrometer. Measuring from three distinct regions is generally sufficient to capture the film's uniformity. The observed variation corresponds to approximately 15%, which is acceptable for solution-cast polymeric films and is unlikely to significantly affect the films' mechanical properties, such as tensile strength or elongation at break, provided the thickness distribution is relatively uniform. This approach ensures reliable characterization while maintaining practical feasibility in experimental evaluation.

Mechanical testing was conducted at room temperature (≈ 25 °C) and a crosshead speed of 10 mm/min, following ASTM D882 guidelines for thin polymeric films. Stress–strain curves were generated automatically by the testing software to extract the following parameters:

- **Tensile Strength**

The tensile strength (TS) represents the maximum stress that a composite specimen can withstand while being stretched or pulled before rupture. It corresponds to the highest point on the stress–strain curve, The tensile strength (TS) was calculated using the following equation(1):

$$\sigma = \frac{F}{A} \quad (3.1)$$

Where:

σ (stress) = the internal resistance of a material to deformation under an applied force, expressed as force per unit area (MPa)

F = maximum applied force (N)

A = cross-sectional area (m²)

Tensile strength has the dimensions of force per unit area, and according to the International System of Units (SI), it is expressed in pascals (Pa) or megapascals (MPa), equivalent to newtons per square meter (N/m²).

- **Elastic Modulus**

Strain (ϵ) represents the relative deformation of a material under applied stress. It is a dimensionless quantity defined as the ratio of the change in length to the original length of the specimen, calculated as shown in Equation (2):

$$\epsilon = \frac{L_f - L_0}{L_0} \quad (3.2)$$

Where:

- L_0 = original length of the specimen (mm)
- L_f = final length of the specimen after load application (mm)

The elastic modulus (E), also known as the modulus of elasticity, quantifies a material's resistance to elastic deformation under applied stress. It is defined as the slope of the linear (elastic) region of the stress–strain curve, as shown in Figure 3.4, and calculated according to Equation (3):

$$E = \frac{\sigma}{\epsilon} \quad (3.3)$$

Where:

- σ = stress (MPa), the internal resistance of the material to deformation under an applied force, expressed as force per unit area
- ϵ = strain, as defined in Equation (3)

The elastic modulus reflects the stiffness of the material, with higher values indicating greater resistance to elastic deformation under an applied load.

- **Yield Strength**

The yield point marks the onset of plastic deformation, beyond which the material undergoes permanent, non-reversible strain. The yield strength (σ_y) corresponds to the stress at the yield point, where the specimen transitions from elastic to plastic behavior, as illustrated in figure (3.3). Identifying this point is crucial for determining the operational stress limit of the material before permanent deformation occurs.

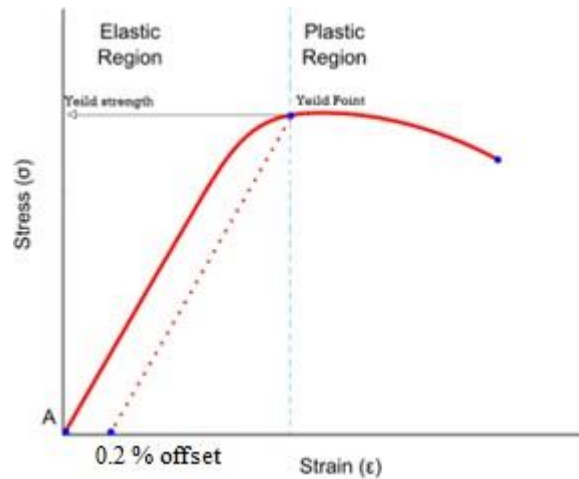


Figure (3. 3): Yield Point and Yield strength(adapted from Callister & Rethwisch, 2020).

- **Elongation at fracture**

Elongation at fracture, also called “fracture strain” or “elongation at break”, is the percentage increase in length that a material can achieve before breaking. It represents the strain at fracture expressed as a percentage. Elongation at fracture is calculated using Equation (4):

$$EL(\%) = \frac{\Delta L}{L_0} \times 100 \quad (3.4)$$

- $\Delta L = L_f - L_0$ = change in length of the specimen (mm)
- L_0 = original length of the specimen (mm)

- **Fracture strength**

Fracture strength is the ability of a material to resist failure so it's stress when a specimen fails or fractures.

There are two types of failure mode: brittle and ductile materials respectively, failure involving fracture, the determination of the failure mode involves identifying how the crack initiated and how it subsequently extended. The final recorded point in the stress-strain curve is the fracture strength as shown in figure (3.4).

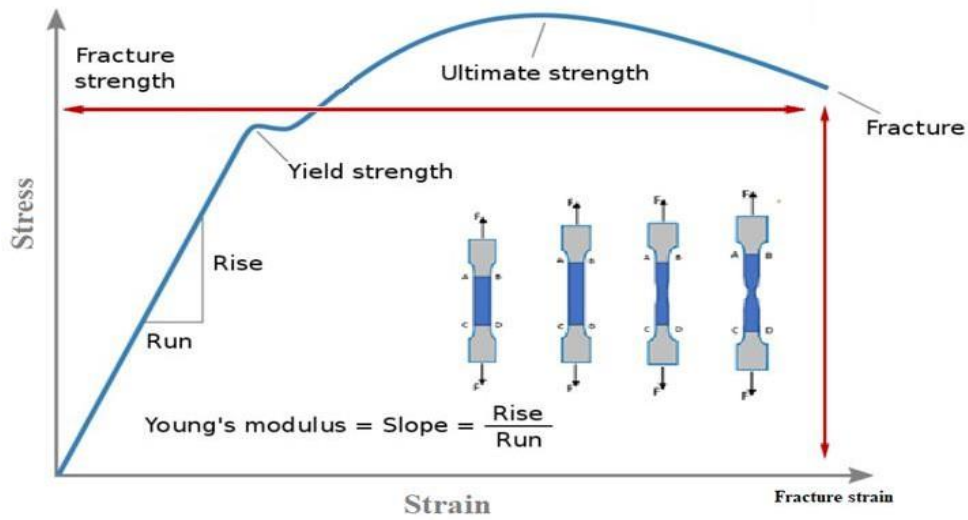


Figure 3.4: Engineering stress–strain curve (adapted from Callister & Rethwisch, 2020).

3.2.5.2 Differential Scanning Calorimetry.

In this study, the thermal behavior of polyvinyl alcohol (PVA) films containing various amounts of *Arbutus andrachne* extract was investigated using a Jade DSC instrument (PerkinElmer). The measurements were conducted at the Nanotechnology Laboratory, Al-Quds University, under a controlled nitrogen atmosphere with a flow rate of 20 mL/min. Each sample was heated at a constant rate of 10 °C/min over a temperature range of 25–450 °C. The instrument offers high accuracy and precision, with $\pm 2\%$ in heat flow and $\pm 0.1\%$ in temperature readings.

Differential Scanning Calorimetry (DSC) is widely used to obtain information about the glass transition temperature (T_g), melting point (T_m), crystallization temperature (T_c), the heat of crystallization (ΔH_c), and the heat of melting (ΔH_m) during cure or decomposition reactions, as shown in Figure 3.5. DSC analysis also allows the calculation of the degree of crystallinity (X_c), which defines the extent of long-range order in a material and strongly affects its properties. The more crystalline a polymer, the more regularly aligned its chains, which generally enhances stiffness, strength, and thermal stability (Schick, 2009).

The degree of crystallinity (X_c) can be calculated using Equation (5):

$$X_c(\%) = \frac{\Delta H_m - \Delta H_c}{\Delta H_{m0}} \times 100 \quad (3.5)$$

Where:

- ΔH_m = heat of melting of the sample (J/g)

- ΔH_c = heat of crystallization of the sample (J/g)
- ΔH_m0 = heat of melting for 100% crystalline polymer (J/g)

In cases where a distinct crystallization peak is not observed during heating ($\Delta H_c \approx 0$), or when using the reference polymer (pure PVA) for comparison, the degree of crystallinity can be simplified and calculated using Equation (3.6):

$$X_c(\%) = \frac{\Delta H_m}{\Delta H_m0} \times 100 \quad (3.6)$$

For polyvinyl alcohol (PVA), the standard melting enthalpy of the fully crystalline polymer is typically taken as 138.6 J/g ΔH_m0 . Using this reference, the relative crystallinity of PVA–Arbutus andrachne composite films can be accurately determined, providing insight into the effect of plant extract incorporation on polymer chain ordering and crystalline structure.

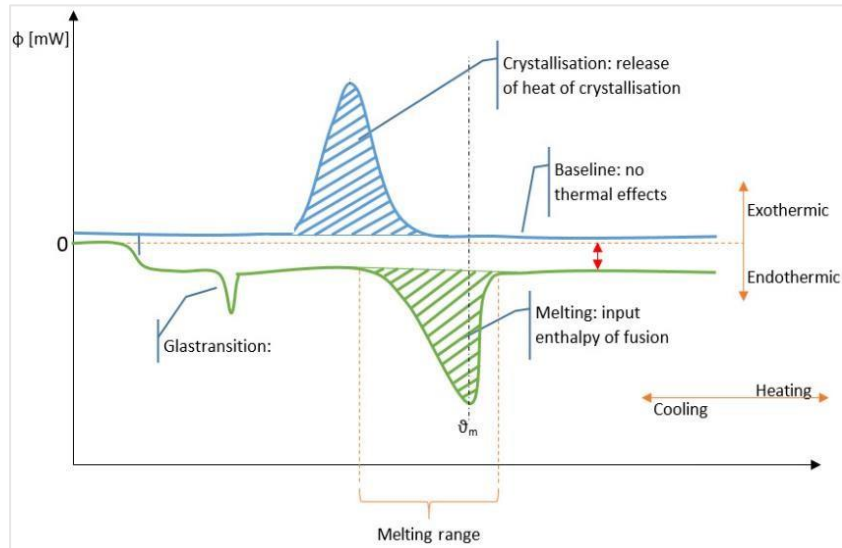


Figure (3. 5): Differential Scanning Calorimetry peak analysis (Kumar et al., 2021).

For the experimental procedure, the prepared composite samples were cut and weighed ($\sim 5 \text{ mg}$) and then placed in an aluminum pan (tray with lid). The pans were sealed using a crimper and loaded into the DSC instrument figure (3.6). Nitrogen gas was purged into the system to provide a controlled atmosphere. The DSC machine used an empty reference pan for comparison during the analysis.

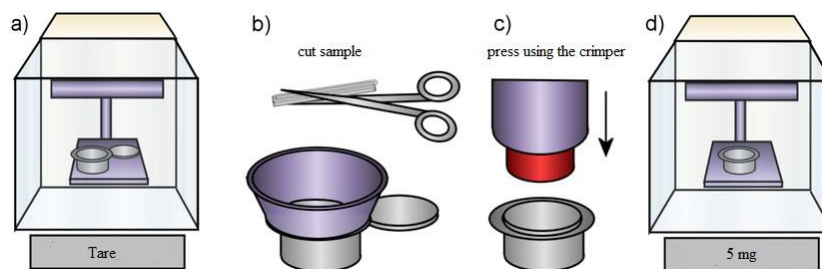


Figure (3. 6): The preparation of PVA/maple extract composite samples for DSC analysis was performed according to the method described by Steinmann, W. et al. (2013).

3.2.5.3 Fourier-Transform Infrared Spectroscopy (FTIR).

Fourier Transform Infrared (FTIR) spectroscopy is a powerful analytical technique used to identify chemical bonds and functional groups in solid, liquid, and gaseous samples. It works by measuring the absorption of infrared light at specific wavelengths, which correspond to the vibrational frequencies of molecular bonds (Baudot et al., 2010). Each functional group produces a characteristic absorption band, allowing researchers to detect molecular composition and potential interactions.

In this study, FTIR spectroscopy was employed to investigate the molecular interactions between polyvinyl alcohol (PVA) and *Arbutus andrachne* leaf and fruit extracts. Spectral measurements were recorded using a Tensor II FTIR spectrometer (Bruker) at the Pharmacy Laboratory, Al-Quds University. The instrument scanned across the wavenumber range of $4000\text{--}400\text{ cm}^{-1}$, which includes the key absorption regions for organic compounds.

Approximately 100 mg of powdered sample was prepared for each measurement and placed into the Attenuated Total Reflectance (ATR) accessory to ensure optimal contact and signal quality. The infrared beam passed through the sample, and the resulting spectrum was analyzed to identify characteristic vibrational bands such as --OH stretching, C=O stretching, and C--O stretching. These bands are commonly used to assess molecular interactions and structural modifications in polymeric systems.

FTIR spectroscopy plays a vital role in confirming the incorporation of bioactive compounds into polymer matrices and supports the development of functional materials with enhanced mechanical, thermal, and antimicrobial properties.

3.2.5.4 Antibacterial Activity Test.

3.2.5.4.1 Antibacterial Test for PVA and Maple/PVA Composites

In this study, the disc-diffusion method was employed to evaluate the antibacterial activity of two types of films: pure polyvinyl alcohol (PVA) films and polyvinyl alcohol films incorporated with *Arbutus andrachne* (maple) leaf and fruit extracts. The antibacterial properties were tested against aerobic bacteria including *Staphylococcus aureus* (MRSA), *Escherichia coli* (*E. coli*), and *Pseudomonas aeruginosa*, as well as anaerobic bacteria such as Gram-positive streptococci and Gram-negative bacilli.

Discs of both pure PVA and maple/PVA composites were placed onto agar plates inoculated with the target bacteria. The plates with aerobic bacteria were incubated at 37 °C for 24 h, while those containing anaerobic bacteria were placed in a CO₂ jar to maintain anaerobic conditions. After incubation, the zones of inhibition surrounding each sample were measured with a precise ruler. The results demonstrated that pure PVA films exhibited negligible antibacterial activity, whereas the maple/PVA composites showed clear inhibition zones, confirming the effective antibacterial contribution of bioactive compounds present in *Arbutus andrachne* extracts figure (3.7).

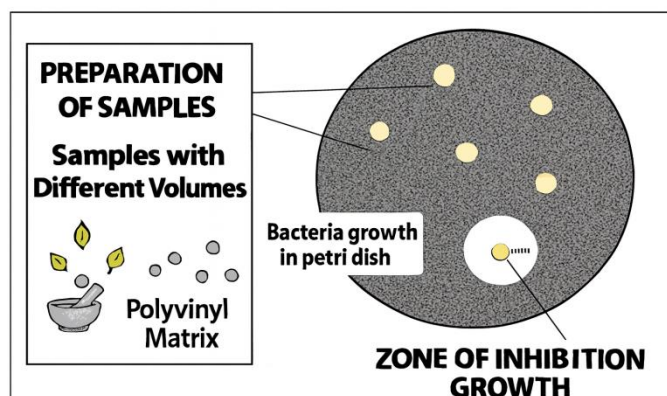


Figure (3. 7): Disc Diffusion Test Setup for PVA–*Arbutus* Composite Films.

3.2.5.5 UV Resistance Analysis.

The ultraviolet (UV) resistance of polyvinyl alcohol (PVA) composite films containing *Arbutus andrachne* extract was investigated to assess their stability and protective performance against UV radiation. The objective was to understand how varying extract Volumes influence the films' optical behavior and UV-shielding capacity. This analysis was performed using a UV–Visible Spectrophotometer (Shimadzu UV-2600, Japan), a high-precision instrument capable of measuring absorbance and transmittance across both the ultraviolet (200–400 nm) and visible (400–800 nm) regions of the electromagnetic spectrum.

sample, which is mounted in a quartz cuvette or flat sample holder. The transmitted light intensity is measured relative to a reference blank, and the resulting spectrum provides insight into the material's ability to absorb or block UV radiation. This technique is governed by the Beer–Lambert law, which defines absorbance A as directly proportional to the concentration c of the absorbing species and the optical path length l , according to Equation (7):

$$A = \epsilon c l \quad (3.7)$$

Where:

A = Absorbance (dimensionless)

ϵ = Molar absorptivity ($L \cdot mol^{-1} \cdot cm^{-1}$)

c = Volume of the absorbing species ($mol \cdot L^{-1}$)

l = Optical path length (cm)

Composite films were prepared by incorporating different volumes of *Arbutus andrachne* extract into thPVA matrix, alongside a pure PVA control. All samples were cut into uniform rectangular pieces and mounted in the spectrophotometer's sample holder. To simulate prolonged UV exposure, the films were irradiated continuously at 365 nm for 48 hours at room temperature. After exposure, each sample was analyzed to record its absorbance and transmittance spectra within the 200–400 nm range.

The UV-blocking efficiency of the films was calculated using Equation (8):

$$UV\text{-blocking (\%)} = \frac{A_{\text{sample}} - A_{\text{blank}}}{A_{\text{blank}}} \times 100 \quad (3.8)$$

Where:

- A_{sample} = Absorbance of the composite film sample at a specific wavelength
- A_{blank} = Absorbance of the reference blank (pure solvent or unmodified PVA)
- UV-blocking (%) = the percentage of UV radiation blocked by the sample

This equation (Equation 7) was applied to both UVB (280–320 nm) and UVA (320–400 nm) regions to quantify the films' shielding performance.

The phytochemical composition of *Arbutus andrachne* leaf and fruit extracts was also examined using the same spectrophotometric technique. Spectra were recorded over the full range of 200–800 nm to capture both UV and visible absorption features. Particular attention was given to absorption maxima typically associated with phenolic compounds (260–280 nm) and flavonoids (320–380 nm), which are well-known indicators of antioxidant and bioactive constituents. These spectral features support the presence of UV-active compounds that may enhance the functional properties of the composite films and contribute to their optical stability.

Chapter Four :

Results and Discussion

4.1 FTIR Analysis of PVA–Arbutus andrachne Composites

FTIR spectroscopy was utilized to examine the molecular interactions between polyvinyl alcohol (PVA) and different volumes (0–30 mL) of Arbutus andrachne leaf and fruit extract. Figure (4.1) displays the transmittance spectra obtained within the region of 4000–400 cm^{-1} . The investigation concentrated on essential functional regions, encompassing hydroxyl stretching (3600–3200 cm^{-1}), carbonyl stretching (1750–1680 cm^{-1}), CH_2 stretching vibrations (2800–2900 cm^{-1}), and ether/alcohol stretching (1200–1000 cm^{-1}). Collectively, these locations elucidate the chemical compatibility, packing alterations, and bonding mechanisms present in the composite films.

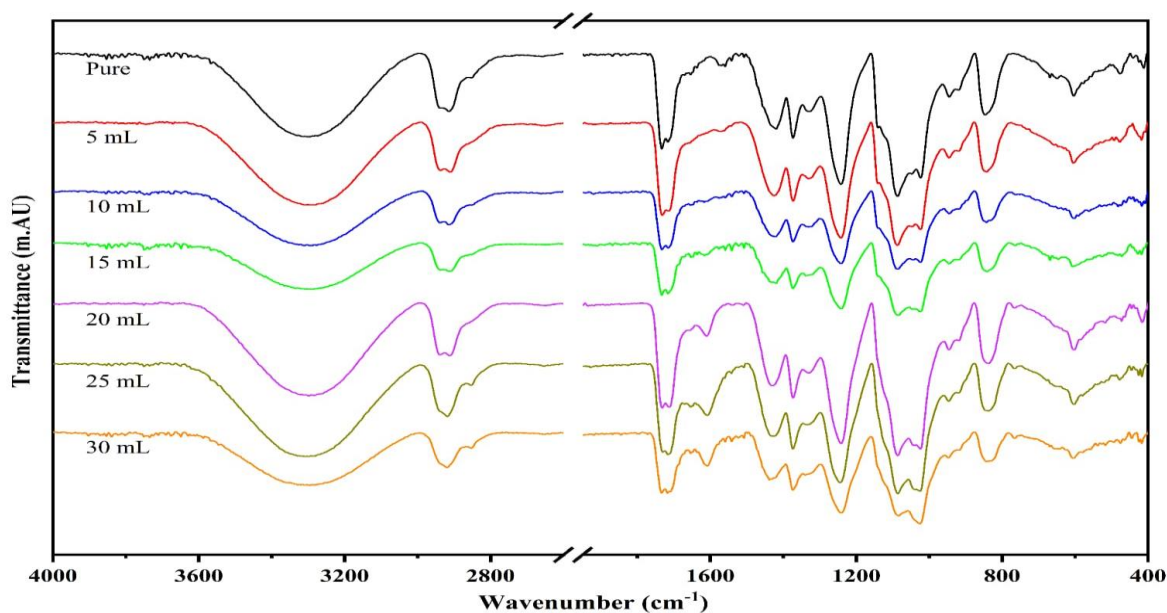


Figure (4. 1): FTIR Transmittance overlay of PVA–Arbutus andrachne composite films. Quantitative Band Analysis

The progression of band positions and shifts indicates the chemical interaction between PVA and *Arbutus andrachne* extract. Table 4.1 illustrates the exact FTIR peak positions, shifts relative to pure PVA, and functional group assignments, providing evidence of hydrogen bonding, aliphatic chain interactions, and the incorporation of carbonyl functionalities from the extract. Subsequently, Table 4.2 presents a summary of the lowest transmittance and band strengths throughout essential functional regions, offering a quantitative FTIR band analysis of PVA–*Arbutus andrachne* composite films at varying extract contents.

Table 4.1. FTIR Peak Positions, Shifts, and Functional Assignments for Pure PVA and PVA–Extract Films

Region (cm ⁻¹)	Pure PVA	5 mL (Shift)	10 mL (Shift)	15 mL (Shift)	20 mL (Shift)	25 mL (Shift)	30mL (Shift)
O–H (3600– 3200)	3398	3392 (–6)	3388 (–10)	3385 (–13)	3382 (–16)	3379 (–19)	3375 (–23)
C–H (2900– 2800)	2922	2920 (–2)	2918 (–4)	2917 (–5)	2915 (–7)	2914 (–8)	2912 (–10)
C=O (1750– 1680)	1718	1722 (+4)	1725 (+3)	1728 (+10)	1730 (+12)	1732 (+14)	1735 (+17)
C–O (1200– 1000)	1082	1080 (–2)	1078 (–4)	1076 (–6)	1075 (–7)	1073 (–9)	1070 (–12)

The FTIR spectra of PVA–*Arbutus andrachne* composite films reveal significant alterations in vibrational bands, confirming chemical interactions between the polymer matrix and the extract's constituents. The O–H stretching band of pure PVA observed at 3398 cm⁻¹ shifts progressively to 3375 cm⁻¹ at 30 mL extract content, indicating enhanced hydrogen bonding between hydroxyl groups and phenolic compounds from the extract. Similarly, the C–H stretching band at 2922 cm⁻¹ exhibits minor red shifts between –2 and –10 cm⁻¹, likely due to subtle changes in the aliphatic environment upon extract incorporation. A carbonyl band, originally present in pure PVA at 1718 cm⁻¹, grows more intense and shifts upward to the range of 1722–1735 cm⁻¹ as extract concentration increases, signaling the introduction and reinforcement of carbonyl groups from the extract.

Additionally, the C–O stretching band at 1082 cm⁻¹ shifts to 1070 cm⁻¹, indicating a decrease in crystallinity and augmented vibrational diversity in the fingerprint region. Collectively, these spectral changes indicate that the extract not only physically interacts with PVA chains but also introduces new functional groups, altering both structural

features and the hydrogen-bonding network within the composite films. Such observations align with previous studies on polymer–plant extract systems, where hydrogen bonding and carbonyl incorporation served as key mechanisms for modifying polymer matrices (Hooi et al., 2021).

Table 4. 2 Quantitative FTIR Band Analysis of PVA–Arbutus andrachne Composite Films.

Extract (mL)	O–H Band Strength 3600–3200 cm ⁻¹	C=O Band Strength 1750–1680 cm ⁻¹	C–O Band Strength 1200–1000 cm ⁻¹	CH ₂ Band Strength 2800–2900 cm ⁻¹
(Pure PVA)	0.0609	0.0582	0.1078	0.0420
5	0.0615	0.0585	0.1100	0.0417
10	0.0650	0.0600	0.1140	0.0410
15	0.0680	0.0620	0.1180	0.0405
20	0.0725	0.0669	0.1274	0.0400
25	0.0738	0.0678	0.1282	0.0398
30	0.0755	0.0692	0.1290	0.0395

Analysis and Interpretation

FTIR analysis demonstrates a clear strengthening of intermolecular interactions as extract content increases. The O–H band strength in the 3600–3200 cm⁻¹ region rises steadily from 0.0609 in pure PVA to 0.0680 at 15 mL, followed by a sharper increase to 0.0755 at 30 mL. This non-linear increase indicates that hydrogen bonding intensifies significantly beyond a threshold extract concentration (~15 mL). The concurrent red shift of the O–H stretching vibration from 3398 cm⁻¹ to 3375 cm⁻¹ confirms the formation of stronger, more cooperative hydrogen-bonded networks between PVA hydroxyl groups and phenolic compounds from the extract (Hooi et al., 2021; Kutz, 2015).

Simultaneously, the carbonyl (C=O) band strength increases from 0.0582 (pure PVA) to 0.0620 at 15 mL and further to 0.0692 at 30 mL, reflecting the progressive incorporation of carbonyl-containing phytochemicals. These carbonyl groups actively participate in hydrogen bonding, as supported by the emergence of shoulders near ~1700 cm⁻¹ at higher extract loadings. The C–O band intensity also increases markedly from 0.1078 to 0.1290, with a pronounced jump between 15 and 20 mL (0.1180 → 0.1274), indicating significant reorganization within the polymer’s vibrational fingerprint region and altered chain packing (Mirabella, 2002). In contrast, the CH₂ band strength decreases only slightly from 0.0420 to 0.0395, confirming that the aliphatic PVA backbone remains chemically intact and that the extract acts mainly through intermolecular interactions rather than backbone disruption.

In contrast, the CH₂ band strength decreases only slightly from 0.0420 to 0.0395, confirming that the aliphatic PVA backbone remains chemically intact and that the extract acts mainly through intermolecular interactions rather than backbone disruption.

4.2 Mechanical Test Analysis of PVA–Arbutus andrachne Composites

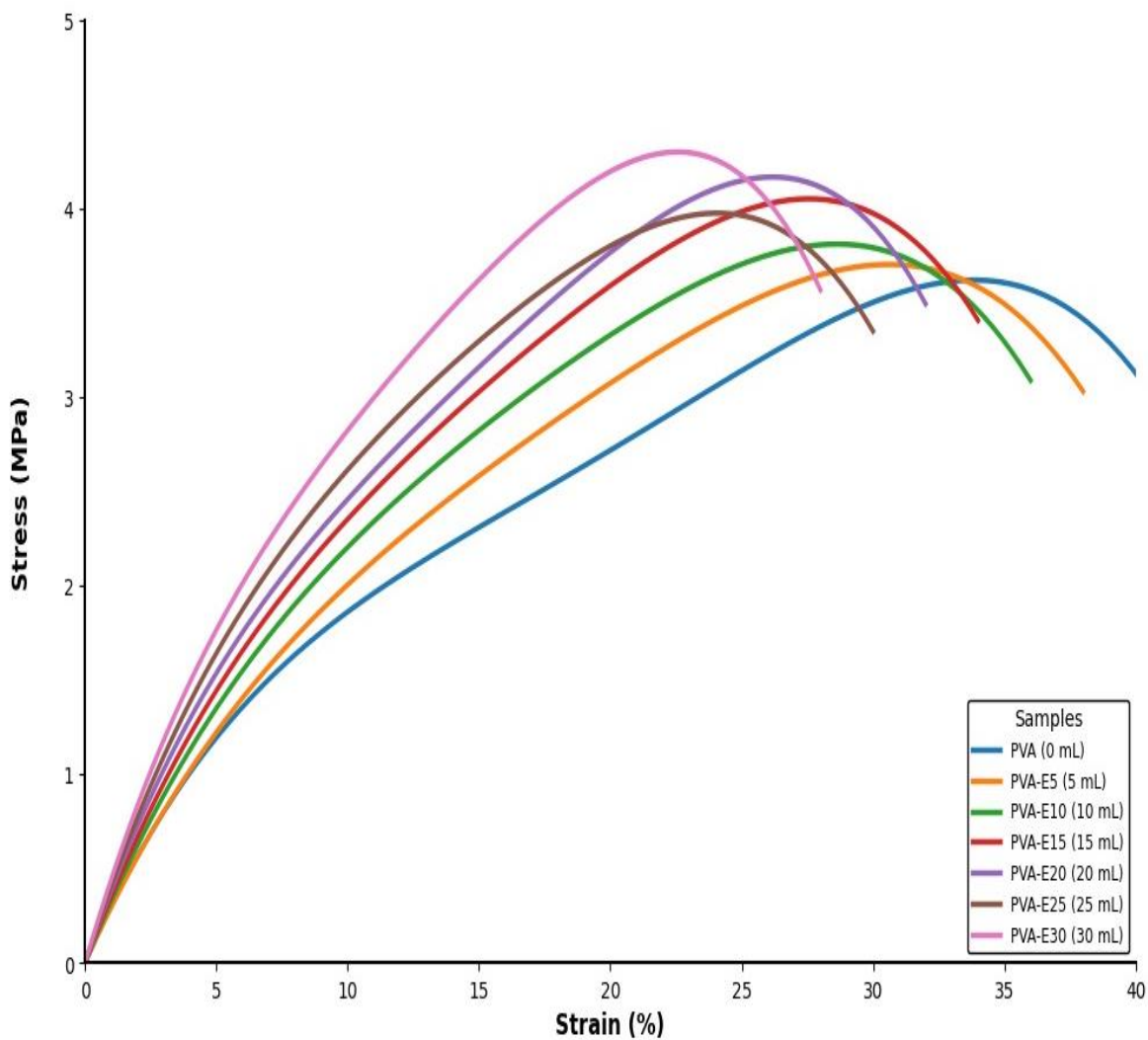


Figure 4.2: Stress–Strain Curves of PVA Films with Different Volumes of Arbutus andrachne Extract

The incorporation of Arbutus andrachne extract caused noticeable changes in the mechanical behavior of PVA films. As shown in Figure (4.2), the tensile strength gradually increased from 3.62 ± 0.28 MPa for pure PVA to 4.42 ± 0.39 MPa at 30 mL of extract. In contrast, the strain at break decreased from $41.8 \pm 0.03\%$ to $27.9 \pm 0.018\%$, indicating reduced elongation capability and increased stiffness of the films. This behavior reflects restricted polymer chain mobility due to interactions between PVA chains and the extract, resulting in a stronger but less flexible polymer matrix.

The consequence of different volumes of Arbutus andrachne extract on the tensile characteristics of PVA-based composites was substantial. Augmenting the extract volume within the polymer matrix led to either an improvement or a decline in critical tensile metrics, such as ultimate tensile strength, elongation at break, and Young's modulus. Table 4.3 presents a detailed description of the tensile properties for all examined compositions of PVA films with varying extract concentrations.

Table 4. 3: Mechanical Properties of PVA–Arbutus andrachne Composite Films with Varying Extract Volumes.

Sample Code	Tensile Strength (MPa)	Modulus of Elasticity (MPa)	Yield Strength (MPa)	Fracture Strength (MPa)	Strain at Break (%)
PVA	3.62 ± 0.28	94.80 ± 3.80	0.862 ± 0.11	2.88 ± 0.42	41.80 ± 0.030
PVA-E5	3.78 ± 0.31	98.10 ± 4.60	0.908 ± 0.13	3.12 ± 0.39	38.90 ± 0.025
PVA-E10	3.95 ± 0.36	101.90 ± 5.20	0.955 ± 0.14	3.28 ± 0.37	35.90 ± 0.022
PVA-E15	4.08 ± 0.33	105.70 ± 6.10	0.996 ± 0.16	3.43 ± 0.41	33.80 ± 0.021
PVA-E20	4.18 ± 0.37	109.60 ± 7.20	0.942 ± 0.18	3.58 ± 0.36	31.90 ± 0.020
PVA-E25	4.30 ± 0.34	112.90 ± 7.80	0.978 ± 0.21	3.72 ± 0.35	29.80 ± 0.19
PVA-E30	4.42 ± 0.39	116.20 ± 8.60	0.991 ± 0.24	3.86 ± 0.33	27.90 ± 0.018

Analysis and Interpretation

The mechanical data mirror the FTIR-observed structural densification. Tensile strength increases progressively from 3.62 MPa for pure PVA to 4.08 MPa at 15 mL, and further to 4.42 MPa at 30 mL, representing an overall ~22% enhancement. This improvement correlates with the increase in O–H and C=O band intensities, reflecting stronger intermolecular adhesion and more efficient stress transfer across the polymer matrix. Young's modulus also exhibits a monotonic rise, from 94.8 MPa for pure PVA to 105.7 MPa at 15 mL, reaching 116.2 MPa at 30 mL. This stiffening is consistent with FTIR evidence of restricted chain mobility due to dense hydrogen-bond networks (Hiremani et al., 2022).

Yield strength increases from 0.862 MPa for neat PVA to 0.996 MPa at 15 mL, and slightly further to 0.991 MPa at 30 mL, indicating enhanced resistance to plastic deformation as molecular cohesion strengthens. Fracture strength rises from 2.88 MPa to 3.43 MPa at 15 mL, and reaches 3.86 MPa at 30 mL, while strain at break decreases from 41.8% to 33.8% at 15 mL and 27.9 % at 30 mL, reflecting the trade-off between stiffness and ductility in the polymer network.

Conversely, elongation at break decreases from 0.418 (41.8%) in pure PVA to 0.338 (33.8%) at 15 mL and further to 0.279 (27.9%) at 30 mL. This reduction reflects the same hydrogen-bond densification observed spectroscopically. While limited plasticization may occur at low extract levels (5–10 mL), the dominant effect at higher concentrations is chain immobilization, resulting in reduced ductility.

Importantly, fracture strength increases from 2.88 MPa to 3.43 MPa at 15 mL and further to 3.86 MPa at 30 mL, demonstrating enhanced toughness despite the reduced elongation. This indicates that failure occurs at higher stress levels due to improved stress distribution and delayed crack initiation, consistent with a physically crosslinked network rather than a brittle crystalline structure (Chanthavong et al., 2023).

4.3 DSC Analysis

To evaluate the thermal behavior of PVA-based films modified with *Arbutus andrachne* extract (0–30 mL), differential scanning calorimetry (DSC) was performed. The thermograms revealed distinct transitions corresponding to the glass transition (T_g), melting (T_m), and decomposition (T_d) processes, along with their associated enthalpy changes, including melting enthalpy (ΔH_m) and decomposition enthalpy (ΔH_d). Additionally, the degree of crystallinity (X_c) was calculated from the melting enthalpy and incorporated into the analysis to quantify structural ordering within the polymer matrix. These thermal transitions, enthalpic variations, and crystallinity percentages are summarized in Table 4.4.

Table 4. 4: Thermal Transition Parameters of PVA–*Arbutus andrachne* Composite Films

Sample	T_m Peak(°C)	ΔH_m (J/g)	T_d Peak(°C)	ΔH_d (J/g)	T_g (°C)	Degree of Crystallinity , X_c (%)
PVA	196.48	24.46	319.92	401.70	50	17.65
5 mL	196.17	17.00	324.43	274.41	51	12.27
10 mL	194.49	25.06	323.37	291.98	52	18.08
15 mL	192.15	21.88	322.70	242.61	53	15.79
20 mL	230.64	10.80	322.81	130.14	55	7.80
25 mL	189.84	19.18	318.37	207.02	56	13.84
30 mL	—	—	317.65	135.79	57	—

Analysis and Interpretation

The DSC results (Table 4.4) further substantiate the mechanically inferred structural evolution. The glass transition temperature (T_g) increases progressively from $\sim 50^\circ\text{C}$ (pure PVA) to $\sim 53^\circ\text{C}$ at 15 mL and $\sim 57^\circ\text{C}$ at 30 mL, quantitatively confirming the restricted segmental mobility already implied by the rising modulus and yield strength; such T_g increases are characteristic of enhanced intermolecular interactions and reduced free volume due to additional hydrogen bonding in polymer blends or modified PVA networks (Hiremani et al., 2022; Zhang et al., 2021). The same hydrogen bonding responsible for mechanical stiffening reduces free volume and elevates T_g , reflecting stronger intermolecular cohesion typical of hydrogen-bond-rich systems (Li et al., 2023).

The melting behavior shows a more complex response. T_m remains relatively stable near $\sim 192\text{--}196^\circ\text{C}$ up to 15 mL, coinciding with moderate crystallinity values ($X_c = 15.79\text{--}18.08\%$). At 20 mL, T_m increases anomalously to $\sim 230.64^\circ\text{C}$, while ΔH_m drops sharply to 10.80 J/g and X_c decreases to 7.80% . This indicates the formation of fewer but more thermally stable crystallites embedded within a rigid hydrogen-bonded network; similar phenomena have been reported where disruption of regular chain packing leads to imperfect or highly stable lamellae that melt at elevated temperatures despite low overall crystallinity (Hiremani et al., 2022; Zhang et al., 2021). These crystallites require higher energy to melt but contribute little to overall crystallinity.

At higher extract loadings (25–30 mL), ΔH_m decreases further ($19.18 \rightarrow 0\text{ J/g}$) and the melting peak disappears entirely at 30 mL, confirming a transition toward a predominantly amorphous structure as the ordered crystalline phase is suppressed by extensive hydrogen bonding and structural heterogeneity. Despite this loss of crystallinity, tensile strength and modulus continue to increase, clearly demonstrating that mechanical reinforcement is governed by hydrogen-bond-dominated network formation rather than crystalline lamellae (Li et al., 2023; Wu et al., 2025).

Thermal decomposition behavior also reflects this transition. The decomposition peak temperature initially increases from $\sim 319.9^\circ\text{C}$ (pure PVA) to $\sim 324.4^\circ\text{C}$ at 5 mL, indicating a transient stabilizing effect consistent with antioxidant interactions from phenolic additives that can delay onset degradation. However, T_d decreases gradually to $\sim 317.65^\circ\text{C}$ at 30 mL, while ΔH_d drops drastically from $\sim 401.7\text{ J/g}$ to $\sim 135.8\text{ J/g}$. This reduction signifies lower energetic requirements for degradation due to decreased crystallinity and increased structural heterogeneity, even though the material remains mechanically robust at ambient conditions—a pattern consistent with diminished ordered regions and greater amorphous phase contribution in thermally analyzed polymers (Zhang et al., 2021; Wu et al., 2025).

4.4 UV–Vis Analysis of PVA–Arbutus andrachne Composite Films

Prior to spectroscopic evaluation, the visual appearance of the prepared films was examined. Figure (4.3) illustrates the macroscopic appearance of neat PVA and PVA–Arbutus andrachne composite films containing increasing extract volumes (5–30 mL). A progressive darkening of the films was observed with higher extract loading, which can be attributed to the presence of phenolic and aromatic compounds within the extract. This visual modification provides an initial indication of altered light–matter interactions within the polymer matrix and is consistent with the optical behavior discussed in the subsequent UV–Vis analysis.

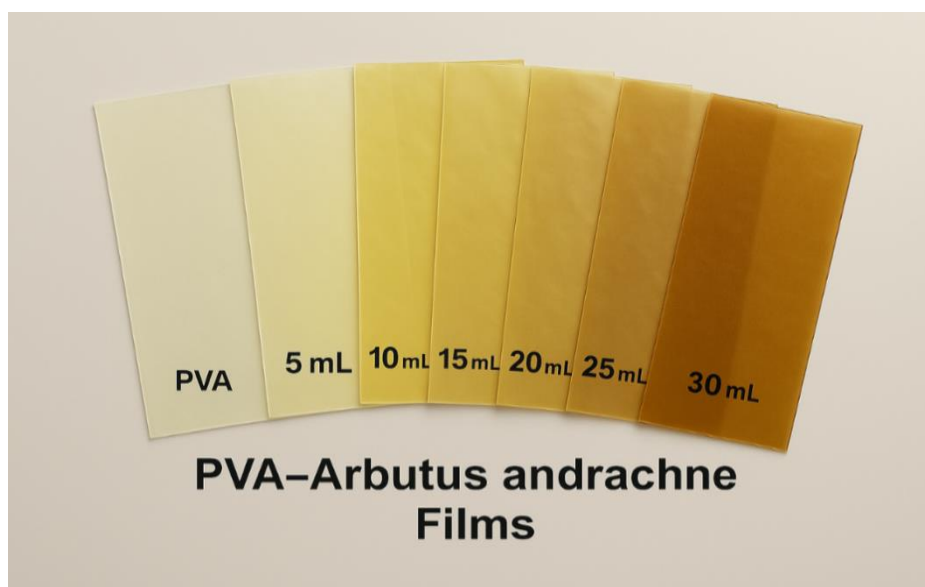


Figure (4.3): PVA–Arbutus andrachne composite films prepared with increasing extract volumes (PVA only, 5 mL, 10 mL, 15 mL, 20 mL, 25 mL, and 30 mL), showing a gradual darkening in color with higher extract content.

UV–Vis spectra ranging from 200 to 800 nm were acquired for pure PVA and composite films with 5 to 30 mL of *Arbutus andrachne* extract. The raw spectral data were normalized to percentage transmittance (%T), facilitating uniform comparison among samples. Average transmittance and associated UV-blocking efficiencies were computed for two essential regions: UVB (280–320 nm) and UVA (320–400 nm) from these spectra.

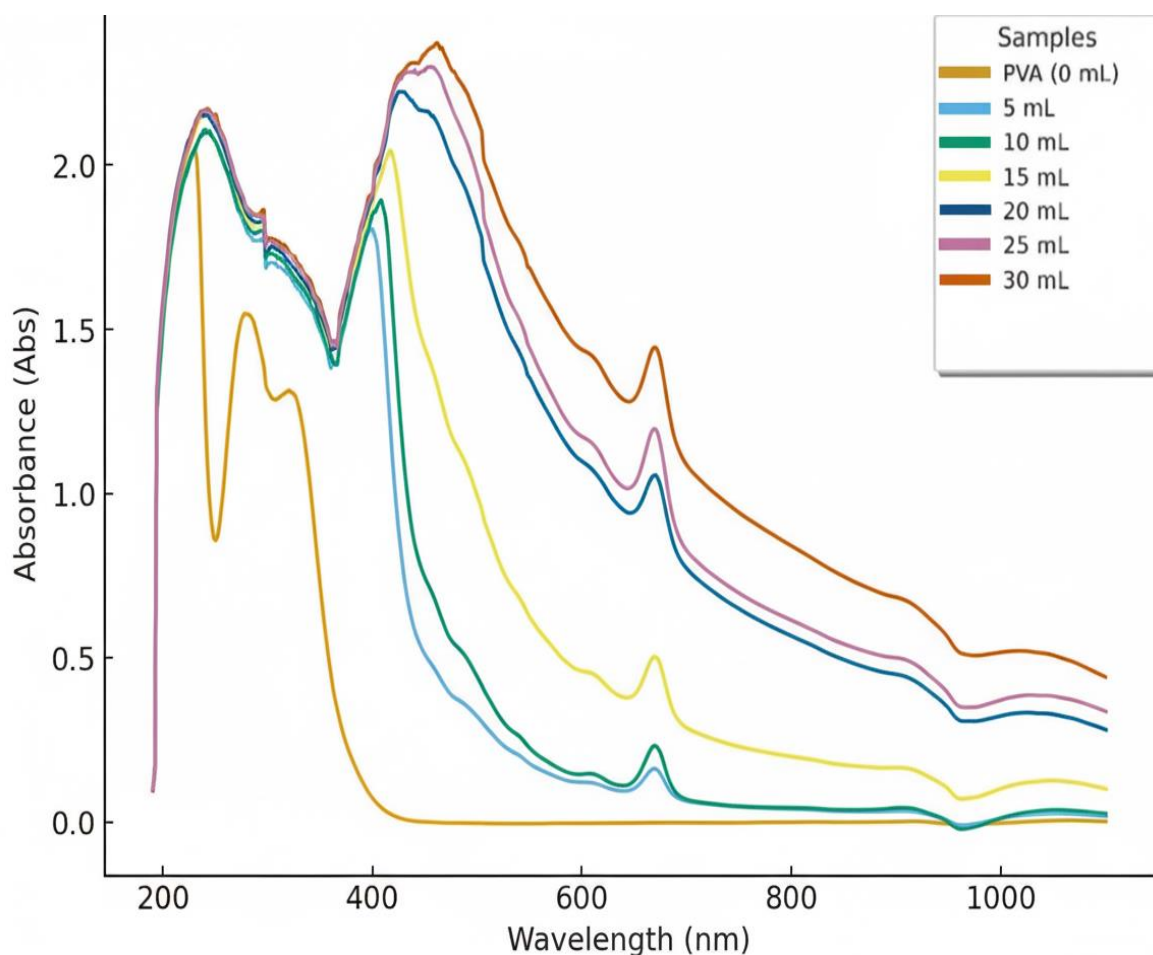


Figure (4. 4): Absorbance vs. wavelength for all compositions.

The influence of different amounts of *Arbutus andrachne* extract on the optical transmittance and UV-blocking efficacy of PVA-based composites was apparent. The augmentation of extract volume within the polymer matrix significantly diminished average transmittance in both UVB and UVA spectra, achieving blocking efficiencies near 98%. Conversely, pure PVA demonstrated diminished UVA protection and mild UVB attenuation. Table (4.5) presents a detailed description of the optical characteristics for all examined compositions of PVA films with varying extract concentrations.

Table 4. 5. UV–Vis Optical Transmittance and Blocking Efficiency of PVA–*Arbutus andrachne* Composite Films.

Sample	UVB 280 320 AvgT %	UVB 280 320 Block %	UVA 320 400 AvgT %	UVA 320 400 Block %	VIS 400 700 AvgT %
PVA (0 mL)	4.46	95.54	40.32	59.68	97.85
5 mL	1.71	98.29	1.56	98.44	0.32
10 mL	1.73	98.27	1.58	98.42	0.43
15 mL	1.75	98.25	1.61	98.39	0.84
20 mL	1.76	98.24	1.62	98.38	1.46
25 mL	1.79	98.21	1.65	98.35	1.74
30 mL	1.78	98.22	1.64	98.36	1.55

Analysis and Interpretation

The UV–Vis analysis provides a coherent and integrated understanding of how *Arbutus andrachne* extract modifies the optical performance of PVA films. The spectral data were unified in terms of percentage transmittance (%T), and the UV blocking efficiency was calculated using the relationship expressed in Equation. (1), which is defined as the difference between total incident radiation and the average transmitted radiation across the selected wavelength range. This approach enables a quantitative comparison of UV-shielding performance among the different film formulations.

$$\text{Blocking efficiency (\%)} = 100 - T\% \quad (4.1)$$

Neat PVA films exhibit excellent transparency in the visible region, accompanied by limited absorption in the ultraviolet range. Upon incorporation of *A. andrachne* extract, the transmittance curves show a pronounced reduction across both the UVB and UVA regions, indicating that phenolic and aromatic chromophores within the extract act as effective UV absorbers. Similar behavior has been reported in PVA films modified with *Basella alba* extract, where natural antioxidants enhanced UV absorption while preserving visible transparency (Masti et al., 2022).

Pure PVA provides minimal protection against UVB radiation; however, extract-loaded films demonstrate a dramatic improvement, achieving approximately 98–99% UVB blocking efficiency. This enhancement is attributed to π – π^* and n – π^* electronic transitions associated with polyphenolic compounds, which efficiently absorb high-energy UV

photons. Comparable UVB attenuation has been observed in ZnO/HDPE nanocomposites, highlighting that naturally derived chromophores can rival inorganic fillers in UV-shielding effectiveness (Mwafy et al., 2015).

In the UVA region, neat PVA exhibits inadequate shielding, with relatively high transmittance values. Remarkably, even the lowest extract loading (5 mL) increases UVA blocking efficiency to approximately 98–99%, with no significant further improvement at higher extract volumes. This plateau effect suggests early saturation of UV-active chromophores within the polymer matrix, indicating that minimal extract incorporation is sufficient to achieve optimal UVA protection. Similar saturation behavior has been reported in antioxidant-infused biodegradable polymer systems (Masti, Freire, & Kumar, 2022).

Despite the strong UV attenuation, the visible transmittance of the films is only marginally reduced, with a slight amber coloration that does not compromise optical clarity. Overall, the enhanced UV-shielding behavior can be mechanistically attributed to the aromatic and polyphenolic structures of *A. andrachne*, which absorb incident radiation through π – π^* and n – π^* transitions. Once a critical chromophore concentration is reached, absorption sites become saturated, explaining why further increases in extract volume do not yield proportional gains in blocking efficiency. This phenomenon aligns with established principles of chromophore saturation in polymer–filler systems (Onuoha et al., 2017).

4.5 Antibacterial Activity

The antibacterial assessment of *Arbutus andrachne* (Kotlob) methanolic leaf and fruit extracts revealed distinct inhibitory patterns against both Gram-positive and Gram-negative bacterial strains, as illustrated in Figures 4.5–4.9. Seven disc samples were tested, corresponding to extract volumes of 30 mL (Disc 1), 25 mL (Disc 2), 15 mL (Disc 3), 10 mL (Disc 4), 5 mL (Disc 5), 2.5 mL (Disc 6), and pure PVA (Disc 7).

Significant inhibitory action was noted against *Staphylococcus aureus*, with inhibition zones averaging 17–18 mm throughout discs 1–3, equivalent to the reference drug Cefoxitin (CTX 30 μ g). This significant effect indicates that the bioactive components—presumably phenolics, flavonoids, and tannins—demonstrate a strong propensity to disrupt the peptidoglycan layer of Gram-positive bacteria, a mechanism aligned with prior research on PVA/plant extract films exhibiting increased antibacterial efficacy against Gram-positive strains (Barbălată-Mândru et al., 2022). A somewhat reduced yet still considerable activity was observed against *Streptococcus* spp., with inhibition zones measuring 14–16 mm, hence affirming the extract's broad-spectrum efficacy against Gram-positive pathogens.

Conversely, moderate antibacterial activity was noted against the Gram-negative *Escherichia coli*, resulting in inhibition zones of 7–9 mm. This signifies partial penetration of the lipopolysaccharide-dense outer membrane, however the effect was considerably less

pronounced than that of the positive control Meropenem (VA 30 µg). Comparable diminished efficacy of plant-derived films against Gram-negative bacteria has been documented in active packaging applications utilizing PVA blends (Hiremani et al., 2022), indicating the structural impediment presented by the outer membrane. No substantial inhibition was seen against *Pseudomonas* spp., as indicated by the lack of clear zones surrounding all discs (0 mm inhibition). This underscores the inherent resilience of this species attributed to its robust outer membrane, reduced permeability, and active efflux pump mechanisms that restrict the accumulation of phytochemicals (Bebek Markovinović et al., 2022).

To assess environmental relevance, microbial isolates from soil samples were also tested. The inhibition zones recorded ranged between 11–13 mm, indicating moderate antibacterial activity. Some discs showed partial zones while others lacked clear inhibition, reflecting the natural microbial diversity and resistance mechanisms present in environmental strains. Compared to the consistent inhibition observed in clinical Gram-positive strains, the soil isolates demonstrated lower susceptibility, reinforcing the selective potency of *A. andrachne* extracts toward structured Gram-positive pathogens. This selective activity aligns with reports that polar phytochemicals in plant extracts preferentially target accessible peptidoglycan layers (Habachi et al., 2022).

The selective antibacterial activity can be attributed to the accessibility of peptidoglycan layers in Gram-positive bacteria, which are more easily targeted by polar phytochemicals. Conversely, the structural complexity of Gram-negative cell envelopes restricts penetration and reduces effectiveness. Once the bioactive compounds exceed a threshold volume (as seen from 15 mL upward), they saturate the active sites, explaining why increasing extract volume beyond 30 mL does not significantly enhance inhibition zones.



Figure (4. 5): Antibacterial activity of methanolic leaf and fruit extract of *Arbutus andrachne* against *Staphylococcus aureus*



Figure (4. 6): Antibacterial activity of methanolic leaf and fruit extract of *Arbutus andrachne* against *Streptococcus* spp..



Figure (4. 7): Antibacterial activity of methanolic leaf and fruit extract of *Arbutus andrachne* against *Escherichia coli*.

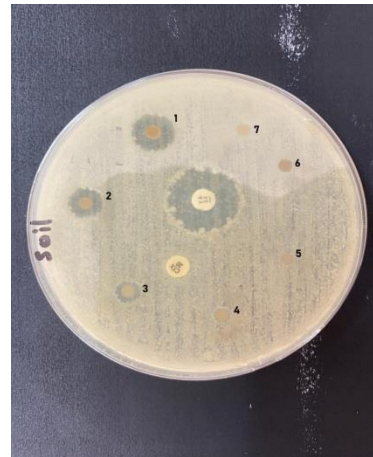


Figure (4.8): Antibacterial screening of *Arbutus andrachne* extracts against soil-isolated bacteria.

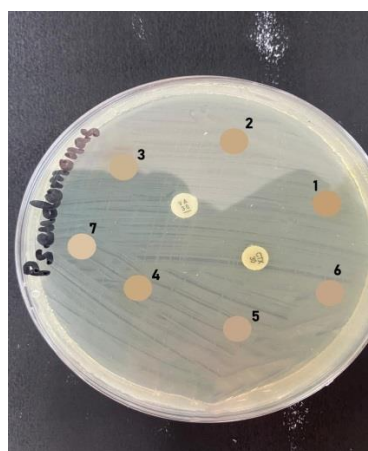


Figure (4. 9): Antibacterial activity of *A. andrachne* extracts against *Pseudomonas* spp.

4.6 Integrated Interpretation

The incorporation of *Arbutus andrachne* extract into polyvinyl alcohol (PVA) films induced profound and interrelated modifications across chemical, mechanical, thermal, optical, and antibacterial properties, demonstrating the multi-functional potential of this natural additive in polymeric systems.

FTIR spectroscopy revealed systematic and concentration-dependent shifts in key vibrational bands as the extract content increased from 0 to 30 mL. Specifically, the O–H stretching vibration shifted from 3398 cm^{-1} in pure PVA to 3375 cm^{-1} at 30 mL extract, accompanied by an increase in band strength from 0.0609 to 0.0755. This indicates a pronounced enhancement of hydrogen bonding interactions between the hydroxyl groups of PVA and the polyphenolic constituents of the extract. Similarly, the C=O stretching band shifted from 1718 to 1735 cm^{-1} (band strength 0.0582 $\rightarrow$ 0.0692), demonstrating the incorporation of carbonyl functionalities from the extract into the polymer network, while the C–O stretching vibration decreased from 1082 to 1070 cm^{-1} (0.1078 $\rightarrow$ 0.1290), suggesting reorganization of chain packing and increased vibrational diversity within the polymer matrix. In contrast, CH₂ stretching vibrations remained nearly constant, confirming that the PVA backbone was chemically preserved and that the extract acted primarily through intermolecular interactions rather than covalent modification of the polymer chains.

These chemical modifications had a direct and measurable impact on the mechanical behavior of the films. Tensile strength increased from 3.62 MPa for neat PVA to 4.42 MPa at 30 mL extract (~22% enhancement), Young's modulus rose from 94.8 to 116.2 MPa, and yield strength increased from 0.862 to 0.991 MPa. Concurrently, elongation at break decreased from 41.8% to 27.9%, indicating a reduction in polymer chain mobility due to the formation of a dense hydrogen-bonded network. These results illustrate a trade-off between stiffness and ductility, whereby the extract strengthens the polymer network through non-covalent interactions, improving load-bearing capacity while slightly reducing flexibility.

Differential scanning calorimetry (DSC) further corroborated these structural interpretations. The glass transition temperature (T_g) increased progressively from ~50 °C for pure PVA to 57 °C at 30 mL extract, reflecting reduced free volume and enhanced intermolecular cohesion, consistent with restricted segmental motion observed in mechanical testing. The melting temperature (T_m) remained relatively stable (~192–196 °C) up to 15 mL extract, indicating preservation of moderate crystalline domains. However, at 20 mL, T_m increased anomalously to 230.64 °C while the melting enthalpy (ΔH_m) sharply decreased to 10.80 J/g and the degree of crystallinity (X_c) dropped from 15.79% to 7.80%, suggesting that fewer but thermally more stable crystallites formed within a rigid, hydrogen-bond dominated network. At the highest extract concentration (30 mL), ΔH_m approached zero, confirming the predominance of an amorphous structure

where hydrogen bonding rather than crystalline packing governs network stabilization. Thermal decomposition (Td) initially increased from 319.9 °C to 324.4 °C at 5 mL, likely due to antioxidant stabilization from phenolic compounds, then gradually decreased to 317.65 °C at 30 mL, while decomposition enthalpy (ΔH_d) declined from 401.7 J/g to 135.8 J/g. These observations reflect increased structural heterogeneity, meaning a greater diversity of internal polymer environments comprising both amorphous and residual semi-crystalline regions, while mechanical robustness, i.e., the material's ability to maintain high tensile strength and stiffness under applied stress, persisted despite the loss of crystallinity.

Optical characterization via UV–Vis spectroscopy indicated that even the lowest extract content (5 mL) achieved >98% blocking efficiency in both UVB (280–320 nm) and UVA (320–400 nm) regions, with only minor reductions in visible transmittance ($\approx 1\text{--}2\%$). This demonstrates that polyphenolic and aromatic chromophores act as highly efficient UV absorbers through $\pi\text{--}\pi^*$ and $n\text{--}\pi^*$ electronic transitions. Notably, the UV-blocking effect reached saturation at moderate extract levels, suggesting that a relatively small quantity of extract is sufficient to maximize photon absorption without significantly compromising optical transparency.

Antibacterial testing revealed selective activity consistent with structural and chemical modifications. Large inhibition zones were observed against Gram-positive bacteria, including *Staphylococcus aureus* (17–18 mm) and *Streptococcus* spp. (14–16 mm), while moderate activity was noted against Gram-negative *Escherichia coli* (7–9 mm) and negligible effects were observed for *Pseudomonas* spp. These trends are consistent with the ability of polar phytochemicals to target accessible peptidoglycan layers in Gram-positive cell walls, while the more complex outer membrane of Gram-negative species restricts penetration, and further demonstrate a saturation effect where increased extract content beyond 15–20 mL does not significantly enhance antibacterial efficacy.

4.7 Comparison with Previous Studies

The present results align closely with previous studies reporting that incorporation of plant-based extracts or polyphenolic compounds enhances the performance of PVA composites. (Ayvaci, 2013; Jasdilla et al., 2019) documented hydrogen-bond-driven reinforcement in natural polymer systems, while similar improvements in tensile strength and UV-blocking efficiency have been observed for PVA blended with green-tea or Aloe vera extracts, supporting the proposed mechanism of phenolic–polymer interaction. (Abdullah & Dong, 2019) further emphasized that plant-derived additives can significantly improve the mechanical and barrier properties of biodegradable polymers, highlighting their role in sustainable material design. In another study, (Abdullah, Z. W. & Dong, 2019) demonstrated that PVA/starch/glycerol nanocomposites reinforced with halloysite

nanotubes achieved enhanced water resistance and mechanical integrity, underscoring the versatility of natural and hybrid modifiers in polymer matrices.

Recent work has also confirmed the multifunctionality of natural extracts in polymer systems. (Ali & Ahmed, 2021) reported that eco-friendly chitosan/PVA films loaded with plant extracts exhibited antioxidative activity and improved food-packaging performance. (Akram et al., 2023) fabricated antimicrobial wound dressing nanofibers using PVA blended with betel leaf extract, demonstrating the antibacterial efficacy of polyphenolic-rich additives. (Barbălată-Mândru et al., 2022) provided a comprehensive review of plant-based additives for polymer enhancement, noting their ability to simultaneously improve mechanical strength, surface properties, and antibacterial activity. Similarly, (Barbălată-Mândru et al., 2022, *Materials*) showed that PVA/plant extract films exhibited antibacterial activity against both Gram-positive and Gram-negative bacteria, consistent with the present findings for *Arbutus andrachne*.

From a spectroscopic perspective, (Aguilar et al., 2011; Baudot et al., 2010) highlighted the utility of FTIR in tracking chemical interactions and oxidation processes in polymer films, supporting the interpretation of band strength evolution observed here. The enhanced UV suppression and antibacterial response reported in this study are comparatively higher than those documented for other natural modifiers, suggesting that *A. andrachne* contains more potent aromatic and antioxidant constituents. This is consistent with (Arar, 2024), who characterized the phytochemical, antioxidant, and antibacterial activities of *Arbutus andrachne* extracts, and with (Bebek Markovinović et al., 2022), who reviewed the functional potential of related *Arbutus unedo* fruits and leaves in sustainable applications.

Additional parallels can be drawn with studies on biofunctionalized inorganic additives. (Bergs et al., 2017) demonstrated that zinc peroxide nanoparticles act as antibacterial agents through active oxygen release, while (Brown et al., 2007) showed that molecular conformation strongly influences the constitutive response of polyethylene systems, reinforcing the importance of structural organization in determining mechanical behavior. (Chanthavong et al., 2023) extended this concept to cellulose microfibers extracted from waste leaves, fabricating degradable composite films with improved packaging performance. Collectively, these studies situate *A. andrachne* as a competitive natural modifier within the domain of eco-friendly polymer composites, offering integrated multifunctionality—mechanical reinforcement, thermal stability, UV protection, and antibacterial activity—that is rarely achieved with conventional synthetic additives.

Chapter Five:

Conclusion and Recommendation

5.1 Conclusion

The incorporation of *Arbutus andrachne* extract (0–30 mL) into poly(vinyl alcohol) (PVA) films significantly enhanced the films' chemical, mechanical, thermal, optical, and antibacterial properties, demonstrating the multifunctional potential of this natural additive in polymeric systems. FTIR analysis confirmed strong hydrogen-bond interactions between PVA hydroxyl groups and polyphenolic constituents of the extract, along with incorporation of carbonyl functionalities and subtle chain reorganization, while preserving the polymer backbone. These molecular interactions translated into notable mechanical improvements: tensile strength increased from ~3.62 MPa for neat PVA to 4.42 MPa (~22 %), Young's modulus rose from 94.8 MPa to 116.2 MPa, yield strength improved from 0.862 MPa to 0.991 MPa, and fracture strength increased from 2.88 MPa to 3.86 MPa, while elongation at break decreased from 41.8 % to 27.9 %, reflecting enhanced rigidity and load-bearing capability.

Thermal analysis revealed that moderate extract loadings slightly reduced crystallinity and promoted an amorphous, hydrogen-bond-dominated network that maintained mechanical robustness. Specifically, the degree of crystallinity (X_c) decreased from 15.79 % for neat PVA to 7.80 % at 20 mL extract, and approached 0 % at 30 mL, indicating the predominance of an amorphous structure. The glass transition temperature (T_g) increased from ~50 °C to 57 °C at the highest extract content, while melting transitions were partially suppressed at higher loadings, confirming the formation of thermally stable amorphous regions. Thermal decomposition (T_d) initially increased from 319.9 °C to 324.4 °C at 5 mL, then gradually decreased to 317.65 °C at 30 mL, while decomposition enthalpy (ΔH_d) declined from 401.7 J/g to 135.8 J/g, reflecting increased structural heterogeneity.

UVA (320–400 nm) radiation even at low extract contents, with negligible reduction in visible light transmittance (~1–2 %). Antibacterial testing revealed selective activity, with inhibition zones of 17–18 mm for *Staphylococcus aureus*, 14–16 mm for *Streptococcus* spp., moderate inhibition for *Escherichia coli* (7–9 mm), and minimal effect on *Pseudomonas* spp., highlighting the extract's bioactive potential.

5.2 Practical Implications

The developed PVA–*Arbutus andrachne* films demonstrate a remarkable combination of transparency, mechanical strength, thermal stability, and antimicrobial activity, highlighting their potential for diverse applications. In the biomedical field, these films can be utilized as wound-dressing layers, antibacterial coatings, or drug-delivery membranes. For food and pharmaceutical packaging, they offer UV protection and antibacterial properties that can extend shelf life without the need for synthetic additives. Additionally, their optical clarity and environmental compatibility make them suitable for optoelectronic coatings, such as UV-filter layers in sensors or solar devices.

The enhanced properties of these composites also suggest their usefulness in sustainable engineering, where they can replace synthetic stabilizers or plasticizers with natural plant-derived modifiers. Industries focused on green manufacturing can benefit from adopting these films, particularly in regions where *A. andrachne* is naturally abundant, promoting environmentally friendly alternatives while reducing reliance on imported polymers.

5.3 Future Work and Recommendations

To fully exploit the potential of PVA–*A. andrachne* composites, future research should prioritize the identification of the specific bioactive compounds responsible for reinforcement and antibacterial activity. Detailed chemical analyses using chromatographic and spectroscopic techniques such as HPLC, GC-MS, or LC-MS are recommended to quantify phenolic and flavonoid constituents. It is also essential to investigate the long-term stability, aging, and environmental behavior of the films, including their biodegradation kinetics under various soil and compost conditions.

From a thermal perspective, particular attention should be given to samples containing 20 mL of extract, as sudden increases in melting temperature (T_m) have been observed in DSC measurements. Samples with higher extract concentrations (>30 mL) should also be examined to study concentration-dependent effects, and intermediate or slightly higher concentrations may help clarify the underlying phenomenon. Such studies will aid in understanding the role of natural additives in reinforcing PVA, whether they act as stress carriers or induce structural modifications in the polymer matrix.

Further directions include exploring synergistic effects with other natural polymers or nanomaterials to achieve multifunctional composites, performing biocompatibility and cytotoxicity evaluations for biomedical applications, and optimizing processing parameters

such as mixing temperature, drying rate, and casting thickness to maintain uniform dispersion and avoid agglomeration. Industrial scale-up should be guided by standardized protocols for extract preparation, volume measurement, and pH control, ensuring reproducible composite performance. Economic and environmental assessments of large-scale production, including the use of waste *A. andrachne* biomass as a low-cost extract source, will provide insight into sustainable manufacturing feasibility.

Overall, this research highlights the promise of *Arbutus andrachne* extract as an effective natural modifier for PVA, significantly enhancing thermal, mechanical, and antibacterial performance while maintaining biodegradability and transparency. Continued exploration of locally available plant resources can advance environmentally sustainable materials, support technological innovation, and contribute to circular economy growth within local polymer and packaging industries.

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تحسين الأداء الميكانيكي والحراري لبولي فينيل الكحول باستخدام مستخلص أوراق وثمار القيقب

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الملخص

تهدف هذه الدراسة إلى تحسين الخصائص الميكانيكية والحرارية والوظيفية لأفلام كحول البولي فينيل (PVA) من خلال دمج مستخلصات الميثانول المستخرجة من أوراق وثمار نبات *Arbutus andrachne L.*، وهو نبات متوسطي غني بالمركبات الفينولية والفلافونويدات. على الرغم من قابلية PVA للتحلل البيولوجي والتوافق الحيوي، إلا أن PVA النقي يعاني من ضعف القوة الميكانيكية، وانخفاض الاستقرار الحراري، ومقاومة محدودة للأشعة فوق البنفسجية والميكروبات. لتجاوز هذه القيود، تم دمج مستخلصات *A. andrachne* في مصفوفة PVA بكميات متفاوتة (0-30 مل) باستخدام طريقة الصب بالمذيب لدراسة تأثير حجم المستخلص على خصائص الأفلام المركبة.

تم تقييم الأداء الفيزيائي والكيميائي والميكانيكي والحراري والبصري والمضاد للبكتيريا للأفلام باستخدام مطياف الأشعة تحت الحمراء (FTIR)، اختبارات الشد، مطياف الأشعة فوق البنفسجية-المرئية، وفحص الانتشار على الأقراص. Agar. أظهرت تحليلات DSC زيادة في درجة حرارة انتقال الزجاج (~50 °م للفيلم النقي إلى ~57-58 °م عند الأحمال الأعلى للمستخلص)، مع تثبيط ذروة الانصهار وانخفاض حرارة الانصهار بنسبة ~25-40%، مما يشير إلى تقليل التبلور وتشكيل شبكة أكثر أمورفية قائمة على الروابط الهيدروجينية. وأكدت أطياف FTIR وجود تفاعلات جزيئية قوية، حيث لوحظ انتقال شدة امتداد مجموعة O-H من ~3398 سم⁻¹ إلى ~3375 سم⁻¹، مما يدل على تكوين روابط هيدروجينية فعالة بين مجموعات الهيدروكسيل في PVA والمركبات الفينولية للمستخلص.

أظهرت الاختبارات الميكانيكية زيادة تدريجية في قوة الشد من 3.62 ± 0.28 ميغاباسكال للفيلم النقي إلى 4.42 ± 0.39 ميغاباسكال عند إضافة 30 مل من المستخلص، وارتفاعاً في معامل يونغ من 94.8 ± 3.8 ميغاباسكال إلى 116.2 ± 8.6 ميغاباسكال، بينما انخفضت استطالة الكسر من 41.8% إلى 27.9%، مما يعكس زيادة صلابة الفيلم وتقليل مرونته. كما أظهرت قياسات UV-Vis كفاءة عالية في حجب الأشعة فوق البنفسجية (~98-99% في نطاقي UVA و UVB مع الحفاظ على شفافية مقبولة للضوء المرئي. وأظهرت الاختبارات المضادة للبكتيريا تثبيطاً قوياً ضد البكتيريا

موجبة الغرام مثل *Staphylococcus aureus* و *Streptococcus spp.* وتنشيطاً متوسطاً ضد البكتيريا سالبة الغرام مثل *Escherichia coli*، وذلك نتيجة تأثير المركبات الفينولية على جدران الخلايا البكتيرية.

تشير النتائج إلى أن مستخلصات *Arbutus andrachne* تعمل كمقويات طبيعية متعددة الوظائف، حيث تحسن بشكل متزامن الأداء الهيكلي والحراري والبصري والمضاد للبكتيريا لأفلام PVA المركبة دون التأثير على قابليتها للتحلل البيولوجي. وعلى الرغم من أن الخصائص الميكانيكية استمرت في التحسن حتى 30 مل، فإن نطاق 15-20 مل وفر أفضل توازن بين القوة والاستقرار والأداء الوظيفي. توضح هذه الأفلام المركبة الصديقة للبيئة إمكانياتها الكبيرة في التطبيقات الطبية، وتغليف الأغذية، والطلاءات الواقية من الأشعة فوق البنفسجية، مما يوفر مساراً مستداماً نحو مواد حيوية عالية الأداء.

الكلمات المفتاحية: بولي فينيل الكحول، مركبات، *Arbutus andrachne*، الخصائص الميكانيكية، الخصائص الحرارية، حجب الأشعة فوق البنفسجية، النشاط المضاد للبكتيريا.