Ref. Ares(2026)5452866 - 29/05/2026

THESEUS

Hub4Circularity

A First of a kind Hub for circularity demonstrator for Attica and peripheral regions

Grant Agreement No 101178059

DELIVERABLE 5.10:

Biobased thin films

Work Package: 5

LEAD BENEFICIARY:

NCSR (NATIONAL CENTER FOR SCIENTIFIC RESEARCH "DEMOKRITOS")

Delivery Date: 31.05.2026

Document Sheet

Project acronym	**Theseus**
Project full title	A First of a kind Hub for circularity demonstrator for Attica and peripheral regions
Programme	Horizon Europe
Topic	HORIZON-CL4-2024-TWIN-TRANSITION-01-38
Type of Action	HORIZON Innovation Actions
Grant Agreement	101178059
Start day	1 December 2024
Duration	60 months

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Document Information

Deliverable number	D5.10
Deliverable name	Biobased thin films
Lead beneficiary	NCSR Demokritos
WP	5
Related task(s)	5.4
Type	R
Reviewers	Dr. Filippos Tourlomousis (NCSR), Christopoulou Georgia (TSIMIS), Dr. Constantinos Samios (TSIMIS)
Delivery date	27.05.2026
Main author(s)	Dr. Nefeli Pontillo (NCSR)
Contributor(s)	-

Dissemination level

PU	Public	X
SEN	Sensitive, limited under the conditions of the Grant Agreement	
Classified R-UE/EU-R	EU RESTRICTED under the Commission Decision No2015/444	
Classified C-UE/EU-C	EU CONFIDENTIAL under the Commission Decision No2015/444	
Classified S-UE/EU-S	EU SECRET under the Commission Decision No2015/444	

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Document history

Version	Date	Changes	Reviewer/Contributor
V0.1	01.05.2026	Initial Version	Dr. Nefeli Pontillo (NCSR)
V1.0	27.05.2026	Final Version	Dr. Nefeli Pontillo (NCSR), Babis Manousiadis (ICCS)

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Publishable summary

This deliverable reports the progress achieved during the first year of Task 5.4, which focuses on the development of biobased thin films for packaging applications within the THESEUS project. The work contributes to Pilot #2, Biobased Materials and Packaging Symbiotic Flows, and aims to identify sustainable film-forming systems that can incorporate biobased fractions recovered from the Blue Bins of Attica.

During the first reporting period, NCSR Demokritos established the practical and methodological basis for the task. The target film requirements were defined in consultation with the end-user partner, TSIMIS S.A., with emphasis on structural, mechanical and surface properties, and stability during printing. A sustainability-oriented laboratory strategy was adopted, prioritising biobased, biodegradable, abundant, and relatively affordable materials, as well as mild processing routes based mainly on water or aqueous systems.

A set of commercial biopolymers and additives was selected to establish reproducible preparation and characterisation workflows before introducing the waste-derived fractions in later stages. The polymers investigated included carboxymethyl cellulose (CMC), hydroxyethyl cellulose (HEC), gelatin, and two chitosans of different molecular weight. Several additives, including glycerol, lignin, sorbitol, urea, and ascorbic acid, were examined to assess their effect on film properties. Solution casting followed by solvent evaporation was selected as the main preparation route. A tensile-testing workflow was also established, including specimen preparation, mechanical testing, and data-processing procedures.

The experimental results showed that the mechanical behaviour of the films depends strongly on both polymer identity and composition. The high MW chitosan showed the highest stiffness among the neat polymers and, together with gelatin, gave the highest tensile strength. CMC-based films showed a promising combination of stiffness, tensile strength, and elongation at break, supporting their selection as a starting point for further optimisation. Additive effects varied considerably across the different systems.

In parallel with the experimental work, progress was made on the design of an autonomous platform intended to support and accelerate film preparation and preliminary testing. This work was carried out through close collaboration between the laboratory team and the engineering team within NCSR, allowing the manual film-development workflow to inform the platform design, while engineering considerations helped refine the process for future automation. The proposed platform is based on two coordinated Geneva wheels and includes stations for solvent preparation, polymer addition, dissolution, film formation, basic testing, optical checking, and cleaning for reuse. Although the platform remains under development, its design provides a foundation for future automated preparation, structured data generation, and data-driven optimisation of biobased film compositions.

The next stage of the work will focus on refining the most promising polymer-additive systems, expanding material screening, assessing additional target properties, and continuing the assembly and integration of the autonomous platform.

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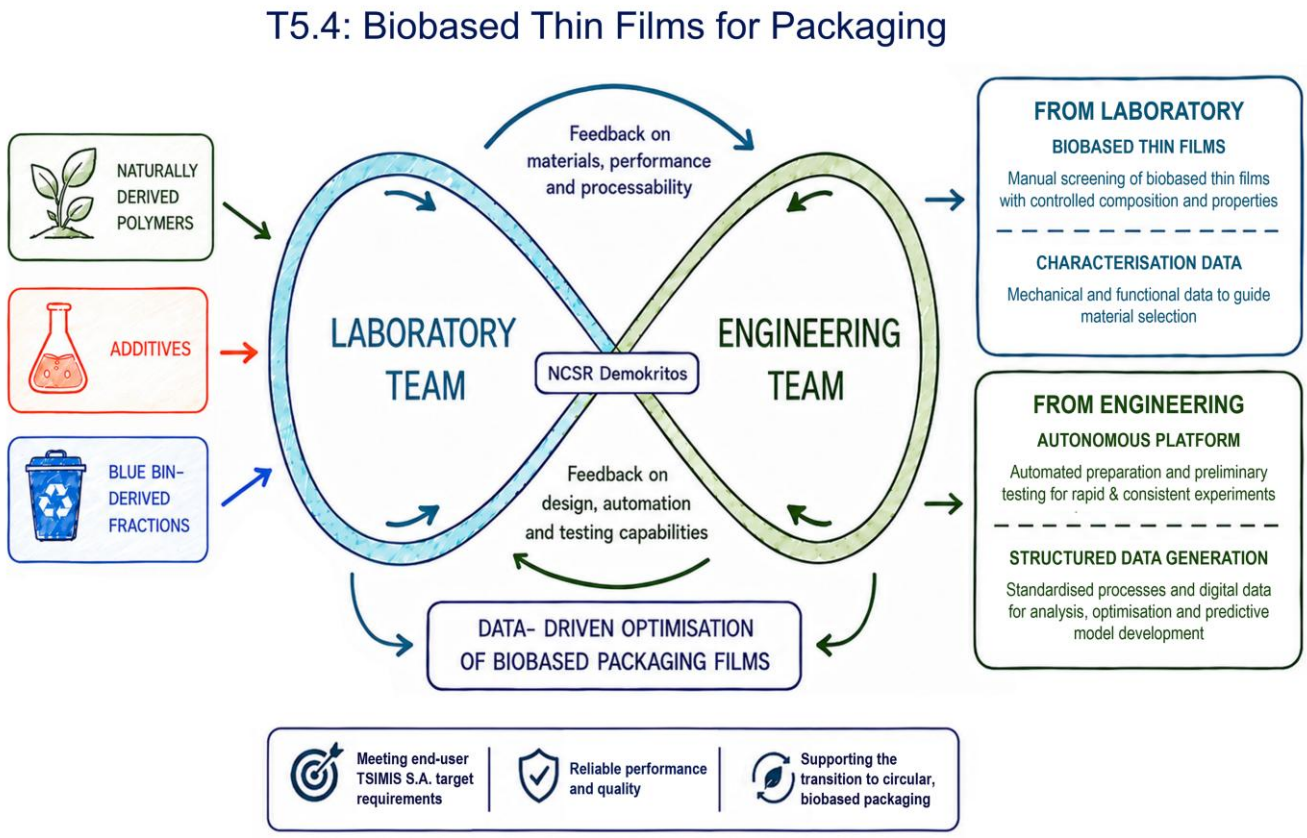


Figure 1. Overview of the Task 5.4 approach for developing biobased thin films for packaging

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Abbreviations

Abbreviations	Full name
CMC	Carboxymethylcellulose
HEC	Hydroxyethylcellulose
MW	Molecular Weight
UTS	Ultimate tensile stress
PEG	Polyethylene glycol
SDS	Safety Data Sheet
WP	Work Package

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1 Introduction

This report presents the progress achieved during the first year of Task 5.4 - *Biobased thin films for packaging*, within the THESEUS project. Task 5.4 is part of Work Package 5 - *Material Processing Technologies* and contributes to Pilot #2 - *Biobased Materials and Packaging Symbiotic Flows*. Within this pilot and among other activities, mixed recycled materials collected from Blue Bins are separated into distinct material streams, including the biobased fraction relevant to the present work.

NCSR Demokritos (NCSR) is responsible for developing biobased thin films for packaging applications, using materials derived from the relevant biobased streams. The task is carried out in collaboration with TSIMIS S.A. (TSIMIS), the end-user partner, which has contributed to defining the target film specifications and practical application requirements.

Task 5.4 started in Month 6 and has a total duration of 42 months. Three deliverables are planned for Months 18, 36, and 42. The present document is the first deliverable of the task and summarises the progress made during its first twelve months. At this stage, the work has focused on defining the application requirements, establishing film preparation and characterisation workflows, screening candidate materials and preparation routes, and aligning the manual laboratory methodology with the requirements of the future autonomous platform.

1.1 Purpose of Task 5.4

The overall objective of this Task is to develop biobased thin films for packaging applications. The work involves identifying suitable polymer systems and assessing how biobased fractions recovered from the Blue Bins can be incorporated into these films. During the first reporting period, the work focused on defining a development strategy that can accommodate heterogeneous and variable feedstocks. This included the use of selected commercial model polymers and additives to establish reproducible preparation and characterisation workflows before introducing Blue Bin-derived fractions in later stages. In line with the circularity and sustainability principles of the THESEUS project, the work has prioritised biobased, biodegradable, abundant, and relatively affordable materials, alongside processing routes that minimise the use of harsh chemicals and avoid unnecessarily energy-intensive steps.

A further objective of the task is the development of an autonomous platform capable of supporting film preparation and basic testing in a structured and repeatable manner. For this reason, the work carried out during this reporting period has addressed both material development and alignment of the laboratory methodology with the requirements of future automation.

Accordingly, the main objectives of the work carried out during this reporting period can be summarised as follows: (i) to define the target requirements in collaboration with the end-user; (ii) to establish the laboratory infrastructure and workflows required for film development; (iii) to screen candidate biobased polymers and additives, using model systems that can later support the incorporation of Blue Bin-derived biobased fractions; (iv) to identify a practical and sustainability-oriented baseline strategy for further optimisation; and (v) to support the design and initial development of an autonomous platform capable of reproducing and accelerating the development process in later stages of the project.

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2 Year 1 Development Strategy and Preparatory Activities

The development of biobased thin films for packaging involves a broad formulation space, with many possible combinations of polymers, plasticisers, additives, and processing conditions. Material behaviour depends on intrinsic properties, as well as on factors such as molecular properties, concentration, solvent system, drying conditions, and composition. In addition, procedures that are feasible at laboratory scale may not always be directly transferable to an automated process. A structured development strategy is thus needed to identify the most promising options and to ensure that the selected preparation routes remain compatible with the future autonomous platform.

2.1 Stepwise strategy adopted by NCSR

The work carried out by NCSR during the first year of Task 5.4 was conceived as an initial screening and methodology-building phase. It aimed to define realistic development pathways, identify promising baseline materials, and establish consistent criteria for comparing candidate systems.

To address the complexity of the formulation space, a stepwise development strategy was adopted. The first step was to clarify the application requirements, so that the experimental work could be guided by realistic end-use targets. The practical basis for systematic experimentation was then established through the procurement of key chemicals and consumables, preparation of the laboratory infrastructure, and evaluation of suitable film-fabrication methods.

Five candidate biopolymers and six additives were selected for screening. Different preparation methods and compositions were explored to identify an approach that was robust, reproducible, and consistent with the sustainability objectives of the project. In parallel, the mechanical testing workflow was established, as mechanical performance was identified early as the most suitable initial screening criterion for assessing the different compositions.

The experimental work also informed the design of the autonomous platform, allowing possible incompatibilities between manual and automated workflows to be identified at an early stage. This helped ensure that decisions on fabrication methodology, material systems, and process steps remained realistic for both laboratory implementation and later automation.

2.1.1 Application Requirements and Technical Targets

The target film properties were defined in consultation with TSIMIS, which provided the desired performance specifications and information regarding the process flow currently used at its facility. This input has served as a core reference point for the material-development strategy and has helped ensure that the experimental work remains aligned with realistic application needs.

Mechanical properties were selected as the main screening criterion, as they represent a key requirement for the intended application and provide a practical basis for comparing candidate materials. Other target properties, including thermal behaviour, optical transparency, solvent resistance, and surface tension, remain relevant and are expected to be examined experimentally during the next stages of the task, once the most promising formulations have been identified.

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The main target requirements for the films developed within Task 5.4 are summarised in Table 1. The target material is a thermoplastic film with a thickness in the range of 20-50 µm. From a thermal point of view, the film should remain dimensionally stable when exposed to temperatures of up to 80 °C. With respect to mechanical performance, the film must be sufficiently stiff to maintain its shape during handling and printing. This is expressed through Young's modulus (E), which is a measure of stiffness: the higher the value, the stiffer the material and the better it resists deformation. For the intended application, the target is a Young's modulus of at least 1 GPa.

In addition, transparency is considered desirable, although it is not essential. The material should also be resistant to solvents commonly encountered in the intended application. Finally, the target surface tension is 38 dynes/cm (mN/m), which is important for printing because it helps inks spread evenly across the film surface.

Table 1. Target film properties set in consultation with TSIMIS

Property category	Requirement / target
Material type and geometry	Films should be produced from thermoplastics rather than thermosetting plastics. The target film thickness is between 20 and 50 µm.
Thermal behaviour	The film should not exhibit permanent deformation when exposed to temperatures up to 80 °C. Shrinkage should remain minimal, with a maximum of about 1–2% in both the Machine Direction (MD) and the Transverse Direction (TD).
Mechanical properties	A relatively high Young's modulus is desired in order to ensure sufficient stiffness without making the film excessively fragile. A safe condition is that more than 100 N force should be required to reach 0.5% elongation for a film of 1000 mm width and 20 µm thickness. This requirement reflects the need to avoid permanent deformation during the printing process.
Optical properties	Transparency is desirable, although it is not considered an absolute requirement at this stage.
Additional performance requirements	Thermo-welding properties are not required. The final material should be resistant to solvents such as ethyl acetate, ethanol, and propanol.
Surface properties	The target surface tension is 38 dynes/cm (mN/m), so that inks can fully wet the film surface without forming droplets.

2.1.2 Multidisciplinary development approach

The manual laboratory development of the films is closely linked to the future autonomous synthesis and testing platform. Accordingly, the work carried out by NCSR has relied on close interaction within the group between two complementary strands of expertise: materials and laboratory science, and design and engineering.

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The materials and laboratory team has been responsible for material selection, literature review, manual handling, film fabrication, experimental setup, testing, and the definition of the processing strategy. In parallel, the engineering group, comprising software, mechanical, design, and electrical engineers, has been responsible for the development and future fabrication of the autonomous platform. The two groups have worked closely together to translate the laboratory process into a form compatible with automation. This has involved continuous exchange between the teams, with repeated discussion of what could or could not be implemented in practice from both the laboratory and engineering perspectives. As a result, the manual workflow has been progressively refined in a way that remains realistic for later automation.

This multidisciplinary collaboration has supported a holistic approach, enabling decisions to be taken on the basis of both scientific performance and implementation feasibility. This approach helps prevent the development of manual procedures that may later be difficult to automate and better connects material discovery with process reproducibility and future AI-assisted composition optimisation.

2.2 Perspective for future pilot-scale validation

The possibility of future pilot-scale testing was also considered. The end-user has indicated willingness to explore such testing if a promising film formulation is identified. However, pilot-scale implementation would be highly challenging. The current work is being carried out at laboratory scale, whereas any transition to pilot scale would require a much more extensive investigation of robustness of the selected composition, processing conditions, and scalability. As such work lies beyond the present scope of the task, pilot-scale validation should be regarded as a possible future opportunity rather than an expected project outcome.

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3 Laboratory Setup and Preparatory Activities

During the first reporting period, an important part of Task 5.4 focused on establishing the practical basis for the systematic development of biobased thin films. Before extensive formulation screening could begin, the necessary chemicals, consumables, and laboratory resources had to be secured, and the laboratory had to be prepared for film preparation and characterisation.

3.1 Selection of Materials and Film Fabrication Approach

The selection of materials and the development of the fabrication methodology were approached in parallel. This was necessary in order to ensure that the candidate material systems were relevant for film formulation, but also compatible with a practical and reproducible preparation route. The work carried out in this phase therefore aimed to establish a rational baseline set of polymers, additives, and processing conditions.

The fabrication process was designed with sustainability as a central consideration. Accordingly, water was selected as the main solvent, and harsh chemicals and unnecessary energy-intensive processing steps were avoided. Although such an approach was not defined as an immediate technical requirement in the proposal, a more sustainability-oriented methodology was deliberately adopted in the broader spirit of the THESEUS project and in alignment with the principles of green chemistry.

3.1.1 Baseline polymers and additives investigated

Candidate systems needed to be capable of forming continuous films, to be processable under mild conditions, and to offer the potential to achieve the required mechanical performance after formulation optimisation. Priority was given to materials that are biobased, biodegradable, abundant, and relatively affordable.

An initial group of biobased polymers known for their film-forming ability was selected and procured for screening. These included carboxymethyl cellulose (CMC), hydroxyethyl cellulose (HEC), gelatin, and chitosan of different molecular weights. These polymers were considered relevant because they represent distinct classes of biobased materials with different expected mechanical, processing, and functional characteristics and, thus, are widely used in film-forming systems.

- Chitosan is a biodegradable, film-forming polysaccharide with generally good mechanical and thermal properties, which makes it attractive for packaging films and membranes. Its main limitation is its hydrophilic character, so chitosan films are often modified or blended to improve moisture resistance, barrier performance, and active functionality. [1-4]
- Carboxymethyl cellulose (CMC) is a water-soluble cellulose derivative valued for its low cost, biodegradability, transparency, and excellent film-forming ability, making it relevant for edible coatings and food-packaging films. However, neat CMC films may have limited water resistance, so reinforcement with fibres, nanomaterials, or active additives is often required. Despite these limitations, CMC remains highly attractive as a baseline film-forming polymer because of its processability under mild aqueous conditions. [4-6]
- Hydroxyethyl cellulose (HEC) is a water-soluble, non-ionic cellulose derivative with good film-forming, adhesive, and moisture-protecting properties, and it has applications in food, pharmaceutical, and coating systems. In neat-film form, however, HEC may show low toughness and

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limited mechanical strength, so composite design is often used to improve performance. For this reason, HEC was considered of interest both as a stand-alone matrix and as a possible blending component within a broader formulation strategy. [7-10]

- Gelatin is a protein-based biopolymer with strong film-forming ability and wide use in edible and active packaging films. Neat gelatin films are not fully hydrophobic, but their properties can be substantially improved through blending or the addition of functional agents to enhance hydrophobicity, barrier behaviour, and active-packaging performance. [3-4,11-12]

Taken together, these polymers provide a useful starting set for formulation screening because they combine practical film-forming ability with complementary property profiles. In addition, all are processable in water or aqueous systems, which is consistent with the sustainability-oriented methodology adopted in this task. In parallel with the baseline polymers, selected additives were also investigated in order to understand how the properties of the films could be tuned. Glycerol was introduced as a plasticiser to improve flexibility and handling, while water-soluble alkali lignin was explored as a functional additive of interest because of its relevance to the selected feedstock strategy and its potential to contribute additional functionality. In addition to their relevance for film development, these materials were also considered suitable from a practical and safety perspective. According to the corresponding Safety Data Sheets (SDS), the selected materials are classified as non-hazardous. They are also naturally derived, biodegradable, compatible with the intended application, and generally regarded as non-toxic. Furthermore, all selected materials are commercially available, and their corresponding safety documentation (SDS) can be readily accessed online.

3.1.2 Feedstock strategy

An additional activity during the reporting period concerned the identification of a relevant waste-derived material stream from the Blue Bins to be incorporated for the future development of the films. This required discussion with the project coordinators, as the selected route needed to reflect both the scientific interest of the task and the types of material that could realistically be extracted and supplied by the recycling-related partners within the project. Following these discussions, wood pieces were identified as a suitable candidate stream for NCSR. This choice was considered relevant because wood is an abundant biobased material and should not be regarded as waste. On the contrary, wood contains valuable structural biopolymers that may be retrieved and reused in high-value material applications. [13-14]

Wood is primarily a lignocellulosic composite composed of three main biopolymer fractions: cellulose, hemicelluloses, and lignin. Lignocellulose constitutes the structural basis of plant cell walls and typically accounts for the majority of the dry matter. Representative compositions often reported in the literature are approximately 50% cellulose, 25% hemicelluloses, and 25% lignin, although the exact proportions depend on plant species, plant part, and growth conditions. [13-14] Cellulose is the main structural polysaccharide and is highly relevant in the context of film-forming biobased materials. Hemicelluloses, which form the second major carbohydrate fraction, are lower-molecular-weight, branched, non-crystalline polysaccharides that contribute to the cohesion and organisation of the lignocellulosic structure, including through interactions with both cellulose and lignin. [13]

Particular interest in the present work is placed on lignin. Lignin is a complex, amorphous, highly branched aromatic biopolymer found mainly in the secondary cell walls of plants, where it contributes to structural rigidity, hydrophobicity, and resistance to degradation. [15-17] From the perspective of sustainable films, lignin is especially attractive because it has been associated with antioxidant, UV-shielding, antimicrobial,

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hydrophobic, and reinforcing effects, which may be translated into valuable active or protective functions in packaging materials. [18-23] At the same time, its practical incorporation is not straightforward, since lignin may also introduce challenges related to structural heterogeneity, aggregation tendency, dark colour, and limited compatibility with some polymer matrices. [18-23]

At the current stage of the work, direct processing of waste wood into controlled film formulations would introduce significant variability and additional complexity. For this reason, the first experimental approach focused on the use of a commercial lignin as a model component, allowing the effect of lignin incorporation to be examined under more controlled and reproducible conditions. In particular, alkali lignin was selected as the initial lignin source.

Alkali lignin is a technical lignin fraction obtained from alkaline pulping or alkaline treatment of lignocellulosic biomass and therefore represents a process-derived, structurally modified form of native lignin. It is attractive for film and composite applications because it is abundant, low-cost, renewable, and functionally active, with reported antioxidant, UV-blocking, and polymer-interacting properties. In biopolymer films, alkali lignin has been reported to improve active functionality, barrier performance, and thermal stability, and may also participate in intermolecular interactions or crosslinking with polymers such as chitosan. [2,12,22,24-25]

The use of commercial alkali lignin in the present stage was a practical and controlled starting point for investigating lignin-containing film formulations. This allowed the contribution of a wood-derived functional fraction to be studied under controlled and manageable experimental conditions. The findings from this model system are expected to later support the use of lignin fractions obtained from the Blue Bins.

3.1.3 Film fabrication approach

Several possible approaches for preparing thin films were explored, including casting, solvent exchange, and microwave-assisted film formation. The purpose of this initial methodological exploration was to identify a fabrication route that would be experimentally practical, reproducible, sufficiently mild for sensitive biopolymers, and consistent with the broader sustainability objectives of the project.

Following this assessment, the preferred preparation route at the current stage was solution casting followed by solvent evaporation. In practice, unless otherwise stated, the polymers and/or additives are dissolved in a solvent to form a film-forming solution, which is then cast onto a surface. The solvent is then allowed to evaporate until a film is obtained. In the systems examined in this task, water was used as the main solvent wherever feasible.

This approach was selected because it combines simplicity, flexibility, and compatibility with the materials under study. It allows formulation parameters to be varied in a controlled manner and is well suited to the screening of polymer–additive systems during the early development stage. In addition, the absence of external heating during drying reduces energy demand and limits the risk of damaging polymers that may be sensitive to elevated temperatures. The method is also compatible with later transfer to a semi-automated or fully automated preparation workflow.

3.2 Establishment of the Mechanical Testing Workflow

Mechanical properties constitute one of the most important performance criteria for thin films intended for packaging applications, as such materials must provide sufficient stiffness and dimensional stability during

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handling, processing, and use. In the present case, the end-user also highlighted the importance of a relatively high Young's modulus in order to avoid permanent deformation during printing. For this reason, mechanical characterisation was selected as the primary screening tool.

3.2.1 Tensile-testing protocol

Establishing the mechanical-testing workflow required adaptation of the available setup to the specific requirements of thin-film samples. A universal testing machine was already available, however, its original configuration was not appropriate for thin-film testing. Following some procurement delays, the setup became fully operational and suitable for systematic film testing. Tensile tests are performed using a Testometric universal testing machine equipped with a 500 N load cell and pneumatic grips, suitable for thin-film specimens.

All tensile tests are carried out according to ISO 527-3 *Plastics - Determination of tensile properties*. More specifically, the films are cut into Type 2 specimens, following the geometry prescribed by the standard. To ensure reliable and reproducible sample preparation, a custom-made 3D-printed cutting tool was designed and fabricated specifically for the films developed in this task. The cutter consists of a 3D-printed ABS body and commercially available razor blades that form the cutting edge. To prepare the specimens, the film is placed on the cutting surface, and the upper part of the cutter is aligned and secured. A vice is then used to apply uniform pressure across the tool surface, enabling consistent cutting and reducing edge defects and dimensional deviations between specimens. The specimens have a width of 17.5 mm and an overall length of 150 mm. The gauge length is 50 mm, and the initial distance between the grips is set to 100 mm.

Once cut, each specimen is measured with a micrometre to determine its thickness before testing. For each specimen, at least five thickness measurements are taken at different positions along the gauge length, and the corresponding average value is used for the subsequent stress-strain calculations. For each film formulation, a minimum of five replicate specimens are prepared and tested in order to ensure repeatability and reproducibility of the results. The tensile tests are performed at a speed of 5 mm/min. The raw force-elongation data generated during the experiment are automatically recorded by the testing system for further processing.

3.2.2 Data-processing and analysis workflow

Given the large number of specimens and formulations expected throughout the task, the team also developed an internal digital tool to accelerate the processing of tensile-test results, containing two processing modules. The first is designed to recognise the raw data exported by the testing system, request the specimen width and thickness from the user, and convert the recorded force-elongation data into stress-strain curves according to the equations defined in the standard. The tool then allows the user to determine whether end trimming of the curve is required and whether toe correction should be applied, thereby improving consistency across multiple datasets. Then, the tool calculates the main tensile parameters considered in this work, namely Young's modulus, ultimate tensile stress, and elongation at break, which are used as practical measures of stiffness, maximum resistance before failure, and extensibility, respectively.

After the analysis of all individual specimens has been completed, a second processing tool is used to compare the results obtained from the different repetitions of a given formulation. This tool generates a

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combined graph representing the mean response together with the corresponding ±1 standard deviation envelope and produces a summary table reporting the mean values and associated variation.

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4 Experimental results

The results presented in this section correspond to the first set of film compositions that were prepared, conditioned, and mechanically tested during the current reporting period. The dataset serves for comparing the behaviour of neat biopolymers, polymer blends, and additive-containing films. All samples were prepared using deionised water as solvent, except for chitosan-based systems, which were dissolved in 2% glacial acetic acid in order to achieve complete solubilisation.

The materials used in the films discussed here were as follows: high MW chitosan (200–600 mPa·s, 0.5% in 0.5% acetic acid at 20 °C, minimum deacetylation value 80.0%, TCI), low MW chitosan (5–20 mPa·s, 0.5% in 0.5% acetic acid at 20 °C, minimum deacetylation value 70.0%, TCI), glycerol (Honeywell), carboxymethyl cellulose (Chemco), hydroxyethyl cellulose (Sigma-Aldrich), gelatin tested according to Ph. Eur. (Sigma-Aldrich), and alkali lignin, water-soluble (Sigma-Aldrich). All samples were dried prior to use in a dryer at 40 °C and were otherwise used as received, without further purification.

4.1 Mechanical performance of neat polymers

The neat polymer films provide the baseline for interpreting all polymer combinations and additive-containing films. The tensile results for the neat polymers are presented in Figure 2.

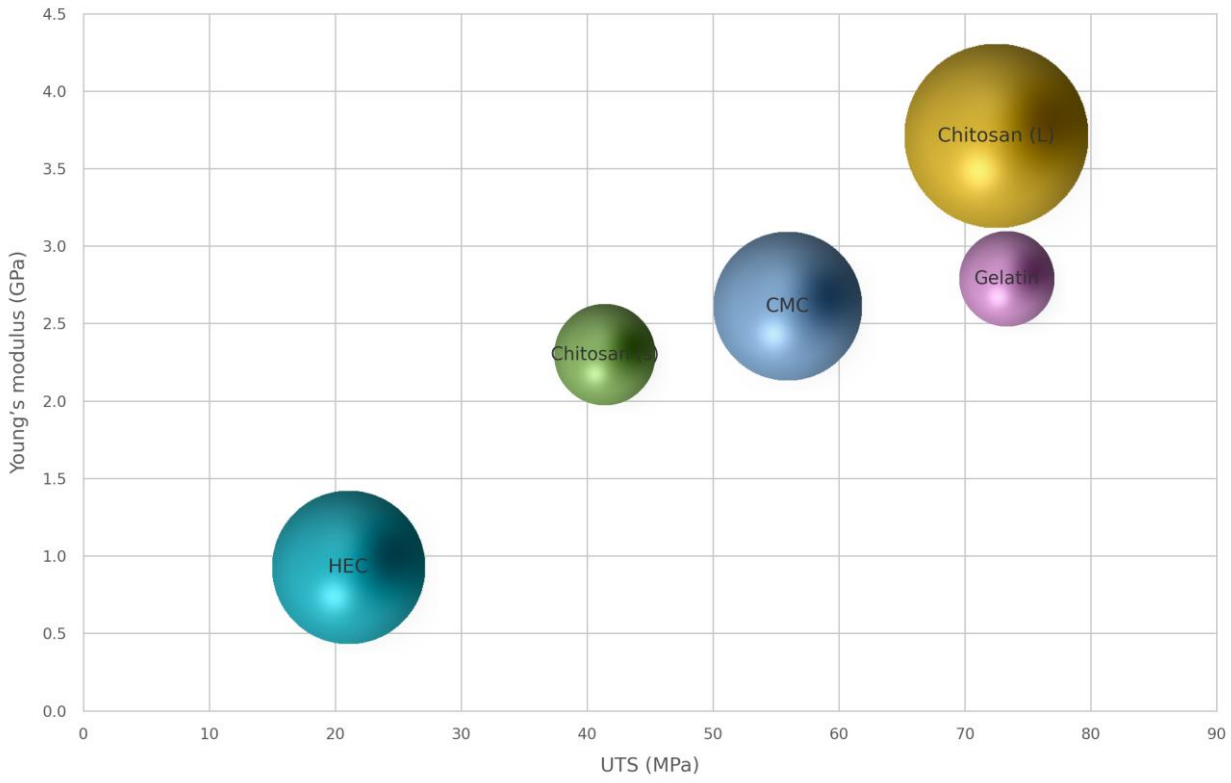


Figure 2. Tensile properties of neat polymers based on Young's modulus (Y-axis), ultimate tensile stress (X-axis), and strain at break (size of the marker)

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Among the selected neat polymers, chitosan (L) showed the highest Young's modulus, with a value of 3.7 ± 0.6 GPa, and was therefore the stiffest neat film. Gelatin and chitosan (L) showed very similar UTS values, 73.3 ± 8.9 MPa and 72.5 ± 10.8 MPa, respectively. Because the values are close and their error ranges overlap, both films are better described as the strongest neat films. CMC also showed relatively high strength, with a UTS of 55.9 ± 10.3 MPa.

Chitosan (L) also showed the highest mean strain at break among the selected neat polymers, 16.0 ± 8.3%. CMC and HEC showed intermediate strain values of 12.1 ± 5.5% and 11.3 ± 4.3%, respectively. Gelatin and chitosan (s) were the least extensible neat films, with strain at break values of 3.9 ± 1.0% and 4.3 ± 2.4%, respectively.

All neat films except HEC exceeded the 1.0 GPa stiffness target. HEC had a Young's modulus of 0.9 ± 0.1 GPa, placing it slightly below the target threshold. Overall, chitosan (L) showed the strongest neat-polymer profile in this selected group because it combined the highest stiffness, one of the highest UTS values, and the highest mean strain at break.

In addition to mechanical performance, qualitative visual observations were recorded for the neat polymer films. The CMC and HEC films were colourless and transparent, with slightly textured surfaces. Gelatin films were slightly yellowish but remained clearly transparent and had smooth, uniform surfaces. Chitosan (L) films appeared whitish, whereas chitosan (s) films were yellowish; both were slightly opaque. These observations provide an initial indication of the visual characteristics of the baseline films, although quantitative optical measurements will be required in later stages to assess transparency and haze more systematically.

4.2 Effect of blending of polymers

Polymer-polymer combinations were prepared to assess the effect of blending two film-forming polymers on stiffness, tensile strength, and extensibility. The tensile results for the polymer combinations without additives are summarised in Table 2.

Table 2. Tensile properties of polymer-polymer combinations

Composition	Ratio (% wt)	Young's modulus (GPa)	Ultimate tensile stress (MPa)	Strain at break (%)
CMC/Gelatin	75/25	2.74 ± 0.2	55.86 ± 2.5	4.54 ± 2.1
CMC/Gelatin	50/50	3.41 ± 0.6	63.81 ± 8.5	3.60 ± 1.0
Chitosan (s)/Gelatin	50/50	2.46 ± 0.2	57.53 ± 3.1	4.70 ± 0.7
HEC/Gelatin	50/50	1.99 ± 0.2	27.93 ± 5.5	2.04 ± 0.5
HEC/CMC	50/50	1.67 ± 0.1	33.80 ± 5.0	10.22 ± 3.5
HEC/chitosan (L)	50/50	2.72 ± 0.1	57.19 ± 8.3	8.48 ± 5.2

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The behaviour of the blends was broadly consistent with the mechanical profiles of the corresponding neat polymers. When gelatin was combined with CMC or low MW chitosan, the resulting films showed relatively high tensile stress, but low strain at break. This indicates that gelatin improved strength but reduced the flexibility of the film.

The CMC/gelatin series is shown in Figure 3 as a function of CMC content. The endpoints correspond to neat gelatin and neat CMC, while the intermediate points represent the polymer blends. Increasing the CMC fraction generally increased strain at break, whereas gelatin-rich compositions showed lower extensibility. This confirms that gelatin contributed to a stronger but more brittle response, while CMC helped retain a more effective mechanical profile.

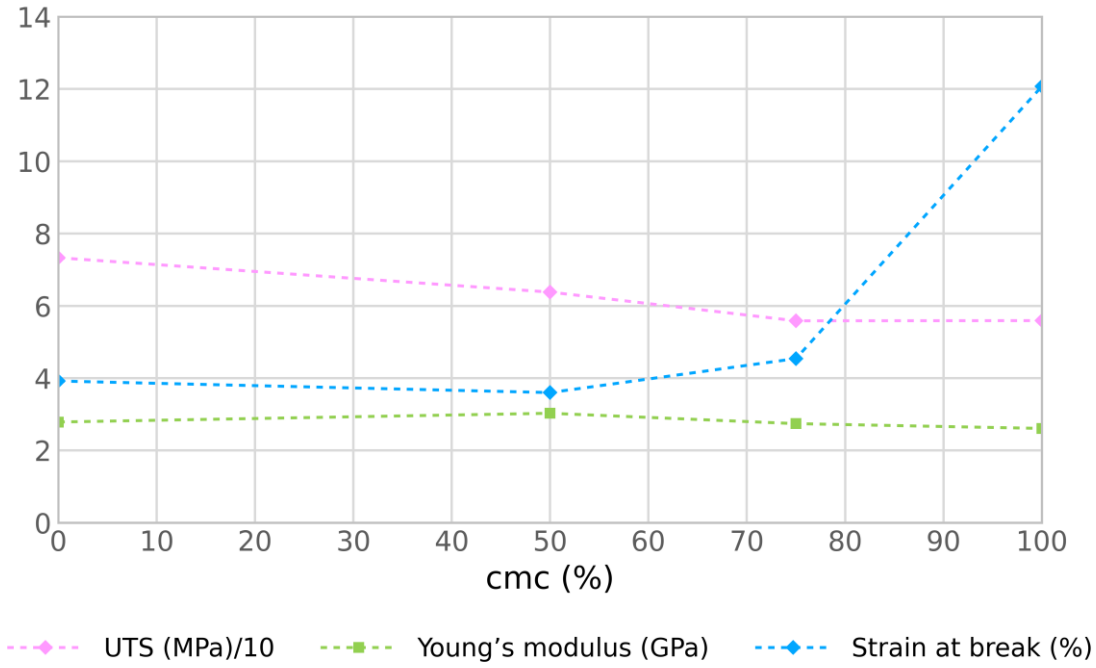


Figure 3. Effect of CMC content on the tensile properties of CMC/gelatin films. UTS values were divided by 10 to allow comparison on the same scale

The HEC-containing blends followed the behaviour expected from the neat components. Blending HEC with gelatin increased stiffness and strength compared with neat HEC but caused a substantial reduction in strain at break. In contrast, the HEC/CMC blend achieved higher stiffness and strength without the same loss of extensibility. Compared with neat HEC, the blend had higher stiffness and strength, together with relatively high strain at break.

It should also be noted that not all polymer combinations were compatible with the present solution-casting methodology. In particular, direct blending of chitosan with CMC was not feasible under the conditions used in this work. Chitosan behaves as a positively charged polymer in acidic media, whereas CMC carries negatively charged groups. When the two solutions are mixed, these opposite charges can interact strongly and lead to the formation of chitosan-CMC polyelectrolyte complexes rather than a homogeneous mixed solution. This behaviour is well documented in the literature. [26-28] In the present work, this incompatibility

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was observed experimentally as the formation of white, soft, gel-like domains after mixing the chitosan solution in acetic acid with the aqueous CMC solution.

4.3 Effect of additives

Additives were investigated as a way to modify the tensile properties of the films. This section discusses the effects of glycerol, lignin, PEG, sorbitol, urea, and ascorbic acid. Overall, the results show that additive performance depended strongly on the polymer matrix. This matrix-dependent behaviour is expected for hydrophilic biopolymer films.

The effect of an additive depends not only on the additive itself, but also on its compatibility with the polymer matrix, the strength of polymer-polymer interactions, hydrogen-bonding behaviour, water retention, chain mobility, and the microstructure formed during drying. Plasticisers generally reduce intermolecular forces and increase chain mobility, thereby reducing brittleness, but this can also reduce stiffness, tensile strength, cohesion, and barrier performance. In addition, water can itself act as a plasticiser in hydrophilic films, meaning that additives which alter moisture retention may indirectly affect tensile behaviour. At higher additive contents, compatibility limits may also be exceeded, leading to non-uniform films or weaker mechanical response. [29,30]

4.3.1 Effect of glycerol plasticisation

Gelatin followed the general plasticisation trend, but its response should be interpreted in relation to the neat gelatin baseline. Neat gelatin was strong but brittle, with high tensile stress and the lowest strain at break among the neat polymers. Glycerol addition reduced stiffness and strength and increased extensibility. At low glycerol content, the brittleness of gelatin was slightly reduced whereas at higher glycerol contents, however, the decrease in stiffness and strength became much more pronounced. This suggests that glycerol can reduce the brittleness of gelatin, but excessive plasticisation weakens the mechanical resistance of the film. The effect of glycerol content on gelatin and high MW chitosan films is summarised in Figure 4.

In chitosan-based films, glycerol caused a strong reduction in stiffness and strength, accompanied by a substantial increase in strain at break. This effect was particularly clear for high MW chitosan. The neat high MW chitosan film had the highest Young's modulus among the neat polymers and also showed high tensile stress. After glycerol addition, this strong and stiff profile was progressively replaced by much softer and more extensible behaviour. At higher glycerol contents, the material became highly deformable but lost much of its original mechanical resistance.

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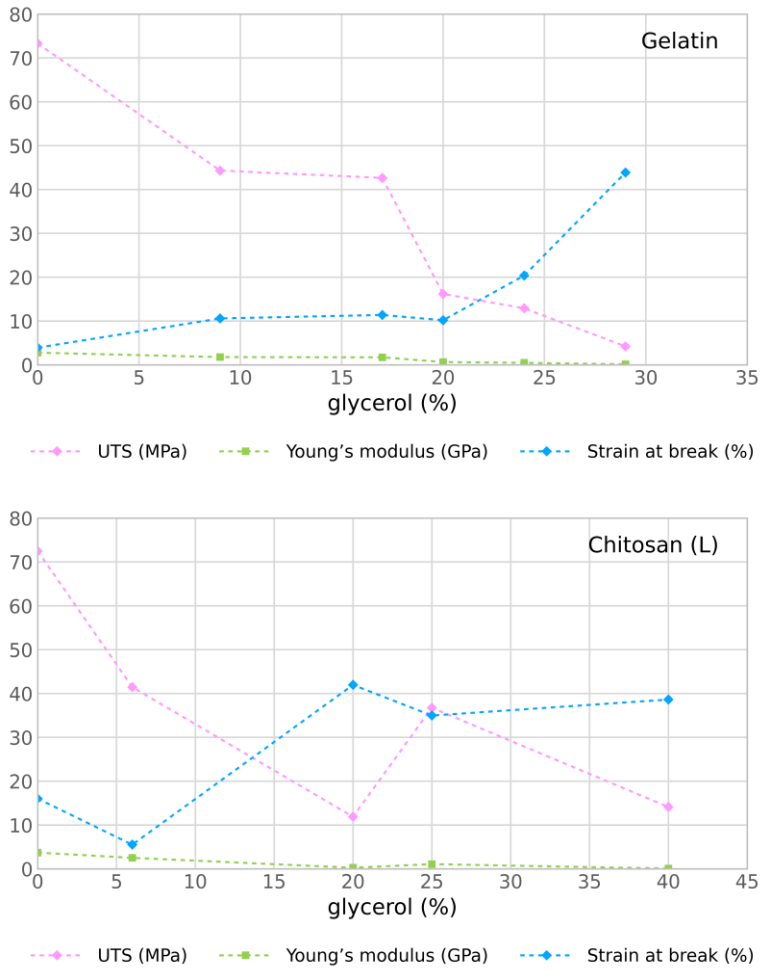


Figure 4. Effect of glycerol content on the mechanical properties of gelatin (top) and high MW chitosan (bottom) films

Similarly, compared with the neat low MW chitosan film, glycerol addition caused a clear reduction in stiffness and tensile stress, accompanied by a strong increase in strain at break. Young's modulus decreased from 2.3 ± 0.4 GPa to 0.2 ± 0.0 GPa, and UTS decreased from 41.4 ± 4.5 MPa to 13.7 ± 2.2 MPa. At the same time, strain at break increased from 4.3 ± 2.4% to 44.1 ± 8.8%. This confirms that glycerol had a consistent plasticising effect across both chitosan grades.

HEC was the most sensitive matrix to glycerol. Neat HEC already showed the lowest stiffness and tensile stress among the neat polymers, and glycerol addition caused further reduction in both properties. At the same time, strain at break increased markedly. This indicates that glycerol transformed HEC into a very soft and extensible material with limited resistance to loading. Therefore, although HEC retained some extensibility in the neat state, its already low stiffness made it particularly vulnerable to further softening by glycerol.

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4.3.2 Effect of lignin addition

Lignin was investigated as a biobased additive in selected systems. Lignin can act as a rigidifying component, filler, binder, or stress-transfer phase, which may increase stiffness or improve mechanical stability when it is sufficiently compatible and well dispersed. However, lignin can also disrupt hydrogen bonding within cellulose-derived networks or form lignin-rich domains when compatibility is poor, leading to lower extensibility or reduced mechanical performance. Therefore, the effect of lignin in cellulose-derived films should be interpreted as strongly dependent on formulation strategy, lignin type, and polymer-lignin compatibility. [31] Its effect was assessed both in non-plasticised formulations and, where applicable, in combination with glycerol. The lignin-containing systems discussed in this section are summarised in Table 3.

4.3.2.1 Gelatin/lignin systems

Gelatin/lignin films without glycerol were prepared several times, but they could not be mechanically tested because they were consistently too fragile to remove from the mould, cut into specimens, and measure. This qualitative observation suggests that lignin had a strong rigidifying or embrittling effect in gelatin when no plasticiser was present.

All measurable gelatin/lignin films therefore contained glycerol. Compared with neat gelatin, the gelatin/lignin/glycerol formulations showed lower stiffness and tensile strength, but higher strain at break, indicating a shift away from the strong but brittle behaviour of neat gelatin towards a more deformable response.

Direct comparisons at fixed gelatin content further support this interpretation. At both 83% and 71% gelatin, replacing part of the glycerol fraction with lignin increased Young's modulus and UTS, while reducing strain at break. Lignin therefore appeared to partly offset the softening effect of glycerol and helped retain stiffness and tensile strength, although this occurred at the expense of extensibility.

The relative balance between glycerol and lignin was also important. Formulations containing similar proportions of glycerol and lignin retained higher stiffness and tensile strength, but showed lower strain at break. In contrast, formulations with higher glycerol than lignin showed lower stiffness and strength, but greater extensibility. This suggests that, in gelatin-based systems, glycerol primarily increased deformability, while lignin helped retain load-bearing capacity but reduced elongation at break.

4.3.2.2 CMC/lignin systems

CMC was tested with lignin alone and with lignin plus glycerol, permitting the comparison between lignin as a single additive and lignin in a plasticised CMC film.

Compared with neat CMC, the addition of 17% lignin only slightly increased Young's modulus, while UTS remained similar. However, strain at break decreased from 12.07 ± 5.52% to 6.73 ± 1.31%, indicating that lignin acted mainly as a rigidifying component in the CMC matrix, increasing stiffness while reducing extensibility.

The comparison between CMC/glycerol and CMC/glycerol/lignin is also important. Young's modulus increased from 1.09 ± 0.07 GPa to 1.23 ± 0.10 GPa, UTS increased from 23.20 ± 3.21 MPa to 32.62 ± 3.33

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MPa, and strain at break increased from 13.45 ± 3.48% to 21.17 ± 5.07%. This is notable because the CMC content was lower in the lignin-containing formulation, decreasing from 80% to 67%. Despite this lower polymer fraction, the lignin-containing film showed higher stiffness and strength, suggesting that lignin helped retain or increase the load-bearing response of the plasticised CMC film. The higher strain at break also indicates that the presence of glycerol changed the way lignin affected the CMC system.

Overall, the CMC results show two different lignin effects depending on whether glycerol is present. Without glycerol, lignin slightly increased stiffness and reduced strain at break, while UTS remained close to that of neat CMC. With glycerol, the lignin-containing film showed higher stiffness, UTS, and strain at break than CMC/glycerol.

4.3.2.3 HEC/lignin systems

HEC was tested with lignin at a HEC/lignin ratio of 91/9 by weight and compared with neat HEC. Lignin increased the Young's modulus, raising the film stiffness above the 1 GPa target, but it also caused a substantial reduction in strain at break. The effect on UTS was less clear, as the mean value increased only slightly and the variability of the HEC/lignin result was high.

Overall, the HEC/lignin system followed the same general trend observed for CMC/lignin. In both cellulose-derived matrices, lignin mainly acted as a rigidifying component: it increased stiffness, had limited effect on tensile strength, and reduced extensibility.

Table 3. Effect of lignin addition

Composition	Ratio (% wt)	Young's modulus (GPa)	Ultimate tensile stress (MPa)	Strain at break (%)
Gelatin/Lignin/Glycerol	83/8/8	1.89 ± 0.1	46.37 ± 4.2	5.29 ± 1.2
Gelatin/Lignin/Glycerol	67/17/17	1.57 ± 0.2	39.60 ± 2.6	8.05 ± 3.0
Gelatin/Lignin/Glycerol	71/11/18	0.61 ± 0.04	16.95 ± 1.72	23.81 ± 5.3
Gelatin/Lignin/Glycerol	62/15/23	0.46 ± 0.03	13.34 ± 1.6	28.43 ± 8.2
CMC/Lignin	83/17	2.91 ± 0.3	54.37 ± 3.9	6.73 ± 1.3
CMC/Glycerol/Lignin	67/17/17	1.23 ± 0.1	32.62 ± 3.3	21.17 ± 5.07
HEC/Lignin	91/9	1.56 ± 0.1	24.34 ± 8.4	3.51 ± 3.3
Chitosan (L)/Lignin, post-treated	Post-treatment	5.74 ± 0.1	107.16 ± 15.7	2.69 ± 1.0

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4.3.2.4 Chitosan/lignin systems

Chitosan/lignin films could not be prepared by the simple solution-casting methodology used for the other systems. Alkali lignin is not soluble under acidic conditions, whereas chitosan requires acidic conditions for dissolution and is not compatible with strongly basic conditions. In addition, alkali lignin makes aqueous solutions strongly basic. As a result, direct incorporation of alkali lignin into chitosan-based casting solutions was not feasible using the same preparation route.

A modified post-treatment route was therefore explored for chitosan (L) to overcome this problem. First, chitosan (L) films were prepared and fully dried. The dried films were then immersed for 3 h in a 3% aqueous lignin solution. [2] During this period, the films were turned over several times to promote more uniform exposure. Because the lignin solution was highly basic and the chitosan had already formed a dry film, the film did not dissolve during the treatment. After 3 h, the films were immersed in water to remove excess lignin and then dried under the usual conditions. The resulting films were brown rather than yellowish but remained transparent rather than opaque.

The cross-linked chitosan (L)/lignin film gave the highest stiffness and strength of all samples discussed in this chapter, with a Young's modulus of 5.7 ± 0.1 GPa and a UTS of 107.2 ± 15.7 MPa. Compared with neat chitosan (L), this corresponds to increases of approximately 54.7% in Young's modulus and 47.8% in UTS. However, strain at break decreased from 16.0 ± 8.3% for neat chitosan (L) to 2.7 ± 1.0%. This indicates a strong increase in load-bearing capacity, but a major loss of extensibility.

Overall, lignin had a strongly polymer-dependent effect. In HEC, it increased stiffness but reduced strain at break. In CMC/glycerol, it improved the overall tensile balance compared with the corresponding glycerol-containing film. In gelatin, lignin-containing films required glycerol to become testable, and the mechanical response depended strongly on the balance between lignin-associated brittleness or rigidification and glycerol-induced plasticisation. In chitosan (L), direct solution casting with lignin was not feasible, but post-treatment in lignin solution produced a cross-linked film with very high stiffness and strength, accompanied by substantially reduced elongation.

4.3.3 Effect of other additives

Following the assessment of glycerol and lignin, other additives were examined to determine whether alternative modifiers could provide improved stiffness, strength and extensibility. This group includes:

- Polyethylene glycol (PEG) can be described as a polyol plasticiser or additive used in polymer and biopolymer film systems. In natural polymer-based films, including cellulose and chitosan systems, PEG has been used to modify film properties, particularly flexibility and mechanical performance. PEG is also commonly described as low-toxicity and biocompatible, with reports describing it as non-toxic, biocompatible, non-immunogenic and non-antigenic. Its plasticising effect is mainly associated with reduced polymer–polymer interactions, increased free volume and improved polymer-chain mobility, which can increase elongation at break and reduce film inflexibility or brittleness. [30,32-33]
- Sorbitol is a hydroxyl-rich polyol plasticiser used in edible and biodegradable biopolymer films. It is added to reduce brittleness and improve flexibility by weakening intermolecular interactions and increasing polymer-chain mobility. Its effects depend on the polymer matrix and concentration: in

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sugar palm starch films, sorbitol improved handling and flexibility, but was less effective than glycerol at equivalent levels. It can also affect moisture content, solubility, water absorption and water-vapour permeability. [34-36]

- Urea is a small molecule used in starch functionalisation and hydrophilic polymer film systems. In film-related studies, it has been reported as a plasticiser or structure-modifying additive, particularly in starch-based materials and often in combination with glycerol. Its effects are mainly associated with hydrogen bonding with polymer chains, improved plasticisation, reduced starch retrogradation or recrystallisation, and changes in thermal behaviour. Mechanical effects are strongly formulation-dependent. [37-38]
- Ascorbic acid, also known as vitamin C, is a naturally derived, food-compatible bioactive compound used in active edible and biodegradable food-packaging films. Unlike conventional plasticisers, it is mainly incorporated for preservation-related functions, particularly antioxidant activity, oxygen scavenging and inhibition of lipid oxidation or browning. In some film systems, vitamin C also contributes to antimicrobial activity and broader shelf-life extension. Its incorporation can modify film properties, including permeability, optical characteristics, thermal and mechanical performance, with the final effect depending on the polymer matrix, concentration and fabrication method. [35,39]

4.3.3.1 Effect of PEG, sorbitol and urea

In gelatin, PEG reduced Young's modulus from 2.8 ± 0.3 to 1.6 ± 0.1 GPa and UTS from 73.3 ± 8.9 to 37.7 ± 1.3 MPa. Strain at break increased from 3.9 ± 1.0% to 7.4 ± 0.7%. This indicates a mild plasticising effect, with reduced brittleness but lower stiffness and strength. In CMC, PEG also reduced Young's modulus and UTS, together with the mean strain at break. Overall, the plasticising effect of PEG was clearer in gelatin.

Sorbitol showed different effects in gelatin and CMC. In gelatin, sorbitol reduced Young's modulus and UTS but increased strain at break from 3.9 ± 1.0% to 12.8 ± 3.4%, indicating a plasticising effect. Compared with glycerol at similar loading, sorbitol retained more stiffness and strength but gave lower elongation.

In CMC, sorbitol did not improve strain at break. The CMC/sorbitol film had a Young's modulus of 2.9 ± 0.1 GPa and a UTS of 48.7 ± 14.2 MPa, but strain at break was only 2.3 ± 0.7%. This was much lower than neat CMC and indicates a more brittle response.

The gelatin/glycerol/sorbitol film showed 20.5 ± 5.7% strain at break, together with low Young's modulus and UTS. Its tensile response was broadly similar to gelatin/glycerol at comparable glycerol content, suggesting that glycerol strongly influenced the measured behaviour in this ternary system.

Urea also behaved differently in CMC and gelatin. In CMC, urea reduced UTS and strongly reduced strain at break, from 12.1 ± 5.5% for neat CMC to 3.6 ± 1.4%. In gelatin, urea increased strain at break compared with neat gelatin, reaching 12.2 ± 4.5%, but this was accompanied by lower stiffness and UTS.

Overall, sorbitol and urea showed a clearer plasticising effect in gelatin than in CMC. In gelatin, both additives improved extensibility but reduced stiffness and strength. In CMC, both additives reduced strain at break, indicating that they did not improve the overall tensile performance of the CMC films under the conditions tested. The comparative effect of the different additives on CMC-based films at an 80/20 polymer/additive ratio is summarised in Figure 5.

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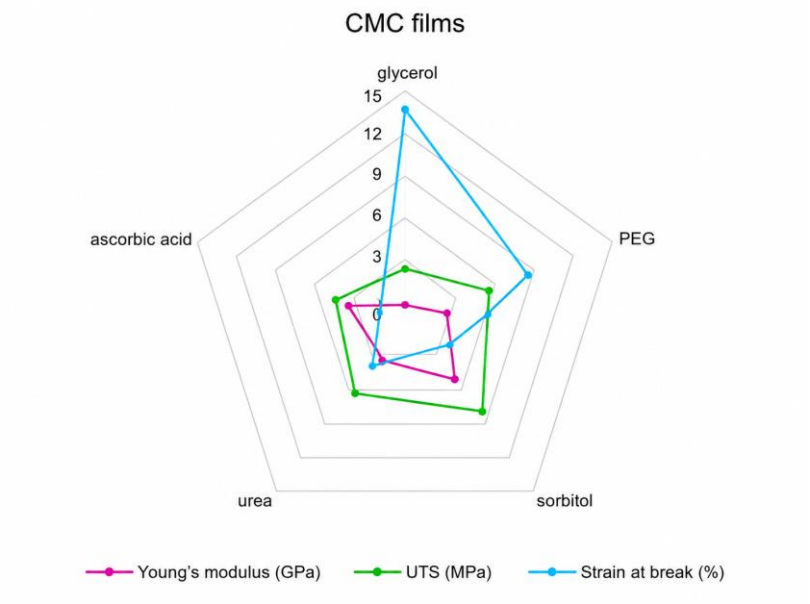


Figure 5. Spider plot comparing Young's modulus, ultimate tensile stress and strain at break of CMC/additive films at an 80/20 ratio. Ultimate tensile stress values were divided by 10 for visual clarity

4.3.3.2 Effect of ascorbic acid

In CMC, ascorbic acid produced a stiff but brittle film. The CMC/ascorbic acid film was very brittle and difficult to test, so only two valid specimens were obtained. The indicative results showed a high Young's modulus of 3.7 ± 0.4 GPa, but very low strain at break, 1.3 ± 0.2%. Compared with neat CMC, this suggests that ascorbic acid increased stiffness but strongly reduced extensibility. However, because of the low number of valid tests, this result should be interpreted cautiously.

The CMC/glycerol/ascorbic acid film showed lower stiffness and UTS than CMC/ascorbic acid, with a Young's modulus of 1.4 ± 0.2 GPa and a UTS of 21.3 ± 7.8 MPa. However, strain at break remained very low, at 1.8 ± 0.6%. Compared with CMC/glycerol, the addition of ascorbic acid was therefore associated with a major reduction in extensibility.

Gelatin/ascorbic acid and chitosan/ascorbic acid films without glycerol or another plasticiser were also prepared, but they could not be mechanically tested because the films broke immediately during handling. The gelatin/glycerol/ascorbic acid film showed a strain at break of 27.0 ± 3.6%, with a Young's modulus of 0.5 ± 0.0 GPa and a UTS of 16.6 ± 1.4 MPa. This sample was more extensible than neat gelatin, but it was below the 1.0 GPa stiffness target and had much lower tensile strength.

In the chitosan (s)/ascorbic acid systems, ascorbic acid was used directly as the acidic component for dissolving chitosan in water. Therefore, no acetic acid was added to these formulations. This distinguishes these samples from the other chitosan-based films, which were prepared from chitosan dissolved in aqueous acetic acid.

The chitosan (s)/ascorbic acid/glycerol 42:42:17 film had low stiffness and strength, with a Young's modulus of 0.2 ± 0.0 GPa and a UTS of 7.4 ± 1.9 MPa, while strain at break was 12.9 ± 5.7%. Increasing the combined additive content further, as in the 33:33:33 chitosan (s)/glycerol/ascorbic acid film, produced a highly

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extensible but mechanically very weak material. This film showed a strain at break of 86.4 ± 11.1%, but its Young's modulus and UTS were only 0.0 ± 0.0 GPa and 1.0 ± 0.3 MPa, respectively.

Overall, ascorbic acid had a strongly matrix-dependent effect. In CMC, it was associated with increased stiffness but severe embrittlement. In gelatin and chitosan, films containing ascorbic acid without plasticiser were too brittle to test. When glycerol was added, testable films could be obtained, but the resulting materials generally showed low stiffness and strength. In chitosan, ascorbic acid was useful as an alternative acidic component for dissolution, but the resulting plasticised films were not among the strongest systems investigated.

4.4 Comparative assessment of the tested film systems

The results show that the tensile behaviour of the films was governed by both polymer identity and composition. The neat polymers established distinct mechanical baselines, and the behaviour of the blends and additive-containing films generally reflected the characteristics of their main components. Gelatin-containing systems, for example, tended to show high strength but low extensibility, while CMC-based systems generally offered a good combination of stiffness, strength, and strain at break. HEC exhibited low stiffness and high sensitivity to plasticisation.

The effect of additives was strongly matrix-dependent. The same additive did not necessarily produce the same response in different polymers. Sorbitol and urea improved extensibility in gelatin but reduced strain at break in CMC, while glycerol softened all matrices but had a particularly strong effect in HEC and chitosan. Lignin improved the tensile balance of plasticised CMC and strongly increased stiffness and strength in post-treated chitosan (L), but reduced extensibility and showed more limited applicability in gelatin. These results confirm that additive performance is governed not only by additive type, but also by compatibility with the polymer matrix, water retention, chain mobility, and the microstructure formed during drying.

A general trade-off was observed across the dataset. Films with higher Young’s modulus and UTS often showed lower strain at break, whereas highly extensible films generally had reduced stiffness and strength. No formulation simultaneously maximised all three mechanical properties. Therefore, future optimisation should focus on matrix-specific formulation strategies that balance stiffness, tensile strength, and extensibility according to the intended application.

From the systems tested so far, CMC-based films appear to offer an interesting and practical starting point for further development. Chitosan (L) also remains of strong interest because of its high baseline stiffness and strength, although its response to plasticisation and post-treatment requires further control. Gelatin provides high tensile strength but requires modification to reduce brittleness, while HEC appears less suitable where the 1.0 GPa stiffness target is critical.

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5 Progress on the Autonomous Platform

In parallel with the experimental development of the thin-film compositions, significant effort has been devoted to the design of an autonomous platform intended to support and accelerate the laboratory workflow. The initial concept behind the platform is that of a laboratory assistant capable of performing multiple assays, thereby increasing efficiency, repeatability and reproducibility. The longer-term objective is to combine standardised preparation and basic testing on the same platform, generate structured datasets from each run, and subsequently use these data to support the identification and optimisation of new film compositions according to the target criteria defined by the end-user.

At the present stage, the autonomous platform should be considered a work in progress. Its design has been completed, a procurement list has been established, and the required accessories have been ordered and delivered. The physical development of the system has already begun; however, this remains at an early stage and therefore the current report focuses primarily on the design logic, the functional modules, and the expected role of the platform within the overall task.

5.1 Platform architecture and coordinated workflow

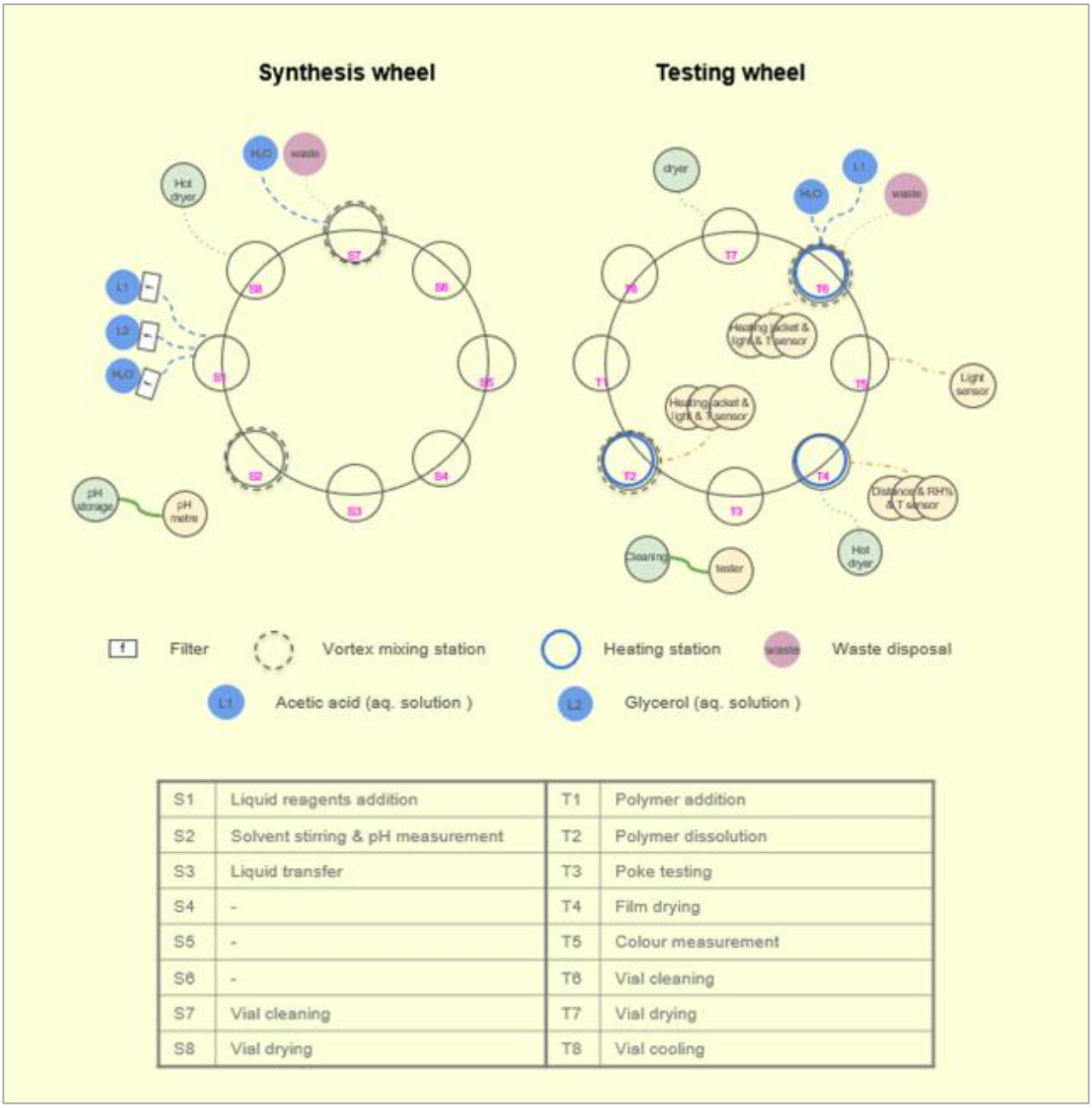
The platform architecture is based on two coordinated Geneva wheels[1], each divided into a series of fixed stations with a dedicated role. The process is organised into two interconnected modules: (a) a synthesis wheel, in which the casting solvent is prepared and conditioned, and (b) a testing wheel, in which polymer dissolution, film formation, and basic testing or quality-control steps take place. In this arrangement, the vials remain stable during each operation, while the Geneva-wheel motion moves them stepwise from one station to the next. This allows the workflow to be organised as a repeatable sequence of discrete operations, including reagent addition, mixing, pH measurement, transfer, drying, testing, cleaning, and reuse. Whenever transfer between the two wheels is required, the prepared liquid is moved by syringe from the synthesis wheel to the testing wheel. Overall, this architecture is intended to minimise unnecessary handling, improve repeatability, and standardise the sequence of operations between runs.

Each wheel is designed to hold eight removable stainless-steel vials positioned within plastic crowns. The vials are supported by bearings and magnets, keeping them securely positioned and magnetically coupled to motors located beneath the wheels. This arrangement enables local vial spinning at selected stations without direct mechanical contact and supports mixing, dissolution, cleaning, and other operations that require controlled agitation. The proposed two-wheel workflow, including the synthesis and testing stations, is shown schematically in Figure 6.

[1] A Geneva wheel is an indexing mechanism that converts continuous rotary motion into intermittent, stepwise rotation. In this mechanism, a rotating drive wheel uses a pin to engage with slots in a driven wheel, advancing it by one fixed step at a time. Between steps, the driven wheel remains locked in position, allowing operations to be carried out at stable, repeatable stations.

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S1	Liquid reagents addition	T1	Polymer addition
S2	Solvent stirring & pH measurement	T2	Polymer dissolution
S3	Liquid transfer	T3	Poke testing
S4	-	T4	Film drying
S5	-	T5	Colour measurement
S6	-	T6	Vial cleaning
S7	Vial cleaning	T7	Vial drying
S8	Vial drying	T8	Vial cooling

Figure 6. Schematic representation of the proposed autonomous platform workflow, showing the two coordinated Geneva wheels and the sequence of dedicated stations used for solvent preparation, polymer processing, film formation, basic testing, cleaning, and reuse

Even though the autonomous platform is being designed as a flexible tool for the preparation and preliminary assessment of different biobased film compositions, the first workflow has been modelled on chitosan/glycerol film preparation. This system was selected as a representative starting case because it is more demanding than the simple dissolution of water-soluble polymers such as CMC or gelatin in water. Chitosan processing requires an acidic environment for dissolution, generally involves longer dissolution

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times, and is strongly affected by the specific chitosan grade used. Different chitosan grades can vary in molecular weight, degree of deacetylation, viscosity, and dissolution behaviour, and these differences can affect both solution preparation and final film properties.

For this reason, chitosan/glycerol provides a useful initial model system for designing a platform able to handle more complex preparation conditions and remain adaptable to simpler polymer systems in later stages. The individual stations of the two wheels therefore reflect the main operations required for this model workflow and provide a modular basis that can later be adjusted to other film-forming systems.

5.1.1 Synthesis wheel

The synthesis wheel is responsible for preparing and conditioning the solvent system before polymer addition. At station S1, liquid reagents are added in measured quantities in order to prepare the casting solvent. Depending on the target formulation, this may include aqueous acetic acid solution, glycerol, and water, up to a maximum total volume of 20 mL. Liquid addition is carried out through a set of needles connected by hoses to syringe pumps, allowing controlled dosing of the liquid components. This step establishes the chemical environment that will later enable chitosan dissolution and influence the properties of the final film, including flexibility and visual appearance.

At station S2, the prepared solvent is homogenised and its pH is measured. pH control is important because it directly affects chitosan dissolution behaviour and helps ensure that batch-to-batch conditions remain comparable. The pH probe is moved along the Y and Z axes, allowing it to be inserted into the vial for measurement and then returned to its storage position. The probe is stored in storage solution and is cleaned with water before and after each measurement. During this stage, vial spinning can be provided by motor-rotating magnets beneath the wheel, supporting homogenisation during or around the pH check.

At station S3, the prepared solvent is positioned for transfer and is subsequently moved to station T1 of the testing wheel, where polymer addition takes place. This transfer is performed using an XYZ moving gantry equipped with a transfer syringe mechanism. The same gantry also carries a powder dispenser, allowing the platform to coordinate liquid transfer and subsequent polymer addition within the automated sequence.

Stations S4 to S6 are currently reserved positions within the synthesis wheel layout. Their inclusion provides flexibility for future additions or modifications to the automated process, should further operations be required as the platform develops.

Station S7 is dedicated to vial cleaning. This station uses a Z-axis moving needle pack that supplies water and removes liquid waste by suction. Vial spinning by motor-rotating magnets is used to assist cleaning by promoting agitation inside the vial. Finally, station S8 provides air-drying of the cleaned vial so that it can be returned to service under reproducible conditions. The drying station uses active heated airflow, with temperature and fan speed monitored during operation.

5.1.2 Testing wheel

The testing wheel carries out polymer addition, dissolution, film formation and basic quality checks. At station T1, precisely weighed chitosan is introduced into the prepared acidic solvent. Controlled addition is preferred over immediate bulk addition, as this helps prevent clumping and supports more uniform dissolution. Polymer addition is performed using the powder dispenser mounted on the XYZ moving gantry, allowing the

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solid component to be introduced into the transferred solvent in a controlled manner. A CAD representation of the proposed platform and the arrangement of its main structural and functional components is presented in Figure 7.

At station T2, the mixture is processed until the polymer has dissolved. This powder-mixing and dissolution station uses a Z-axis moving cap equipped with an infrared temperature sensor for monitoring the solution, an optical sensor for checking dissolution, and induction heating for the vial. Vial spinning is again achieved by motor-rotating magnets beneath the wheel, providing agitation during dissolution. Mild heating is included to accelerate polymer dissolution and improve solution homogeneity, with the aim of obtaining a visually uniform casting solution without undissolved particles.

Station T3 is dedicated to a preliminary mechanical check of the forming material. The station uses a Z-axis moving end-tip mounted on a load cell, allowing a simple poke-type measurement to be performed under controlled conditions. This is not intended to replace full mechanical characterisation, but to provide a rapid practical indication of consistency or early film-forming behaviour within the automated workflow. The station also includes a retractable cleaning mechanism with water supply and waste suction.

At station T4, the polymer solution is dried to form a film. The drying station is based on a Z-axis moving cap with active heated airflow input and output. Temperature, humidity, and fan speed are monitored, and the system also includes an infrared temperature sensor, a time-of-flight level sensor, and induction heating for the vial to support controlled drying. These parameters affect drying rate and film quality. Their control is expected to reduce common defects such as wrinkling, cracking, trapped bubbles, and uneven thickness.

Once the film has formed, station T5 performs a colour or optical check. This station uses a Z-axis moving cap equipped with an optical sensor for colour measurement. This step provides a simple quantitative readout that may help identify variability between batches, such as yellowing, haze, or changes in transparency, which may in turn reflect composition differences or altered drying conditions.

The testing wheel also incorporates cleaning and reuse functions. Station T6 is a vial-cleaning station connected to water input and waste output. In addition, this station includes acid supply, waste suction, induction heating for the vial, and vial spinning by motor-rotating magnets. This combination is intended to improve cleaning effectiveness, particularly after polymer dissolution and film formation steps. Station T7 provides heated drying of vials after cleaning, and station T8 cools the vials to a stable handling temperature before reuse.

A notable feature of the design is the explicit integration of cleaning, drying, and reuse steps within both wheels. This has been included in order to minimise cross-contamination between experiments and to allow repeated operation without excessive manual intervention. By embedding these steps directly into the workflow, the platform is intended (a) to automate the preparation of films and (b) to standardise the conditions under which sequential runs are performed. This is particularly important for a system that aims to generate reliable comparative data for later optimisation.

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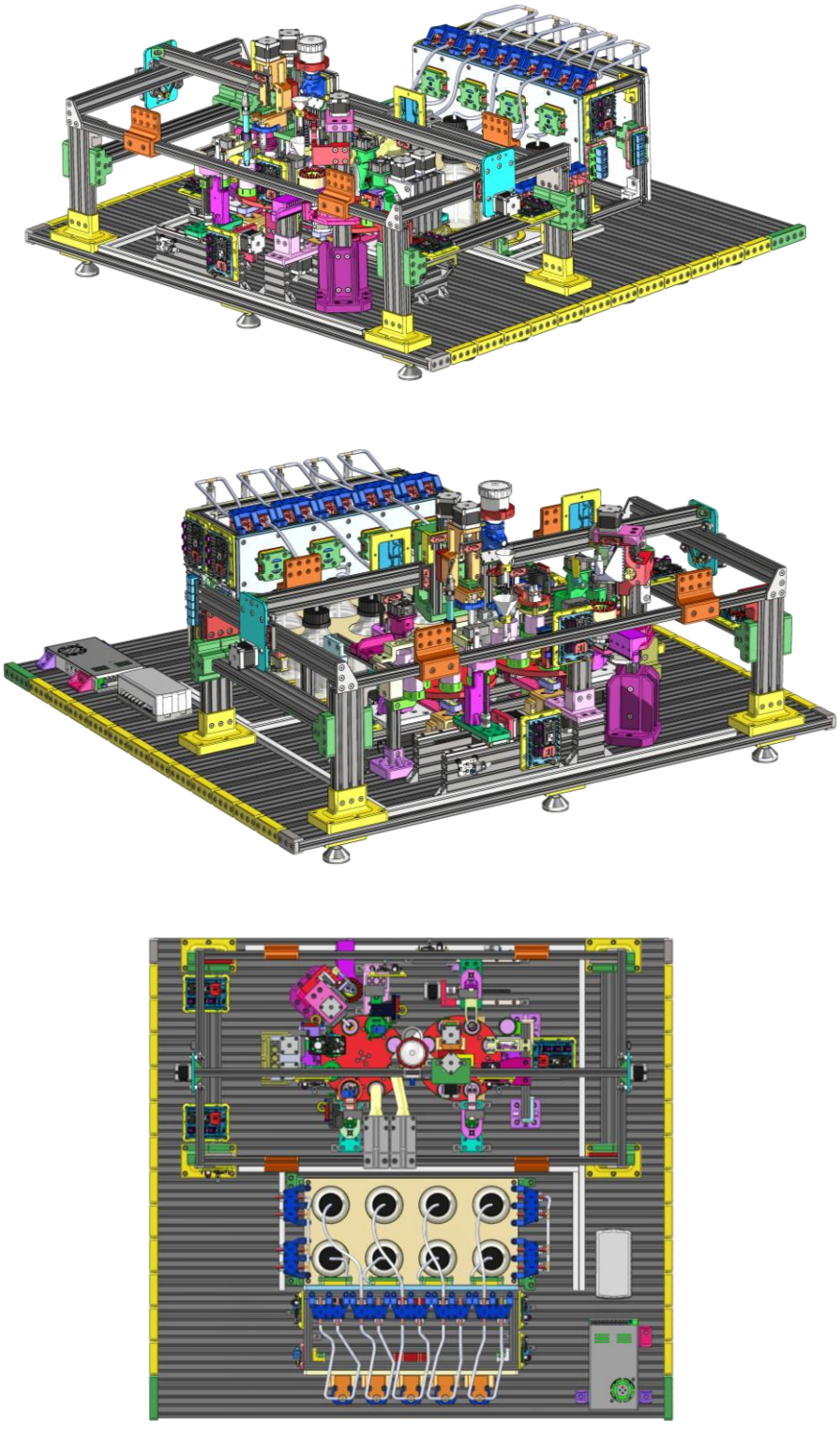


Figure 7. CAD representation of the proposed autonomous platform, showing the arrangement of the main structural and functional components from three viewing angles

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5.2 Data generation, current status, and next steps

The platform has been designed not merely as a preparation device but as a data-generating tool. Samples will be prepared and subjected to simple in-line checks on the same platform, creating the basis for a structured dataset that links composition, processing conditions, and observed response. In a later stage, the intention is to extract these results and use them to feed a computational system capable of suggesting new compositions that may better satisfy the desired property targets. In this sense, the present platform constitutes the experimental backbone upon which future data-driven optimisation can be built.

At the time of writing, the design phase has been completed and the corresponding procurement list has been prepared. The required accessories have been ordered and delivered, and the development of the platform has already started. The next reporting period is expected to include visual documentation of the assembled system, a fuller description of the individual modules, and initial observations from the first stages of implementation and integration.

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6 Conclusions

6.1 Conclusions

The first reporting period of Task 5.4 established the practical and methodological basis for the development of biobased thin films for packaging applications within the THESEUS project. During this period, NCSR defined a stepwise development strategy, aligned the work with the requirements provided by TSIMIS, prepared the necessary laboratory infrastructure, selected candidate biobased polymers and additives, and established the main preparation and mechanical-testing workflows.

A sustainability-oriented approach was adopted from the beginning of the task. Water or aqueous systems were used, harsh chemicals and unnecessarily energy-intensive steps were avoided, and priority was given to materials that are biobased, biodegradable, abundant, relatively affordable, and compatible with mild processing conditions. This approach is consistent with the broader circularity and green-chemistry objectives of the project.

The first experimental results showed that the selected polymers provide distinct mechanical baselines. High MW chitosan gave the highest stiffness among the neat polymers and also showed high tensile resistance. Gelatin showed high tensile stress but low elongation at break. CMC provided a favourable combination of stiffness, tensile stress, and elongation at break. HEC showed lower stiffness and greater sensitivity to plasticisation. These differences confirmed that polymer identity strongly controls the mechanical behaviour of the films.

The study of blends and additive-containing films further showed that the final mechanical response depends strongly on the polymer matrix and the composition. Glycerol produced the expected plasticising effect across all polymer families, reducing stiffness and tensile stress and increasing elongation at break. Lignin showed a more composition-dependent effect, with promising behaviour in selected CMC-based systems. Chitosan/lignin films were not feasible using the current preparation route because acidic chitosan processing is incompatible with the basic conditions associated with alkali lignin. Other additives, including PEG, sorbitol, urea, and ascorbic acid, also produced matrix-dependent effects, confirming that no single additive can be treated as universally beneficial across all systems. These findings show that future work should focus on matrix-specific formulation strategies rather than universal additive selection, since the mechanical response depends strongly on polymer-additive compatibility, water retention, film microstructure, and the balance between stiffness, strength, and extensibility.

Overall, the results demonstrate that film optimisation requires a careful balance between stiffness, tensile resistance, and extensibility. No material system simultaneously maximised all three mechanical properties, and therefore the selection of future compositions should be guided by the target performance requirements rather than by a single property alone. At the present stage, CMC-based systems appear to offer a useful starting point for further optimisation because they retained comparatively good mechanical performance across several compositions. High MW chitosan also remains of strong interest because of its high baseline stiffness and tensile stress, although its response to plasticisation and processing conditions requires further control.

In parallel with the material-development work, significant progress was made on the design of the autonomous platform. The proposed system is based on two coordinated Geneva wheels, organised into

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synthesis and testing modules, with dedicated stations for solvent preparation, pH measurement, transfer, polymer addition, dissolution, film drying, preliminary testing, optical checking, cleaning, and reuse. Although the platform is designed as a flexible tool for different biobased film compositions, the initial workflow has been modelled on chitosan/glycerol films because this system presents a more demanding processing case than simple dissolution of water-soluble polymers. At the time of writing, the platform remains under development, and no validated automated dataset has yet been generated. However, the design provides a clear basis for future automated preparation, quality checking, and structured data generation.

6.2 Next Steps

The next stage of the work will focus on expanding and refining the film-formulation and testing activities. Film preparation and mechanical testing will continue in order to ensure that sufficient information is available to guide the selection of the most promising development pathways. Candidate compositions will be prioritised based on the target requirements agreed with TSIMIS, including stiffness, processability, handling quality, and visual appearance. Promising systems will be repeated where needed to confirm reproducibility before being taken forward for broader characterisation. In addition, processing parameters such as concentration, casting volume, drying conditions, film thickness, and additive loading may be further adjusted. Based on the results obtained so far, additional materials, new polymer-additive combinations, and potentially more complex multi-component systems may also be introduced where necessary.

A major priority for the next reporting period will be the incorporation of material fractions recovered from the Blue Bins. The relevant material stream to be received and incorporated has already been discussed and agreed; however, experimental testing with this material has not yet started. This will now become a central focus of the laboratory work. The initial screening of commercial model polymers and additives has provided a clearer understanding of the behaviour of the material systems under investigation, and this knowledge will support the transition towards Blue Bin-derived fractions. Commercial model systems will also remain useful as reference materials for comparing the behaviour of waste-derived fractions against controlled baselines.

If promising compositions are identified, the characterisation work will be expanded beyond mechanical testing. Additional properties relevant to the intended packaging application, such as water vapour permeability, wettability, optical behaviour, thermal response, solvent resistance, and surface properties, will be investigated. These measurements will provide important information for assessing whether the selected films can meet the broader application requirements, rather than relying on mechanical performance alone.

In parallel, the manufacturing of the autonomous platform will continue. The fabrication process has already started but remains at an early stage. The next steps will follow the completed design, with the aim of assembling and integrating the different modules of the system. The experimental data generated during continued laboratory screening will also be organised in a consistent format so that they can later support structured data analysis and data-driven optimisation.

Once manufacturing and assembly are complete, initial operational testing of the platform will begin. Films prepared and tested using the automated system will be compared with those produced by the laboratory team under corresponding conditions. This comparison will be necessary to confirm that the automated platform can reproduce the laboratory results reliably and can therefore be used as a dependable tool for

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future structured data generation and optimisation. Any differences between the automated and manual results will be used to refine the platform workflow, operating parameters, and testing protocol.

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References

1. Cui, L., Gao, S., Song, X., Huang, L., Dong, H., Liu, J., Chen, F. and Yu, S., 2018. Preparation and characterization of chitosan membranes. RSC advances, 8(50), pp.28433-28439.
2. Ravishankar, K., Venkatesan, M., Desingh, R.P., Mahalingam, A., Sadhasivam, B., Subramaniyam, R. and Dhamodharan, R., 2019. Biocompatible hydrogels of chitosan-alkali lignin for potential wound healing applications. Materials Science and Engineering: C, 102, pp.447-457.
3. Cui, C., Gao, L., Dai, L., Ji, N., Qin, Y., Shi, R., Qiao, Y., Xiong, L. and Sun, Q., 2023. Hydrophobic biopolymer-based films: Strategies, properties, and food applications. Food Engineering Reviews, 15(2), pp.360-379.
4. Shah, Y.A., Bhatia, S., Al-Harrasi, A., Oz, F., Khan, M.H., Roy, S., Esatbeyoglu, T. and Pratap-Singh, A., 2024. Thermal properties of biopolymer films: Insights for sustainable food packaging applications. Food Engineering Reviews, 16(4), pp.497-512.
5. Li, X., Li, J., Shen, X., Cao, M., Wang, Y., Zhang, W., Xu, Y., Ling, Z., Chen, S. and Xu, F., 2024. Transparent cellulose/lignin composite films with adjustable haze and UV-blocking performance for light management. ACS Sustainable Chemistry & Engineering, 12(14), pp.5427-5435.
6. Rincón, E., De Haro-Niza, J., Morcillo-Martín, R., Espinosa, E. and Rodríguez, A., 2023. Boosting functional properties of active-CMC films reinforced with agricultural residues-derived cellulose nanofibres. RSC advances, 13(35), pp.24755-24766.
7. Šimkovic, I., Gucmann, F., Dobročka, E., Filip, J., Hricovíni, M., Dujnič, V., Mendichi, R., Schieroni, A.G., Piovani, D., Zappia, S. and Hricovíni, M., 2024. Properties of quaternized and cross-linked hydroxyethylcellulose composite films. Cellulose, 31(17), pp.10341-10357.
8. Cen, C., Wang, F., Wang, Y., Li, H., Fu, L., Li, Y., Chen, J. and Wang, Y., 2023. Design and characterization of an antibacterial film composited by hydroxyethyl cellulose (HEC), carboxymethyl chitosan (CMCS), and nano ZnO for food packaging. International Journal of Biological Macromolecules, 231, p.123203.
9. Tohamy, H.A.S., El-Sakhawy, M., El-Masry, H.M., Saleh, I.A. and AbdelMohsen, M.M., 2022. Preparation of hydroxyethyl cellulose/mangiferin edible films and their antimicrobial properties. BMC chemistry, 16(1), p.113.
10. Jo, C., Rukmanikrishnan, B., DS, P., Ramalingam, S. and Lee, J., 2021. Cellulose-pulp-based stretchable composite film with hydroxyethyl cellulose and turmeric powder for packaging applications. ACS Sustainable Chemistry & Engineering, 9(40), pp.13653-13662.
11. Núñez-Flores, R., Giménez, B., Fernández-Martín, F., López-Caballero, M.E., Montero, M.P. and Gómez-Guillén, M.C., 2013. Physical and functional characterization of active fish gelatin films incorporated with lignin. Food Hydrocolloids, 30(1), pp.163-172.
12. Zadeh, E.M., O'Keefe, S.F. and Kim, Y.T., 2018. Utilization of lignin in biopolymeric packaging films. ACS omega, 3(7), pp.7388-7398.
13. Brandt, A., Gräsvik, J., Hallett, J.P. and Welton, T., 2013. Deconstruction of lignocellulosic biomass with ionic liquids. Green chemistry, 15(3), pp.550-583.
14. Prado, R., Erdocia, X., De Gregorio, G.F., Labidi, J. and Welton, T., 2016. Willow lignin oxidation and depolymerization under low cost ionic liquid. ACS Sustainable Chemistry & Engineering, 4(10), pp.5277-5288.
15. Vanholme, R., Morreel, K., Ralph, J. and Boerjan, W., 2008. Lignin engineering. Current opinion in plant biology, 11(3), pp.278-285.
16. Ragauskas, A.J., Beckham, G.T., Biddy, M.J., Chandra, R., Chen, F., Davis, M.F., Davison, B.H., Dixon, R.A., Gilna, P., Keller, M. and Langan, P., 2014. Lignin valorization: improving lignin processing in the biorefinery. science, 344(6185), p.1246843.

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This project has received funding from the European Union Horizon Research and Innovation programme under grant agreement No 101178059

17. Li, M., Pu, Y. and Ragauskas, A.J., 2016. Current understanding of the correlation of lignin structure with biomass recalcitrance. Frontiers in chemistry, 4, p.45.
18. He, Y., Ye, H.C., You, T.T. and Xu, F., 2023. Sustainable and multifunctional cellulose-lignin films with excellent antibacterial and UV-shielding for active food packaging. Food Hydrocolloids, 137, p.108355.
19. Shankar, S., Reddy, J.P. and Rhim, J.W., 2015. Effect of lignin on water vapor barrier, mechanical, and structural properties of agar/lignin composite films. International journal of biological macromolecules, 81, pp.267-273.
20. Yang, F., Xu, L., Dai, G., Luo, L., Yang, K., Huang, C., Tian, D. and Shen, F., 2022. Conversion of cellulose and lignin residues into transparent UV-blocking composite films. Molecules, 27(5), p.1637.
21. Li, X., Liu, Y. and Ren, X., 2022. Transparent and ultra-tough PVA/alkaline lignin films with UV shielding and antibacterial functions. International journal of biological macromolecules, 216, pp.86-94.
22. Zhang, X., Liu, W., Liu, W. and Qiu, X., 2020. High performance PVA/lignin nanocomposite films with excellent water vapor barrier and UV-shielding properties. International journal of biological macromolecules, 142, pp.551-558.
23. Pontillo, A.R.N., Chen, S., Ordóñez, D.F., Mac Dowell, N., Lee, K.Y. and Welton, T., 2025. Fabricating lignocellulosic films as potential biobased plastics. RSC Sustainability, 3(5), pp.2301-2310.
24. Guo, D., Wu, S., Lyu, G. and Guo, H., 2017. Effect of molecular weight on the pyrolysis characteristics of alkali lignin. Fuel, 193, pp.45-53.
25. Pińkowska, H., Wolak, P. and Złocińska, A., 2012. Hydrothermal decomposition of alkali lignin in sub- and supercritical water. Chemical Engineering Journal, 187, pp.410-414.
26. Ferreira, D.C.M., Ferreira, S.O., de Alvarenga, E.S., Soares, N.D.F.F., dos Reis Coimbra, J.S. and de Oliveira, E.B., 2022. Polyelectrolyte complexes (PECs) obtained from chitosan and carboxymethylcellulose: A physicochemical and microstructural study. Carbohydrate Polymer Technologies and Applications, 3, p.100197.
27. Li, L., Baig, M.I., de Vos, W.M. and Lindhoud, S., 2023. Preparation of sodium carboxymethyl cellulose–chitosan complex membranes through sustainable aqueous phase separation. ACS Applied Polymer Materials, 5(3), pp.1810-1818.
28. Altam, A.A., Zhu, L., Huang, W., Huang, H. and Yang, S., 2021. Polyelectrolyte complex beads of carboxymethylcellulose and chitosan: The controlled formation and improved properties. Carbohydrate Polymer Technologies and Applications, 2, p.100100.
29. Vieira, M. G. A.; da Silva, M. A.; dos Santos, L. O.; Beppu, M. M. Natural-based plasticizers and biopolymer films: A review. European Polymer Journal 2011, 47(3), 254–263.
30. Malik, G.K., Khuntia, A. and Mitra, J., 2022. Comparative effect of different plasticizers on barrier, mechanical, optical, and sorption properties of hydroxypropyl methylcellulose (HPMC)–based edible film. Journal of Biosystems Engineering, 47(2), pp.93-105.
31. Ruwoldt, J., Blindheim, F.H. and Chinga-Carrasco, G., 2023. Functional surfaces, films, and coatings with lignin–a critical review. RSC advances, 13(18), pp.12529-12553.
32. Faradilla, R.F., Lee, G., Sivakumar, P., Stenzel, M. and Arcot, J., 2019. Effect of polyethylene glycol (PEG) molecular weight and nanofillers on the properties of banana pseudostem nanocellulose films. Carbohydrate Polymers, 205, pp.330-339.
33. Zhang, M., Li, X.H., Gong, Y.D., Zhao, N.M. and Zhang, X.F., 2002. Properties and biocompatibility of chitosan films modified by blending with PEG. Biomaterials, 23(13), pp.2641-2648.
34. Sanyang, M.L., Sapuan, S.M., Jawaid, M., Ishak, M.R. and Sahari, J., 2016. Effect of plasticizer type and concentration on physical properties of biodegradable films based on sugar palm (Arenga pinnata) starch for food packaging. Journal of food science and technology, 53(1), pp.326-336.

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This project has received funding from the European Union Horizon Research and Innovation programme under grant agreement No 101178059

35. Silva-Weiss, A., Ihl, M., Sobral, P.D.A., Gómez-Guillén, M.C. and Bifani, V.J.F.E.R., 2013. Natural additives in bioactive edible films and coatings: functionality and applications in foods. Food Engineering Reviews, 5(4), pp.200-216.
36. Paudel, S., Regmi, S. and Janaswamy, S., 2023. Effect of glycerol and sorbitol on cellulose-based biodegradable films. Food Packaging and Shelf Life, 37, p.101090.
37. Paluch, M.; Ostrowska, J.; Tyński, P.; Sadurski, W.; Konkol, M. Structural and Thermal Properties of Starch Plasticized with Glycerol/Urea Mixture. Journal of Polymers and the Environment 2022, 30, 728–740.
38. Sudheesh, C., Varsha, L., Siddiqui, S.A., Sunooj, K.V. and Pillai, S., 2024. Exploring urea as a prospective auxiliary for starch functionalization: A concise review on modified starch properties and the sustainable packaging films. Food Chemistry, 455, p.139914.
39. Gupta, R. K.; Manisha; Ali, E. A. E.; Areekal, N. N.; Patel, J.; Pipliya, S.; Srivastav, P. P.; Wang, C.-K.; Castro-Muñoz, R. Vitamin C-infused biopolymer films: a multifunctional approach for active food packaging and preservation. Sustainable Food Technol., 2026,4, 25-50.

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