Biobased removable textile prints

Exploration of biobased binders for the purpose of screen printing

Degree project report in Nanotechnology

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Cover: Illustration of a white t-shirt with a black print saying "second chance".

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Abstract

The textile industry has a substantial environmental and climate impact. One approach to reducing this footprint is to increase the reuse and recyclability of post-consumer garments. Vividye addresses this challenge by enabling removable textile prints through a detachable priming layer beneath conventional inks.

This thesis investigates the development of a biobased alternative to the currently used latex binder in the priming layer, with the goal of maintaining key performance properties such as removability, printability, and washability. Four biobased binders were explored: an emulsion of crosslinked castor oil, polylactic acid (PLA) powder, powdered ethyl cellulose, and an aqueous ethyl cellulose dispersion, commercially known as Surelease®.

The most promising formulation used Surelease® as the main binder and PLA as a filler. To improve rheological behavior, a cellulose derivative was added as a thickener. Multiple formulations with varying ratios of these components were prepared and evaluated through film casting and visual inspection. Wettability was assessed using an optical goniometer. While the final formulation did not fully match the film forming and washability performance of the original latex-based system, it demonstrated comparable removability and superior printability. These results demonstrate the potential of biobased alternatives for more sustainable textile printing.

Keywords: Biobased, Formulation, Priming layer, Removable textile print, Reuse, Screen printing, Biobased binders.

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My time at Chalmers has not always been a walk in the park, but after six years, I've made it - and I'm proud. I could not have done this without generous and thoughtful classmates. Thank you for the discussions, support and lunch company.

I would also like to thank my husband and my mom. Daniel, without you I probably would have given up during the pandemic - thank you for stopping me from dropping out to become an arborist or a knitting influencer. Mom, thank you for picking me up while crying after failed exams (more than once), and for always having my back.

It's been a journey. I've learned a lot, and I'm grateful for every part of it - but now, I'm definitely ready for the next step.

Vera Andersson, Gothenburg, June 2025

List of Acronyms

Below is the list of acronyms that have been used throughout this thesis listed in alphabetical order:

BTCA	1,2,3,4-Butanetetracarboxylic acid
DINCH	1,2-Cyclohexane dicarboxylic acid diisononyl ester
DLS	Dynamic Light Scattering
MFFT	Minimum Film Forming Temperature
PDI	Poly(hexamethylene diisocyanate)
PLA	Polylactic Acid

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Introduction

1.1 Background

The textile industry exerts a significant negative impact on both the environment and the climate. Life-cycle assessments reveal that environmental harm occurs at every step of the process, from raw material extraction and textile production to manufacturing, transportation, consumer use (including washing), and final disposal of garments [1].

In parallel, the fast fashion industry is continuously growing and the demand for clothing is expected to rise by 2 % a year [1], but at the same time garments are used fewer times than ever. This trend not only accelerates consumption but also amplifies the environmental footprint of the textile sector.

One notable example is the extensive water consumption required for textile production: it is estimated that the manufacture of a single cotton t-shirt consumes approximately 2700 liters of freshwater [2]. Beyond water use, both textile production and the laundering of synthetic garments release microplastics and other pollutants into aquatic environments. These contaminants pose risks to ecosystems and drinking water supplies. Dyeing and textile treatment is responsible for an estimated 20 % of global industrial water pollution [1], and it contributes to nearly 10 % of annual microplastic emissions [3]. Alarmingly, over half a million tonnes of microplastics from synthetic fabrics are deposited into the oceans each year as a result of washing processes [2].

In addition to pollution, textile waste presents a major challenge. Approximately 87 % of fibers used in clothing production are ultimately incinerated or sent to landfills - equivalent to the disposal of one garbage truck of textiles every second [2].

Increasing reuse and recycling of post consumer garments could mitigate these issues by reducing the demand for new raw materials, water, energy and minimizing waste accumulation [4]. Despite these advantages, several challenges hinder the scalability and effectiveness of current recycling initiatives. From a technical perspective, recycling textiles is complicated by the need to sort garments based on fiber composition. Furthermore many textiles consist of blended materials, which poses challenges for mechanical and chemical recycling processes that typically require uniform input streams [4]. In addition to fiber blends, printed garments represent another challenge. For instance, a cotton t-shirt printed with plastic-based inks becomes a composite material, rendering it unsuitable for standard recycling processes due to the incompatibility of the mixed components.

One innovative solution to address part of this challenge involves the development of printable inks designed for controlled removal. These inks enable the removal of printed designs from garments without damaging the underlying textile, thereby allowing for reprinting or more efficient recycling. This is particularly beneficial when separating synthetic ink residues from natural fibers like cotton, improving the recyclability of the material.

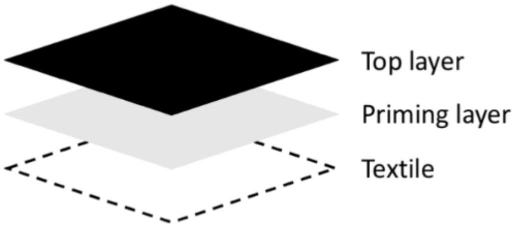


Figure 1.1: Schematic illustration of the layering technique showing the textile, the priming layer and the top layer.

The Swedish company Vividye has developed a novel two-layer ink system to achieve this functionality. The system consists of at least two layers: a priming layer applied directly to the fabric and a top layer containing the ink, as illustrated in Fig 1.1, the ink layer could consist of more than one layer.

The layering technique involves said sacrificial priming layer and a customized removal solution for removal on demand. At Vividye, prior research has led to the development of a proprietary priming layer formulation (referred to as FP03) and a corresponding removal solution (RS03). The components of both FP03 and RS03 are listed in Table 1.1. Due to confidentiality agreements, the exact concentrations of these components cannot be disclosed; however, selected ingredients and their roles is discussed throughout this thesis.

Table 1.1: Components of the original formulations of the priming layer (FP03) and the removal solution (RS03). Ingredients are listed in the order of amounts used from most to least.

Priming layer (FP03)	Removal solution (RS03)
Latex Water Polysaccharide Glycerol 1,2,3,4-Butanetetracarboxylic acid	Water Acetone Propylene carbonate Ethylan 1005 Ethylan 1008 Enzyme specific for degrading of the polysaccharide

As previously discussed, the priming layer plays a key role in enhancing the reuse and recyclability of garments. However, while the current formulation has biobased ingredients, it utilizes a petroleum-based latex binder. From a sustainability perspective, it is highly desirable to develop a fully biobased system, in order to reduce reliance on non-renewable resources and further minimize the environmental impact of textile treatments. This can be achieved through replacement of the binder with a biobased alternative.

1.2 Purpose and objectives

The purpose of this master's thesis is to improve the primer used for textile printing, focusing on environmental impact. This optimization can be divided into two key objectives:

1. Identify a biobased film forming binder as a sustainable alternative to the petroleum-based latex currently used in existing formulations.

2. Maintain the performance of the priming layer concerning removability, printability, and washability.

All modifications will be carried out with the goal of developing a functional, stable, and environmentally friendly product.

1.3 Limitations

To maintain a manageable scope and focus on developing a biobased formulation, several limitations were defined for this study.

No new chemical components were synthesized; only commercially available substances were used in the formulation. To ensure consistency, only cotton fabrics were selected as the substrate, given their relevance and uniform behavior in testing.

Due to time constraints, the formulation is not expected to be fully optimized. Instead, minimum performance requirements were set to evaluate its viability:

1. The formulation should form films upon curing.
2. The films should be somewhat flexible compared to other screen printed films.
3. The films should withstand exposure to water.
4. The formulation should have workable rheological behavior.

Finally, the overarching goal of contributing to a more sustainable textile industry imposes a natural constraint on the choice of ingredients. Only low-toxicity, environmentally friendly substances were used, in line with green chemistry and sustainability principles.

2

Theory

2.1 Screen printing

Screen printing is a widely used and cost-effective printing technique, known for its simplicity and minimal equipment requirements [5]. The process involves the use of a screen composed of a mesh (Fig. 2.1E) coated with a photosensitive emulsion (Fig. 2.1D). Upon exposure to light, the emulsion hardens, creating a stencil in which the unexposed areas correspond to the desired print pattern (Fig. 2.1C). During printing, ink (Fig. 2.1A) is evenly distributed across the screen using a squeegee (Fig. 2.1B). The pressure applied by the squeegee brings the mesh into contact with the substrate, allowing the ink to be transferred through the open areas of the stencil onto the textile surface. After printing, the ink is typically cured at elevated temperatures to ensure proper film formation and adhesion.

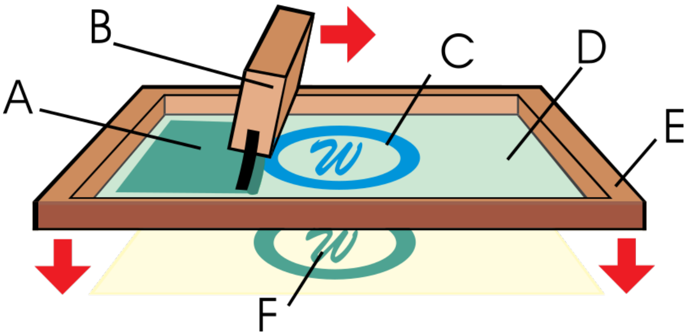


Figure 2.1: Schematic illustration of the setup for screen printing, A) ink, B) squeegee, C) negative image, D) photo emulsion, E) screen and F) printed image. *Image: Harry Wad, CC BY 2.5*

The rheological properties of the printing formulation play a crucial role in achieving clear and well-defined prints [5]. If the formulation is too viscous and not shear thinning, it may hinder ink transfer through the mesh. However, higher viscosity can also contribute to sharper edge definition in the printed pattern. Therefore, optimizing the viscosity is essential to balance printability and print quality.

2.2 Film formation

Film formation is a critical property in printing applications. It refers to the process in which a liquid material is applied to a surface and subsequently cured or dried, resulting in a solid film composed of high molecular weight polymers [6]. In the case of water-based systems, this transformation is referred to as latex film formation [7]. The process progresses from a dilute dispersion of colloidal polymer particles to a concentrated state, leading to particle packing and ultimately the formation of a continuous film. Latex film formation is conventionally described in three stages: drying, particle deformation, and polymer diffusion. These stages are illustrated in Fig 2.2.

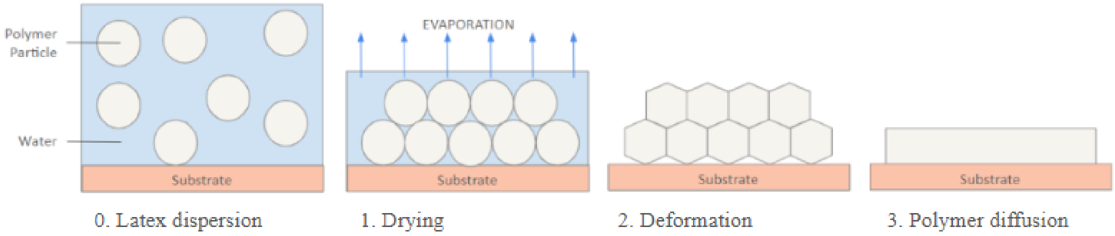


Figure 2.2: A schematic illustration of the three stages of latex film formation: drying, particle deformation, and polymer diffusion.

While most commercial inks are plastisol-based, the priming layer in this project can be classified as a water-based ink. Waterborne systems often underperform compared to solventborne ones in terms of hardness, block and print resistance among others [8]. These limitations stem from the film formation process, particularly the trade-off between the glass transition temperature (T_G) and the minimum film formation temperature (MFFT) of the binder. A high T_G ensures hardness, while a low MFFT is needed for film formation at ambient conditions. This conflict, known as the "film-formation dilemma", is a key challenge in advancing waterborne coatings.

2.3 Colloidal stability

Colloidal stability plays a crucial role in formulation chemistry and the formulation of inks. A stable formulation exhibits an extended shelf-life and resists particle agglomeration. A colloid is defined as a stable dispersion in which one phase is finely distributed within another [9]. The dispersed phase typically comprises particles with sizes ranging from approximately one nanometer to one micrometer. Due to their small size, these particles exhibit a high surface-to-volume ratio.

Achieving a stable colloidal dispersion requires minimizing destabilizing mechanisms such as sedimentation, coagulation, agglomeration and flocculation [9]. The high surface area of colloidal particles, in combination with van der Waals interactions, makes them particularly susceptible to these processes. Two main mechanisms are commonly employed to enhance colloidal stability: electrostatic and steric stabilization. These approaches can also be combined in a method known as electrosteric stabilization, offering improved resistance to coagulation through both electrostatic and steric effects.

Electrostatic stabilization is the mechanism in which the attractive van der Waals forces are counterbalanced by the repulsive Coulomb forces acting between the charged colloidal particles and are highly sensitive to changes in ionic strength [10]. An increase in ion concentration can compress the electrical double layer surrounding the colloidal particles, thereby reducing or eliminating the osmotic repulsion that prevents particle aggregation. As a result, the colloidal system may become destabilized, leading to flocculation or phase separation.

Steric stabilization arises from the adsorption or grafting of polymer chains onto the surface of colloidal particles [10]. These polymer layers create a physical barrier that prevents particles from approaching each other closely enough to dominate the attractive van der Waals forces. In contrast to electrostatic stabilization, sterically stabilized systems are significantly less sensitive to changes in ionic strength, as the stabilization mechanism does not rely on the presence of surface charges. However, flocculation can still occur under certain solvent conditions - such as poor solvent quality or specific interactions between polymer chains and the medium - but such systems often retain the ability to redisperse upon restoration of favorable solvent conditions.

2.4 Wetting and contact angle

Wetting is the ability of a liquid to spread on a surface [10]. The degree of wetting - or *wettability*, depends on the physicochemical properties of both the liquid and the solid substrate, i.e. the surface properties. Wettability is determined by the balance between adhesive forces (between the liquid and the solid surface) and cohesive forces (within the liquid itself). The spreading process continues until the system reaches equilibrium.

In the context of textiles, if a film exhibits high wettability - such as the ability to adsorb water - it is likely to influence the washability of the garment, potentially affecting the durability and removability of applied coatings or treatments.

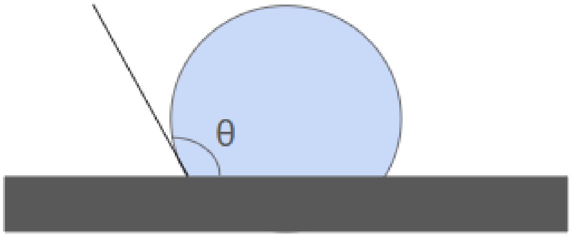


Figure 2.3: Schematic picture of a water droplet on a surface, θ represents the contact angle.

The degree of wetting is often quantified by the contact angle formed between the liquid droplet and the solid surface [10], see Fig 2.3. A low contact angle indicates high wettability, as is typically observed when water is placed on a hydrophilic surface. In contrast, a hydrophobic surface yields a contact angle of 90° or greater, indicating poor wetting behavior. In cases where surface roughness is specific and controlled, it is possible to achieve superhydrophobicity, characterized by contact angles exceeding 150°, which results in extremely low wettability.

It is important to note that a surface can exhibit hydrophobic characteristics while still showing high dynamic wettability. In such cases, the initial contact angle may be high, but the droplet rapidly spreads over time due to favorable surface interactions or topographical features.

2.5 Dissolution and swelling of polymer materials

In order to create a film that can withstand laundering and at the same time be removable, it is important to understand the dissolution and swelling of the polymers in the formulation.

Polymer dissolution in a solvent is generally considered a two-stage process [8]. In the solid state, the polymer is conformationally restricted. Upon dissolution, it gains configurational freedom and can adopt a wide range of energetically similar conformations, influenced by chain flexibility and interactions with the solvent. Flory and Huggins described this process as involving: (1) the transition from an ordered, rigid state to a disordered, flexible one suitable for random placement on a lattice, and (2) subsequent mixing with solvent molecules. However, this behavior differs for cross-linked polymer networks. Unlike linear or branched polymers, which typically dissolve in compatible solvents, cross-linked polymers do not dissolve but instead swell when exposed to suitable liquids. From a thermodynamic perspective, spontaneous dissolution occurs only when the change in Gibbs free energy is zero or negative [11].

Swelling through solvent uptake can be employed as a strategy to apply a controlled, weak mechanical load to polymer materials [12]. This property is intentionally utilized in the film removal process of the layering technology investigated in this project. Swelling occurs as solvent molecules penetrate the polymer network, leading to an increase in volume [12]. In a good solvent, favorable polymer-solvent interactions cause the polymer chains to stretch. According to the Flory-Rehner theory, swelling represents a thermodynamic equilibrium between the entropy loss due to chain deformation associated with chain stretching and the enthalpy of mixing.

3

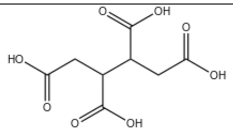
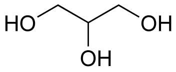
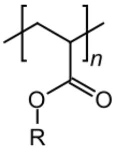
Materials and methods

3.1 Materials

3.1.1 Components of original formulation

Due to confidentiality constraints, the exact quantities of the individual components in the original formulation cannot be disclosed. However, Table 3.1 provides an overview of the components included in the original formulation.

Table 3.1: The components of the original formulation with corresponding chemical structures.

Name	Chemical structure	Source
Polysaccharide	N/A	B. Engelhardt & CO AB
1,2,3,4-Butanetetracarboxylic acid		Sigma Aldrich
Glycerol		Sigma Aldrich
ALBERDINGK® AC 2007		Alberdingk-Boley

3.1.1.1 Latex

Latex functions as a binder within the formulation. It is a colloidal dispersion composed of polymeric particles suspended in water [7]. Typically, these particles constitute approximately 50 *wt*% of the total dispersion. In the present formulation, the latex employed exhibits an average particle size of 100 nm, as determined by dynamic light scattering (DLS). The latex used in this work is ALBERDINGK® AC 2007.

3.1.1.2 Glycerol

Glycerol was used as the plasticizer in the original formulation. It is a hydrophilic plasticizer and an organic compound commonly used to enhance the flexibility and processability of polymeric materials [13], in this formulation the polymeric material is the polysaccharide.

3.1.1.3 1,2,3,4-Buthanetetracarboxylic acid

1,2,3,4-Butanetetracarboxylic acid (BTCA) was used as a crosslinking agent for the polysaccharide in the formulation. This crosslinking enhances the structural integrity and water resistance of the resulting film.

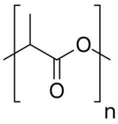
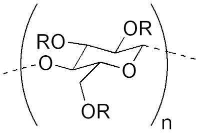
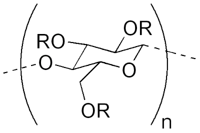
3.1.1.4 Specific polysaccharide

A polysaccharide is incorporated into the formulation to enhance the removability of the film. Upon exposure to the enzyme in the removal solution, the polysaccharide is degraded, allowing for swelling and facilitating easier detachment of the film from the substrate.

3.1.2 Alternative biobased binders

The objective of this project was to identify a biobased binder with properties comparable to those of conventional petroleum-based latex. An ideal binder would enable the formulation to exhibit strong adhesion to cotton substrates, appropriate rheological characteristics, and the ability to cure in 1 to 2 minutes in a temperature range of 150–170 °C. Furthermore, the resulting films should be flexible and durable, capable of withstanding laundering, yet removable using a specific removal solution. An overview of the biobased binders evaluated is provided in Table 3.2.

Table 3.2: Biobased binders with corresponding chemical structures used in this work.

Name	Chemical structure	Source
Polylactic acid		Centexbel
Ethyl cellulose	 R = H or CH_2CH_3	Sigma Aldrich
Surelease® 19040	 R = H or CH_2CH_3 +additives	Colorcon

3.1.2.1 Polylactic acid

Polylactic acid (PLA) has emerged as a widely used biobased alternative to petroleum-based plastics, primarily due to the cost-effective production of renewable materials such as starch [14]. PLA is considered biodegradable; however, its degradation requires specific conditions, particularly exposure to temperatures above its glass transition temperature (approximately 60 °C). Complete degradation typically requires industrial composting facilities where such temperature conditions can be consistently maintained. Although PLA is often labeled as biodegradable, this classification can be misleading, as it does not readily degrade under natural environmental conditions [15].

3.1.2.2 Ethyl cellulose

Two types of ethyl cellulose were introduced as binders: powdered ethyl cellulose and an aqueous dispersion commercially known as Surelease®. While the exact composition of the aqueous dispersion is not disclosed, the technical data sheet specifies a solid content of approximately 25 *wt*%.

Ethyl cellulose is a hydrophobic derivative of cellulose commonly used in pharmaceutical applications, particularly as a coating material for tablets. Its widespread use is attributed to properties such as non-toxicity, chemical stability and its film forming properties [16].

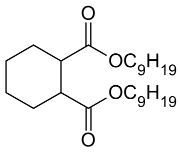
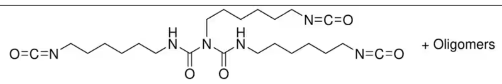
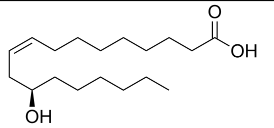
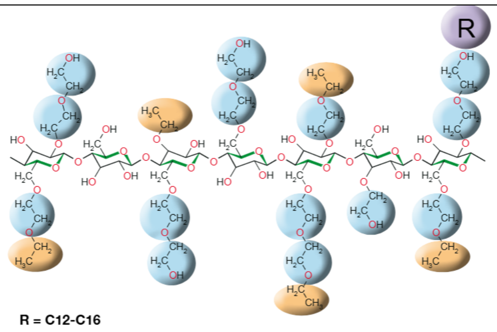
3.1.2.3 Emulsion with crosslinked castor oil

An emulsion based on crosslinked castor oil was evaluated as a potential binder system. Crosslinking was achieved using poly(hexamethylene diisocyanate) (PDI), and the emulsion also contained nanocrystalline cellulose, water, and calcium carbonate.

3.1.3 Alternative additives and components

In addition to binder substitution, several alternative additives and components were incorporated into the original formulation to modify and optimize its performance. A complete list of these components is presented in Table 3.3.

Table 3.3: Other components and additives used within the project and their corresponding chemical structures.

Name	Chemical structure	Source
1,2-Cyclohexane dicarboxylic acid diisononyl ester		BASF
Poly(hexamethylene diisocyanate).		Sigma-Aldrich
Castor oil		Sigma-Aldrich
Bermocoll® EHM500		Nouryon

3.1.3.1 Bermocoll® EHM500

Bermocoll® EHM500 is used as a thickener to improve rheological behavior of latex paints [17]. It is a non-ionic, water-soluble cellulose ether which is dissolved in water prior to use, in this work, the concentration will be 1 *wt*%. The purpose of using EHM500 in this formulation is to increase the viscosity.

3.1.3.2 1,2-Cyclohexane dicarboxylic acid diisononyl ester

1,2-Cyclohexane dicarboxylic acid diisononyl ester (DINCH) is a colorless, hydrophobic plasticizer commonly used as a non-phthalate alternative in various applications [18]. In this study, DINCH was selected as an alternative plasticizer due to its hydrophobic character, offering a contrast to the hydrophilic nature of glycerol previously used in the formulation.

3.2 Formulation of priming layer

The original formulation was prepared at the beginning of the project to develop relevant laboratory skills and to establish a benchmark for comparison with newly developed formulations. It involves several steps to ensure proper function of the formulation.

3.3 Preliminary evaluation of alternative binder

The different binder alternatives were evaluated by preparing formulations and conducting a preliminary assessment based on predefined minimum performance criteria (see Limitations 1.3). Following this initial screening, two binders were selected for more systematic and detailed evaluation.

3.4 Film preparation

Films were prepared using two different methods: in the first, the formulation was spread onto a Teflon-mould using a spatula. In the second, it was applied to a cotton textile using a large spatula. Both types of films were subsequently cured in an oven at approximately 180 °C.

During the preliminary evaluation, layers with a thickness of 200 μm were cured on microscope glass slides for further assessment.

3.5 Study of interactions between ingredients in formulation

To enable efficient formulation development, part of the project focused on understanding the interactions between the various potential components of the formulation.

3.5.1 Light scattering

DLS, or Dynamic light scattering (Litesizer DLS 500, Anton Paar) was performed to determine the particle size distributions of various samples. This analysis aimed to provide insight into the role of individual components within the formulation and thereby enabling informed adjustments to their respective concentrations.

Several of the DLS measurements were conducted as part of a sequential addition series. A stock solution was first prepared and diluted with Milli-Q water to a concentration of approximately 0.1 $wt\%$. 1 mL of this diluted solution was transferred to a cuvette, and the initial particle size distribution was recorded. Subsequently, 20 μL of a second solution - containing components from both the original formulation and the new additives depending on the measurement, at an approximate concentration of 1 $wt\%$ - was added to the cuvette, followed by another measurement. This addition and measurement cycle was repeated, with an additional 20 μL added prior to each step, until a total of six measurements had been performed. This approach allowed for monitoring of particle size changes in response to the incremental introduction of formulation components.

3.5.2 Contact angle measurements

Contact angle measurements were carried out using an optical goniometer (Attension) to evaluate the wettability of the films. Samples incorporating different types and concentrations of plasticizers were analyzed to assess the influence of plasticizer composition on the overall performance of the formulation.

For sample preparation, a film of the formulation was cured directly onto a piece of cotton textile. After curing, the film was cut into strips approximately 1.5 × 3 cm in size. Each strip was then mounted onto a microscope glass slide using double-sided adhesive tape to ensure flatness and stability during measurement.

3.6 Optical microscopy

An optical microscope (Zeiss Stemi 508) was used to examine the cured formulation on textiles to get an understanding of how the priming layer coated the fibers.

3.7 Evaluation of formulations

The formulations were evaluated continuously throughout the development process, based on the previously defined minimum performance requirements. In practice, this included assessments of formulation viscosity, the visual and tactile properties of the cured films, and the wettability of the film surfaces.

3.8 Removability

The original removal solution (RS03) was tested on the formulation that exhibited the most promising properties.

For sample preparation, the film was cured directly onto a piece of cotton textile. After curing, the film was cut into six strips, each approximately 1.5 × 3 cm in size.

Two glass bottles with screw caps were prepared, each containing a different solution. One bottle contained 100 g of water, while the other contained a mixture of 50 g water and 50 g RS03. Both bottles were placed in a convection oven set to 40 °C. Once the solutions reached the target temperature, three textile strips were placed in each bottle, and the bottles were returned to the oven. The condition of the films was monitored every 10 minutes over the course of 1 hour.

3.9 Screen printing

As a last step, the formulation exhibiting the most promising properties was selected for screen printing alongside the original formulation. Screen printing was performed in the lab to compare the rheological behavior and printability of the new formulation relative to the original formulation.

4

Results and discussion

4.1 Preliminary evaluation of alternative binder

Preliminary evaluations of the various binder candidates identified PLA powder and the ethyl cellulose dispersion Surelease® as the most suitable components for the intended formulation. However, both materials exhibited limitations when used individually. PLA demonstrated favorable rheological behavior, and its viscosity could be conveniently adjusted by varying the amount. Nevertheless, films produced with PLA alone were brittle and lacked sufficient flexibility. In contrast, Surelease® exhibited good film forming capabilities but suffered from inadequate rheological performance due to its inherently low viscosity.

To address these limitations, a combined formulation was developed in which Surelease® served as the primary binder and PLA functioned as a filler. This combined the strengths of both materials, resulting in a formulation with improved rheological behavior and better mechanical performance of the cured films.

The castor oil emulsion demonstrated favorable rheological properties, indicating good processability for screen printing applications. However, it produced an oily tactile finish on the printed surface, which is undesirable for textile use. Additionally, the castor oil formulation crosslinked with PDI was excluded from further evaluation due to regulatory and safety concerns. PDI is classified as a potential allergen and is subject to strict handling requirements under EU regulations governing diisocyanates.

The powdered form of ethyl cellulose was also discarded due to solubility challenges. As ethyl cellulose is not water-soluble, it tended to form aggregates within the formulation, negatively affecting homogeneity and processability.

4.2 Evaluation of interactions between components in formulation

4.2.1 Particle size

DLS measurements were conducted in sequential addition series according to the methodology section (3.5.1), and data was plotted on graphs with the relative concentration of the added formulation in Surelease®. The different added solutions were the specific polysaccharide (1 *wt*%), EHM500 (1 *wt*%), BTCA (1 *wt*%) and a mixture of approximately 2.5 % BTCA (1 *wt*%), 72 % EHM500 (1 *wt*%) and 25 % polysaccharide (1 *wt*%).

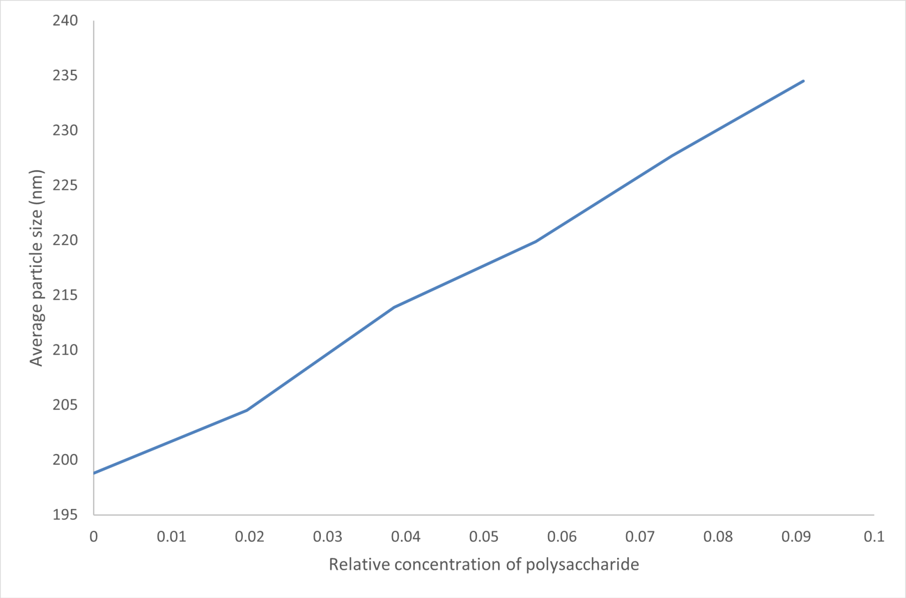


Figure 4.1: Average particle size as a function of relative concentration in a sequential addition series of polysaccharide (1 *wt*%) to Surelease® (0.1 *wt*%).

The addition of the polysaccharide to Surelease® suspension resulted in a slight, approximately linear increase in particle size, as shown in Fig 4.1. The underlying mechanism for this increase is not entirely clear. One plausible explanation is related to the effect of increased viscosity upon the addition of larger quantities of the polysaccharide. Higher viscosity reduces the rate of Brownian motion, which in turn can affect the DLS measurements.

Since DLS estimates particle size based on the diffusion behavior of particles in a suspension, the reduced mobility in more viscous solutions may be misinterpreted by the instrument as an increase in hydrodynamic diameter. Therefore, the observed particle size growth may not reflect actual aggregation or structural changes but rather an apparent effect due to changes in the rheological properties of the medium.

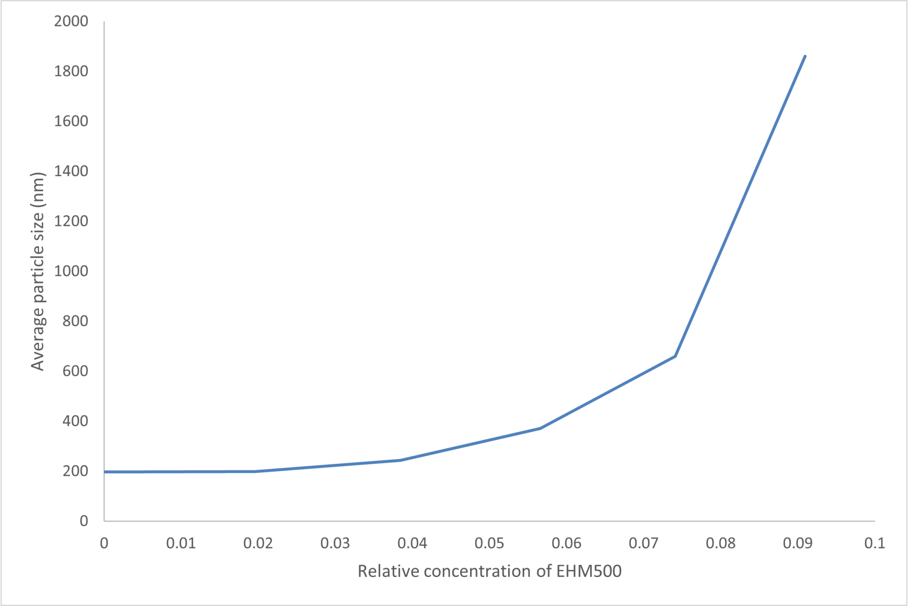


Figure 4.2: Average particle size as a function of relative concentration in a sequential addition series of EHM500 (1 *wt*%) to Surelease® (0.1 *wt*%).

Fig 4.2 illustrates that the particle size of the dispersion increases almost exponentially with the incremental addition of EHM500 to Surelease®. This pronounced growth in particle size is likely a result of interactions between the hydrophilic EHM500 and the hydrophobic components of Surelease®, leading to destabilization or aggregation within the system.

To ensure that the observed increase in particle size was not solely due to the introduction of inherently larger particles, independent DLS measurements were performed on EHM500 (1 *wt*%). The results showed an average particle size of approximately 165 nm, which is smaller than the measured particle size of Surelease® alone (approximately 195 nm). This confirms that the increase in particle size observed during the combination of the two components is due to intermolecular interactions rather than simple additive effects.

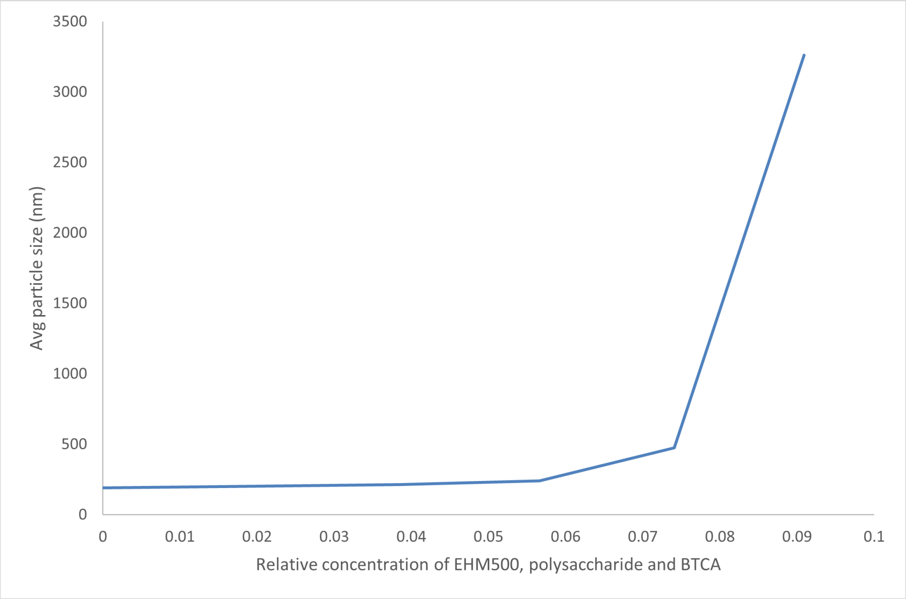


Figure 4.3: Average particle size as a function of relative concentration in a sequential addition series of a mixture of EHM500 (1 *wt*%), polysaccharide (1 *wt*%) and BTCA (1 *wt*%) to Surelease® (0.1 *wt*%).

Fig 4.3 shows a significant increase in particle size following the addition of a mixed solution containing 2.5 % BTCA (1 *wt*%), 72 % EHM500 (1 *wt*%), and 25 % polysaccharide (1 *wt*%). The particle size remains relatively stable throughout the initial measurements, but a sharp increase is observed during the final two additions.

Previous experiments indicated that the interaction between EHM500 and Surelease® alone led to substantial particle growth, with sizes reaching approximately 1900 nm. In contrast, the final measurement in the present experiment shows particle sizes exceeding 3000 nm, suggesting that the presence of BTCA and the polysaccharide contributes to further destabilization or aggregation of the system. This enhanced effect may be the result of synergistic interactions among the hydrophilic EHM500, the crosslinking agent BTCA, and the polysaccharide, which together disrupt the colloidal stability of the Surelease® dispersion more severely than EHM500 alone.

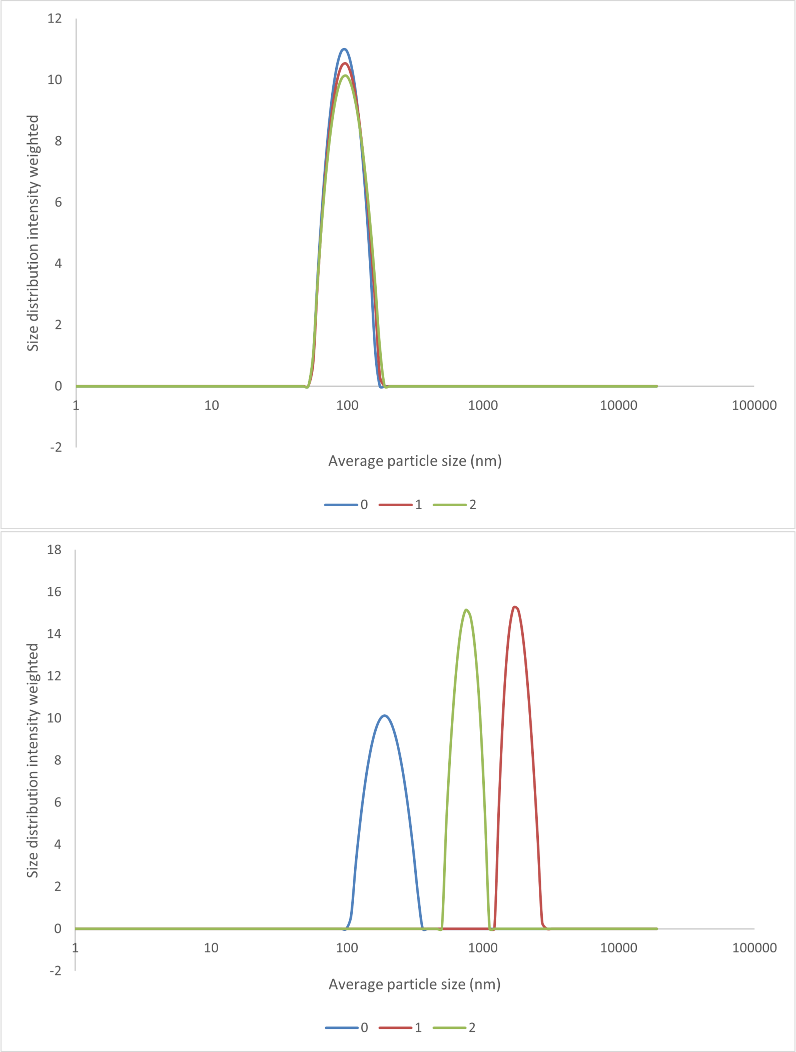


Figure 4.4: DLS measurements showing the size distribution of particle size when BTCA (1 *wt*%) is in introduced in a sequential addition series to the stock solution (latex (top) and Surelease® (bottom)). 0) first measurement (only stock solution), 1) after first addition of BTCA, 2) after second addition of BTCA. The scale of the x-axis is logarithmic.

To understand the synergetic effects of the addition of BTCA and the polysaccharide to the system, further testing was performed. Fig 4.4 shows the effect of adding BTCA to two different binder systems. When BTCA is introduced to the reference latex, no significant change in particle size is observed, indicating minimal or no interaction between BTCA and the latex particles. In contrast, the addition of BTCA to Surelease® results in a pronounced increase in particle size. During the measurement, visible white aggregates were observed in the cuvette, further suggesting colloidal instability. Several factors may account for this behavior. The increase in particle size could be due to a pH shift affecting colloidal stability, or it may result from specific interactions between BTCA and one or more components in the aqueous phase of Surelease®. These components are likely included to stabilize the ethyl cellulose particles and prevent aggregation. Disruption of this stabilization mechanism could explain the observed flocculation.

4.2.2 pH

The pH of Surelease® with added BTCA was measured to investigate whether the observed flocculation was caused by pH-induced destabilization of the system. The pH measurements were conducted using the same sequential addition method as employed in the DLS experiments. Although the precise formulation of Surelease® is proprietary and not publicly available, the observed pH of the diluted dispersion (Fig 4.5) suggests the presence of a basic component, as ethyl cellulose - the primary film forming agent in Surelease® - is inherently pH-neutral.

As shown in the figure, a significant decrease in pH was recorded during the addition of BTCA, starting from approximately pH 10 and decreasing to nearly pH 4 after the full 100 μL of BTCA solution had been added. This substantial shift indicates that the aggregation observed upon BTCA addition may be driven, at least in part, by the resulting change in pH.

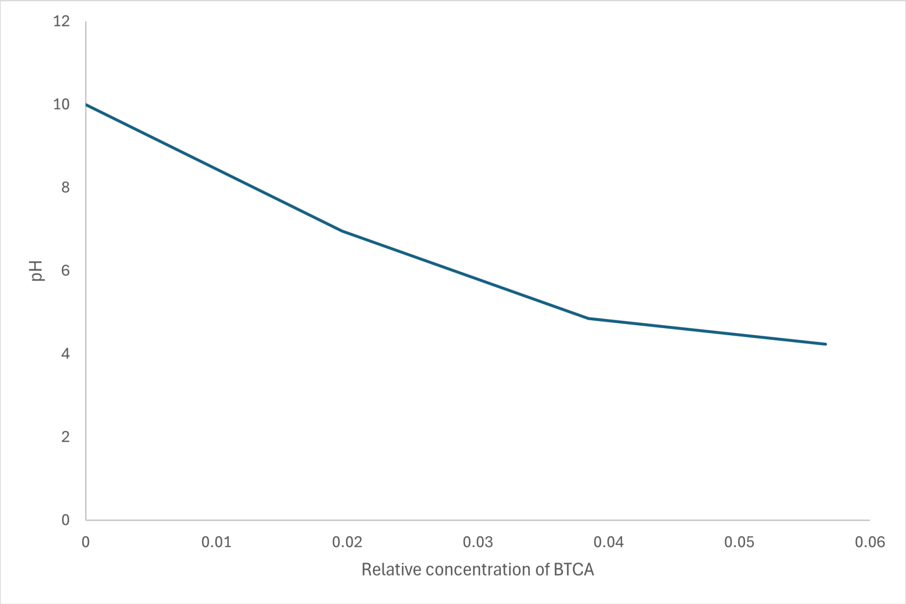


Figure 4.5: pH as a function of relative concentration in a sequential addition series of BTCA (1 *wt*%) to Surelease® (0.1 *wt*%).

However, when BTCA is introduced in combination with EHM500 and the polysaccharide (Fig 4.6), the overall change in pH is minimal. This suggests that under these conditions, pH-induced destabilization is less likely to occur.

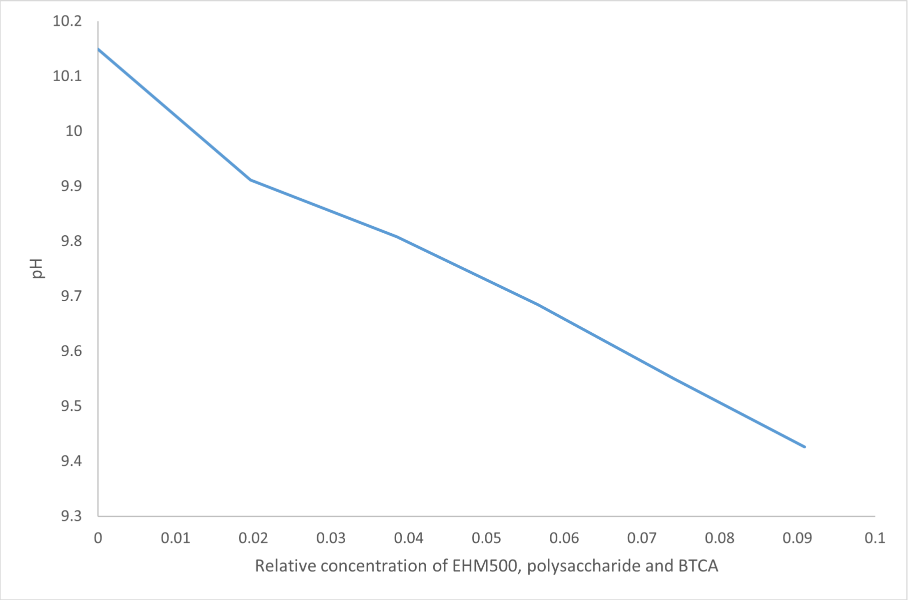


Figure 4.6: pH as a function of relative concentration in a sequential addition series of a mixture of EHM500 (1 *wt*%), polysaccharide (1 *wt*%) and BTCA (1 *wt*%) to Surelease® (0.1 *wt*%).

4.3 Formulations

Based on earlier findings, five formulations were selected for detailed characterization, including film formation and wettability measurements. The compositions, shown in Table 4.1, were chosen primarily for their film forming ability and rheological behavior. As previously mentioned, a combination of PLA and Surelease® showed most promising results. In this formulation, Surelease® acted as the primary binder, while PLA served as a structural filler to enhance mechanical strength and rheology. This blend proved effective: PLA improved the rheological profile, and viscosity could be tuned by adjusting its content. Various binder ratios and two plasticizers - glycerol and Hexamoll® DINCH - were also evaluated.

Table 4.1: Different ratios of the components of the formulations. The percentages corresponds to the amount of latex in the original formulation. The other components from the original formulation is the same amounts as in the original formulation.

Sample name	PLA powder	Surelease®	EHM	Plasticizer
A1	50 %	25 %	-	Glycerol
A2	50 %	20.5 %	4.8 %	Glycerol
A3	25 %	25 %	19 %	Glycerol
B1	50 %	20.5 %	4.8 %	Hexamoll® DINCH
B2	27 %	25 %	19 %	Hexamoll® DINCH

EHM500 was incorporated into the formulation as a thickening agent, with the objective of reducing the required amount of PLA and thereby mitigating the brittleness in the resulting films.

4.4 Film formation

A notable difference between the original formulation and those incorporating PLA is the extent of film coverage on the textile substrate. Microscopic examination of the original formulation reveals that the individual stitches remain clearly visible beneath the applied film. In contrast, formulations containing PLA result in a more continuous coating, obscuring the underlying textile structure such that the stitches are scarcely visible. A visual comparison is presented in Fig 4.7.

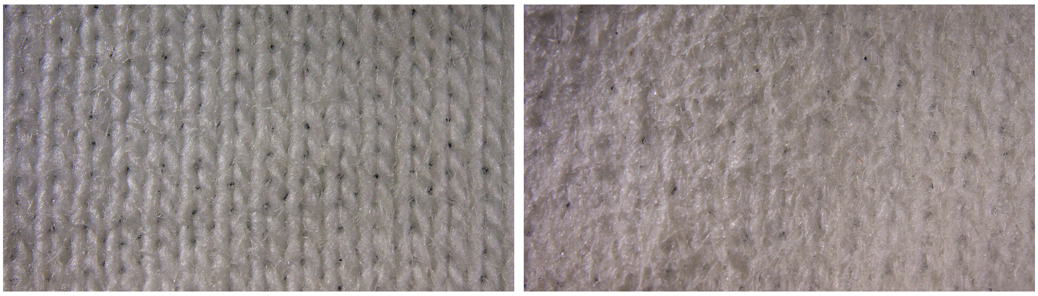


Figure 4.7: Films coating a cotton substrate. The original formulation is to the left and to the right is a formulation containing PLA.

All formulations discussed in the previous section contained both PLA powder and Surelease®. None of these formulations resulted in films that could be successfully peeled from the Teflon-mould and hence produce a self-standing film. Instead, all films were brittle and porous, indicating poor film integrity and cohesion. An example is shown in Fig 4.8, which displays the film obtained from formulation A3, the remaining films exhibited similar characteristics.

Figure 4.8: A3 as a cured film on a Teflon-mould. The uneven surface on the picture to the right is caused by scraping of a spatula.

Although the films did not exhibit satisfactory properties when cured on a Teflon-mould, their performance on textile substrates appeared significantly more promising. When applied to cotton fabric, the films demonstrated high flexibility and adhesion, withstanding substantial bending and stretching without visible cracking.

The difference in performance between the two substrates may be attributed to several factors. One possibility is the reduced film thickness on the textile, which may lead to enhanced flexibility. Another contributing factor could be interactions between the formulation components and the cotton fibers, potentially improving film integrity on the textile surface.

While the formulations did not produce self-standing films with sufficient mechanical strength, this limitation may not be critical, given that the intended application is as a coating on cotton textiles. As long as the films perform adequately under typical conditions for printed textiles, the absence of self-standing film capability does not compromise their suitability for the target application.

4.4.1 Influence of plasticizer

Glycerol was used to plasticize the polysaccharide in the formulation. To investigate the effect of the plasticizer on film properties, varying amounts of glycerol were added to the formulation “A1”, see Table 4.1. The tested ratios included 1, 2, and 5 times the original amount of glycerol. These formulations were cured both on the Teflon-moulds and on textile substrates.

The thicker films cured on the Teflon-moulds were used to assess whether increasing the plasticizer content could reduce the brittleness typically associated with PLA. All samples appeared similar in visual and physical characteristics - brittle and porous, see Fig 4.8. In all cases, the films could be scraped off as brittle flakes using a spatula, but it was not possible to peel off an intact film layer.

Films cured on textile substrates were used for contact angle measurements to evaluate the impact of glycerol content on wettability. The results demonstrated that increasing the amount of glycerol led to higher wettability, as shown in Fig 4.9. Based on these observations, the formulation with the lowest glycerol content (equivalent to that in the original formulation) was selected as the most suitable, since lower wettability is desirable to enhance the film's washability.

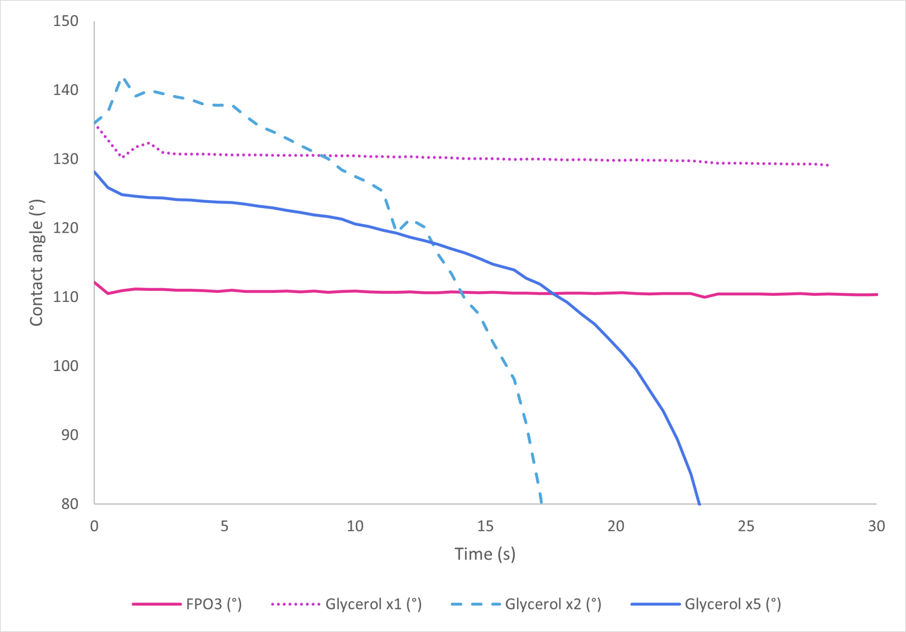


Figure 4.9: Wettability and contact angle measurements of films made of the original formulation and 3 modified formulations containing PLA, Surelease® and varying amounts of glycerol (1×, 2×, and 5× the original concentration).

Given that glycerol is a hydrophilic plasticizer, a more hydrophobic alternative - Hexamoll® DINCH - was tested in its place to target the binder. The same quantity as in the original formulation was used. A new formulation, "B1", (see Table 4.1) was prepared with Hexamoll® DINCH as the only variable. This formulation was also cured on both Teflon-moulds and textile substrates.

The thicker films produced with Hexamoll® DINCH showed a slight improvement in mechanical properties; larger flakes could be peeled off, although the films remained porous and brittle. Contact angle measurements again indicated an increase in wettability when Hexamoll® DINCH was used, as shown in Fig 4.10. The high wettability makes the formulation less suitable in terms of washability.

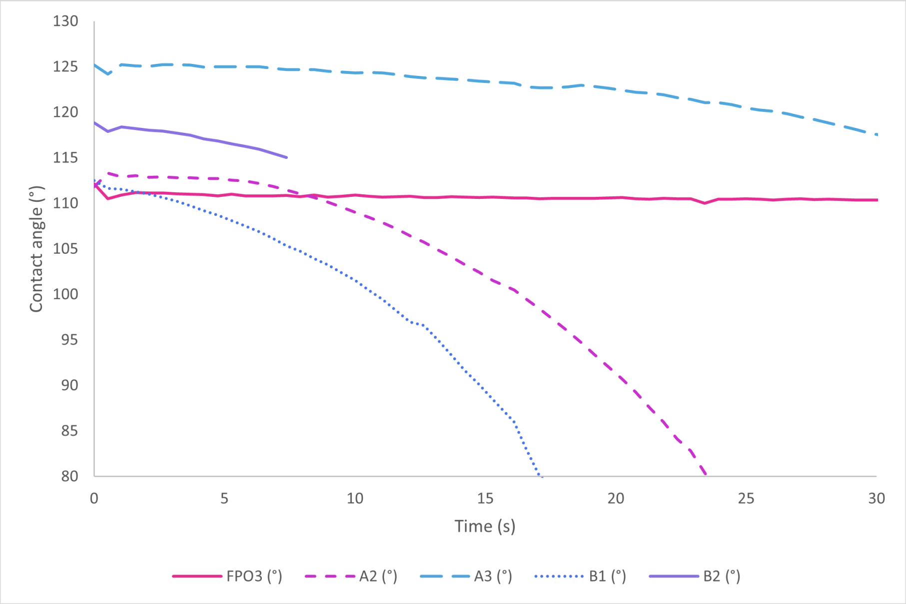


Figure 4.10: Wettability and contact angle measurements of films made of the original formulation and 4 new formulations: A2, A3, B1 and B2.

In summary, the amount of plasticizer did not produce a noticeable effect on the mechanical properties of the thicker films. However, contact angle measurements revealed that increasing the plasticizer content led to higher and undesired wettability. While the incorporation of a hydrophobic plasticizer such as Hexamoll® DINCH improved film forming properties, it also contributed to increased surface wettability, which may be disadvantageous for washability.

Based on these findings, the formulations Glycerol x1 (which is the same as A1) and A3 were identified as the most suitable candidates, offering a favorable balance between film integrity and surface properties for the intended textile printing application.

4.5 Removal solution

To test the removability an experiment according to the description in the methodology was performed. The white film was cured onto a white textile, making it difficult to distinguish the film from the textile. Both the water and the removal solution dissolved the film, however after one hour, both the films emerged in water and the ones emerged in the removal solution had remains of films. The remains on the water-exposed films could not be mechanically removed by a spatula while the films exposed to the removal solution could.

The removal of the water-emerged films shows that the polysaccharide is being exposed to the water, indicating a lack of film formation in the formulation. However, since the water is not enough to remove the film it still shows some promising behavior.

The removal solution appears to increase removability compared to only water, showing promising results, even though complete removal without additional mechanical force would be desired. However, RS03 does not provide full removal of FP03 without mechanical force either.

4.6 Screen printing

Following the evaluation of the formulations, two samples were selected for screen printing: the original formulation FP03 and the modified A3, see Table 4.1 for composition. The printed results are shown in Fig 4.11.

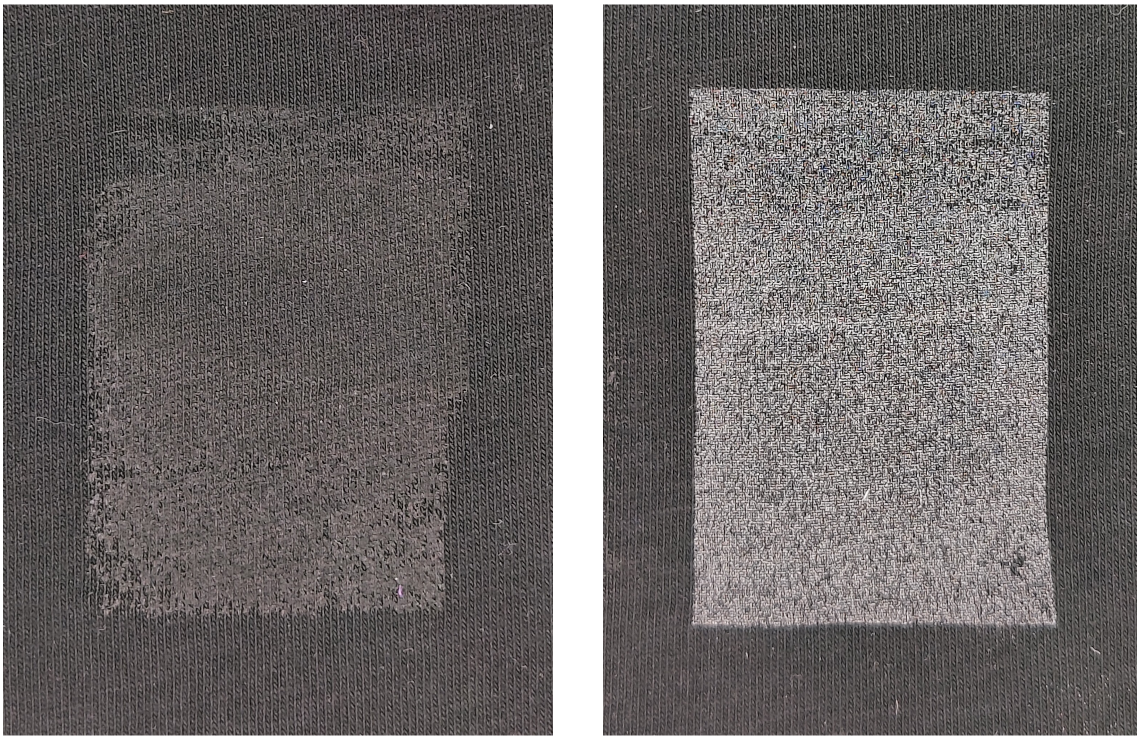


Figure 4.11: Screen printed squares cured on cotton textiles. The original formulation (FP03) is printed to the left and A3 to the right.

A3 demonstrated superior printability. It was the first formulation tested, and it produced a satisfactory print on the initial attempt. In contrast, FP03 proved more difficult to work with, requiring multiple trials to achieve an acceptable print. In addition to improved handling, A3 resulted in prints with sharper edges and more consistent coverage, as clearly observed in Fig 4.11. However, FP03 exhibited higher transparency compared to A3, which can be a desirable property for a priming layer. The increased opacity of A3 is attributed to the presence of PLA particles, which impart a whiter appearance to the formulation as a result of bigger size. If the layer were to be repurposed as a removable ink rather than a priming layer, the improved coverage provided by A3 would be advantageous. Nevertheless, further optimization would be required to achieve the opacity typically needed for high-quality prints.

5

Conclusion

The primary objective of this project was to identify a biobased, film forming binder to replace the currently used oil-based latex binder, without compromising the performance of the priming layer. Performance was assessed based on film formation, flexibility, water resistance, and rheological behavior.

Four different binder alternatives were explored, with the most promising results achieved using a combination of PLA and Surelease® - where Surelease® functioned as the main binder and PLA acted primarily as a filler. While PLA improved the rheological characteristics of the formulation, it also increased the brittleness of the resulting films. Four formulations, varying in their ratios of PLA, Surelease®, and EHM500, and incorporating either glycerol or Hexamoll® DINCH as plasticizers, were prepared and evaluated. Film formation was visually assessed by curing the formulations on a Teflon-mould and textile substrates, and wettability was measured using an optical goniometer.

Among the tested formulations, the one labeled "A3" - which contains glycerol as plasticizer, 25 % PLA, 25 % Surelease®, and 5 % EHM500 relative to the original amount of latex - was selected as the most promising. This formulation underwent additional testing, including removal and screen printing trials. While A3 outperformed the reference formulation FP03 in terms of printability, it did not match its film forming quality. The films produced with A3 were removable using a dedicated removal solution, but only with the application of mechanical force.

5.1 Future outlook

Future work on the formulation should focus on enhancing film forming properties and improving removability. This may involve exploring alternative plasticizers or optimizing the composition of the removal solution to better suit the updated priming system.

Although the newly developed formulation is biobased - meeting the primary objective of the project - it is not biodegradable. While this represents a significant step toward sustainability, the next logical progression would be to identify a biodegradable alternative to PLA, further minimizing environmental impact.

Another promising direction would be to incorporate pigments directly into the priming layer, potentially eliminating the need for a separate layering system. The development of a biobased, removable ink formulation could offer a fully sustainable solution, supporting reuse and recyclability in the textile industry.

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