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Proceeding Paper

Preliminary Modeling Study of a Tape Casting System for Thermoplastic Starch Film Forming †

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Abstract: Thermoplastic starch films (TPS) are an alternative for single-use plastics in packaging. Evaluating large-scale production alternatives that maintain the properties of these bio-based polymers is a crucial factor in understanding their potential industrial use. This preliminary study focuses on testing whether a mathematical model used to predict the drying conditions of ceramic film via tape casting can be adapted to the production of TPS. It also determines the possible drying tape speeds for this type of polymeric film.

Keywords: tape casting; film drying; starch

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1. Introduction

Nowadays, a switch to environmentally friendly raw materials is increasingly necessary, which poses a new challenge for engineering since, in the industry, it becomes necessary to adapt manufacturing processes to the conditions tolerated by these new materials. Such is the case of bioplastics production [1,2], an industry in which some of the raw materials considered as possible substitutes require humidity and temperature conditions that are difficult to adapt in the polymer processing systems existing in the current plastics industry, or else they need pretreatments to acquire the characteristics that allow for their processability, which, at the same time, implies higher costs, such as in the case of thermoplastic starch or TPS [3,4].

There are two general processes for film manufacturing, the wet process or casting and the dry process. The first method consists of pouring a polymer solution onto a substrate and then drying the solvent, a technique also known as solvent casting, often used at the laboratory level. In addition, the second method is based on the thermoplastic properties presented by some biopolymers, such as the extrusion process [5]. Related to the dry method are also processes such as compression molding, injection molding, and blow molding; however, due to costs and characteristics of the final materials, the large-scale processing of TPS is not yet common worldwide, and research is still ongoing to improve both properties and large-scale manufacturing [6].

Therefore, considering that the decomposition temperature of starch (200–220 °C) is lower than the melting temperature (240 °C), which is why it cannot be processed as a polymer in its native state [4], the study of film properties of this biopolymer at the laboratory level generally begins with preparation via the manual casting molding technique, which allows for the use of starch gelatinized with water as a film-forming solution, which is poured into a mold that can be made of glass, acrylic, or Teflon. We would then wait for the solvent to evaporate, finally leaving a film in equilibrium with the humidity of the environment (dry) that can be removed from the mold (Figure 1). With this technique, film formation is favored, avoiding flow problems and the thermal degradation of biopolymer dispersions. Considering that the processing conditions required to transform biopolymers into thermoplastic materials via techniques such as extrusion can affect the properties of some of the components, especially the active ones [5,7], it is practical for experimentation; however, it is not useful in performing larger scale processes, as needed in the industry, since it does not allow for the preparation of film segments of large dimensions and requires long drying times [8]. In addition, some local irregularities are often unavoidable, such as variations in drying speed or final film thickness related to geometric and drying conditions. For these reasons, and due to the long drying time required, this methodology is not suitable for preparing larger films [9,10]. To solve this, solvent methods are used continuously, such as the tape casting method.

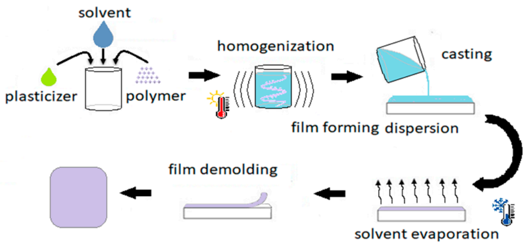

Figure 1. Solvent casting molding.

Although the tape system was developed in 1943 to produce ceramic films, this system has not changed much in recent years and consists of solution preparation equipment and a line of coating machines (Figure 2). The solution preparation equipment is a stand-alone batch operation and includes a thermostated vessel equipped with mixing and feeding systems; the latter consists of a coating applicator, which consists of a nozzle with a guide gate that is adjusted according to the final dry film thickness, usually set in micrometers; the nozzle, in turn, acts as a reservoir for the solution. On the other hand, there is the coating line on which the nozzle rests, which is a drag belt system pulled at a constant speed by a motor, which moves to cause the solution to be dragged between the guide gate and the conveyor belt, the result is a film in solution formed on the belt. This moves through the drying chamber that removes the solvent via evaporation, producing at the end of the process a dry film that is detached from the conveyor belt [5,7]. In recent years, the tape casting system has been considered an alternative for the continuous film production of biodegradable plastics, although there are still few studies on this topic, and despite allowing the formation of a continuous film, it is not common in the conventional plastics industry [6].

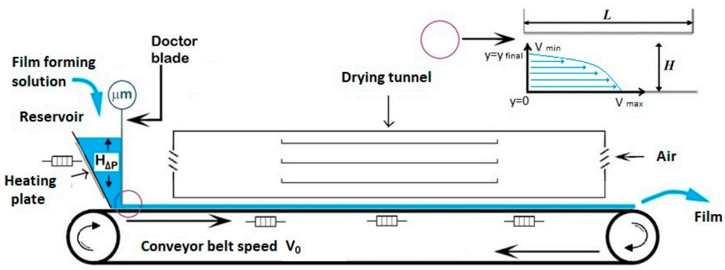

Figure 2. Tape casting system.

Studies have shown that a continuous casting method (blade coating or tape casting) can be used on an industrial scale for bioplastics production because the film-forming suspension is prepared on continuous conveyor belts with effective thickness control. The formed film is dried via heat conduction, convection, or radiation over short periods [11]. With this in mind, the wet method is a coating operation and, as such, pre-existing technologies can be adapted to the production of biopolymer films [5]. The production of edible films via dry methods means productivity and economics. However, considering the severe processing conditions required to transform biopolymers into thermoplastic materials, the properties of some of the components, especially the active ones, may be affected. Therefore, wet processes can offer a moderate and useful way to manufacture edible films, even account for the fact that it is an energy-intensive procedure [11].

When large-scale film preparation is required, it is essential to study the rheological properties and the effect of additives (plasticizers, fillers) on the thermoplastic behavior of film-forming materials in order to select the appropriate processing parameters [11]. Starch gels present thermoplastic behavior, which allows us to process films by applying different thermal and mechanical techniques. However, they present great changes in properties related to the change in starch concentration and its vegetable origin, as well as to the particular characteristics of the added additives. For this reason, specific analyses should be carried out for each type of filmogenic suspension to be processed; among the studies that have been reported for TPS, it is found that according to the rheological study of lecithin and leavening leucocytes, the following stand out: According to the rheological study of le Marcotte et al. (2001) [12], starch gels at different concentrations (4%, 5%, and 6%) present pseudoplastic behavior and absence of thixotropy. They were also reported to retard the sedimentation of solids added to the suspension, which facilitates the inclusion of fillers such as nanoparticles and fibers. On the other hand, differences in the proportion between amylose and amylopectin in starch generate changes in rheological properties, with gels with higher amylose content being more viscous [13], as well having as greater adhesiveness, in addition to presenting changes in thixotropy and pseudoplastic behavior [14]. Implying that a more detailed analysis of this type of gel is necessary when it is desired to scale up the production of films in systems such as tape casting, taking into account the flux pattern of the suspension is the main property related to the formation of good films via this technique [15].

For film formation, according to the needs of the industry, drying times must be taken into account. In addition, despite its frequent use in the food and chemical industry, reports on starch film drying are scarce in the literature; most of these reports refer to convection drying, and this absence is even more evident in studies related to the tape casting technique. We finding very few articles on this subject in the last decade, with most of them being carried out by a single research group.

The drying of starch films depends on several factors; for example, Oliveira de Moraes et al. (2015) [8] and Karapantsios (2006) [16] reported on the influence of temperature and suspension thickness on the conduction drying of starch-based films, considering that the thickness of the spread suspension was the most important variable controlling the properties of the films. De Moraes et al. (2013) used a doctor blade opening of 3-to-4 mm to obtain films with a maximum drying temperature of 60 °C. They found that suspensions of 3-mm thickness can be dried in 2.3 h. Furthermore, in de Moraes et al. (2013) [15], the authors report that the film thickness is always less than the blade spacing and depends on the spreading speed, using conveyor belt movement speeds of approximately 265 cm min and achieving drying times of 5 h, In both articles, shorter drying times are found than those reported in the literature for films prepared via solvent casting (more than 13 h) [17]. Other works, such as Mendes et al. (2020) [18], report that a 1.5 mm-thick layer that was conducted through two stages of oven drying at 90 °C allowed for complete drying. On the other hand, considering the form of heat transfer for film drying, a study by de Moraes and Laurindo (2018) [19] finds that infrared drying, compared to convection drying, as well as the coupling of these, can be a viable procedure for the large-scale production of starch-based films reinforced with cellulosic fibers. The article reports that drying with infrared radiation requires half the time of drying with heat conduction at 60 °C. However, higher infrared heating power may favor crack formation, affecting film properties.

On the other hand, only Elizabeth Gamboni et al. (2021) [5] evaluates the interaction between the conveyor belt material and the starch-based filmogenic solution as a factor in the design of a tape casting equipment. The study reports on the spreadability of the filmogenic solution and its adhesion ability, eventually choosing the polyurethane tape as the most suitable material for its performance and lower cost.

Finally, there is only one article that refers to the modeling of the biofilm production process via tape casting, that of Vogelsang et al. (2014) [7], which describes the preparation of a dextran-based film using water as solvent. The results are compared with a model in which correlations are established between the processing speed and the dry film thickness, which is the variable of interest at the end of the process. The flow is described by a nozzle, which, in its lower part, has a sheet that drags the fluid, forming a velocity profile in which the maximum velocity develops in the drag band, and the velocity in the nozzle is taken as zero assuming that there is no slip in that flow layer.

To develop the conical casting model for pseudoplastic fluids, it is described via the Navier–Stokes equation of motion (1) and the behavior of the viscous fluid with the power law (2), taking into account the solution proposed by Tadmor and Gogos (2006) [20].

$$\rho\left(\frac{\partial V_x}{\partial t}+V_x\frac{\partial V_x}{\partial x}+V_y\frac{\partial V_x}{\partial y}+V_z\frac{\partial V_x}{\partial z}\right)=-\frac{\partial P}{\partial x}-\left(\frac{\partial \tau_{xx}}{\partial x}+\frac{\partial \tau_{xy}}{\partial y}+\frac{\partial \tau_{xy}}{\partial z}\right)+\rho g_x \quad (1)$$

$$\tau_{xy}=k\left(\frac{dv_x}{dy}\right)^n \quad (2)$$

On the other hand, in the work of Tok et al. (2000) [21], a model is used for the processing of ceramics via tape casting, finding the thickness ratio in relation to the conveyor belt speed by performing the non-Newtonian combination of pressure and drag flow model. In this work, the power law equation is also used as a basis for describing the suspended flow, continuing with the coordinates of the Figure 2, where an infinite plane, $x - z$, located at $y = H_0/2$, which presents a maximum fluid velocity $Umax$, is assumed. No-slip conditions are considered, in which the same velocity of motion is presented for both the fluid and the plane. In the above conditions, the fluid presents a static pressure gradient (ΔP). Looking at the way the fluid moves in the reservoir nozzle and on the moving conveyor belt, it is observed that the model in the tape casting system can be described as a flow through parallel plates via Equation (3), where ρ_s and ρ_{tp} are, respectively, the density of the fluid and dry film; L is the channel length; and m and n the power law parameters. The correlation correction factor α is introduced for the reduction of the film extension at the nozzle outlet. In addition, because the thickness between the suspension and the film changes due to the effect of solvent evaporation, the factor β, which considers the correction for mass loss, must also be introduced. Thus, finally, the film thickness corresponds to

$$\delta_{tp}=\left[\frac{2\left(\frac{H_0}{2}\right)^{\frac{1}{n}+2}(\Delta P)^{\frac{1}{n}}}{L\left(\frac{1}{n}+2\right)m^{\frac{1}{n}}U}+\frac{1}{2}(H_0)\right]\frac{\alpha\beta\rho_s}{\rho_{tp}}. \quad (3)$$

Therefore, this preliminary study proposes the modeling of the conditions necessary in order to obtain a given film thickness as a function of the conveyor belt speed in a tape casting system for the production of starch-based bioplastic films, using the model proposed by Tok et al. (2000) for ceramics.

2. Methods

The model of Tok et al. (2000) [21] (Equation (3)) is used to predict the velocities required for starches. To compare the functionality of the model in biopolymers, the results reported by Vogelsang et al. (2014) [7] were used, obtaining the data from the graph reported in the article using Engauge Digitizer software. Velocity increments between 0 to 13 mm/s were used, and the pressure drop in the reservoir (756.25 Pa) was previously determined, using Excel solver to determine the film thickness parameters.

Finally, using the information found for cassava starch with fiber addition reported in Oliveira de Moraes et al. (2015) [8], outlined in Table 1, parameters are proposed for the calculation of speed and nozzle opening conditions in a tape casting system described by Vogelsang et al. (2014) [7] to prepare starch-based film. The values of m (3.545 Pa·s^n) and n (0.604), found in de Moraes et al. (2013) [15] for a temperature of 60 °C, were considered.

Table 1. Experimental data cassava starch film (Oliveira de Moraes et al., 2015) [8]. Temperature: 60 °C; conveyor belt speed: 41.67 mm/s.

Gap (m)	Density (Kg/m^3)	Drying Time (min)	Final Thickness (mm)
0.001	498	36	0.06
0.002	828	52	0.069
0.003	1025	67	0.118
0.004	1190	100	0.148

3. Results and Discussion

3.1. Validation

The experimental data, as well as those in the model, show a lower thickness with higher velocities, assuming then that the speed of the conveyor belt exceeds the flow velocity through the nozzle, which reduces the volume on the belt and, as a consequence, generates a lower thickness; then, the drag flow component becomes the dominant component at high speed. It is found that with the model of Tok et al. (2000) [21], the results found by Vogelsang et al. (2014) [7] can be predicted (Figure 3).

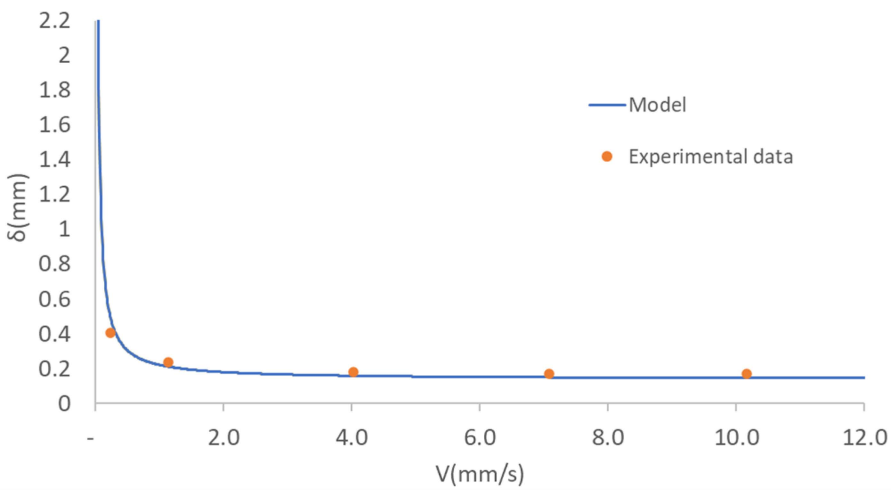

Figure 3. Film thickness vs. conveyor belt speed. Comparison between experimental results obtained by Vogelsang et al. (2014) [7] and theoretical results replicating the model proposed by Tok et al. (2000) [20].

3.2. Prediction of Starch Film Conditions

According to the graph constructed using the model of Tok et al. (2000) [21] (Figure 4), it is inferred that the larger the nozzle opening, the higher the speed required to obtain a thinner film. On the other hand, due to the change in the geometry of the equipment with which the calculations were performed, differences are found with the final thicknesses reported by Oliveira de Moraes et al. (2015) [8]. It is also expected that other conditions, such as drying time, ambient humidity, and length of the conveyor belt, among others, influence the results.

According to the results of the graph, it is expected that a nozzle opening of 3 mm is the most suitable for a starch using a speed of approximately 40 mm/s since it is the one that presents an average thickness, which could be considered acceptable.

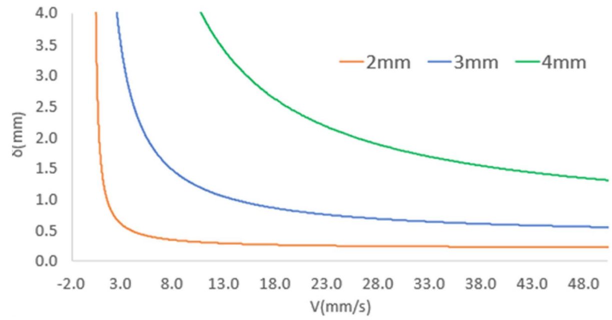

Figure 4. Prediction of cassava starch film thickness vs. conveyor belt speed with different nozzle openings (2 mm, 3 mm, and 4 mm).

4. Conclusions

Models proposed for ceramic materials processed via tape casting can be adapted to bioplastics with the same process.

The modeling of polymer processing via tape casting needs further study; it could present an opportunity to work with materials that require high moisture contents.

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