

State of Knowledge on Starch as an Alternative Solution to Petrochemical Resources—A Review

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Abstract

The use of plastics from petrochemical resources poses environmental impacts, and one of the alternative solutions is the use of starch. The objective of this present work has been to present the literature on starch, and to highlight the debate in the development of composite films. The approach adopted was to present the state of the art on starch and thermoplastic starch matrix composites. The work shows that starch is available worldwide and can be used in the manufacture of biodegradable plastics; the debate remains on the reinforcement of thermoplastic starch to improve its physical and mechanical properties poor; then researchers must diversify the reinforcements to see the impact on the properties of thermoplastic starch.

Keywords

Plastics, Starch, Composite Films, Thermoplastic Starch, Biodegradable

1. Introduction

Plastic is used nowadays in several industries such as food packaging, production of electrical and electronic equipment [1]. Plastic production was 299 million tonnes in 2013 and is expected to increase by 3.9% per year [2]. This is due to the fact that the creation of plastic-based products is very easy thanks to its characteristics of low cost, convenience, lightness and satisfactory elegance which make them very necessary materials. However, there are negative effects on the environment caused by the use of these non-biodegradable plastics generated from petroleum resources such as the depletion of fossil resources, the re-

lease of carbon dioxide, environmental pollution, destruction of the ozone layer, the risk of human health toxicology and the increase in the price of oil [3] [4] [5]. To solve these problems, several avenues have been carried out to explore alternative solutions to materials plastics from petrochemicals. Thermoplastic starch (TPS) has been proved to be the best possible solution. TPS has poor mechanical and water barrier properties and must be reinforced with mineral, organic fillers or fibers [6] [7]. This is how thermoplastic starch (TPS) has been reinforced with Kenaf fibers [8], banana leaves [9], cassava fibers [10], kaolinite [7], pineapple leaf fibers [1] chitosan [11], two- and three-dimensional loads [6]. The objective of this present work was to present the literature on starch and highlight the debate in the development of composite films, in order to facilitate further research in this field.

2. Starch

Very abundant in biomass, starch is a polysaccharide of plant origin constituting the main carbohydrate source of higher plants. It is essential in the diet of living beings and in particular men. It represents a large percentage of dry matter in certain plant reserve organs such as legumes (25% to 50%), tubers (60% to 90%), and cereals (30% to 70%). Half of the world's industrial production of starch is intended for human consumption [12].

Biodegradable, renewable and inexpensive, starch has become in recent years a solution to the problem of plastic packaging, which justifies the interest of the various studies carried out around the world [13] [14] [15]. It is positioned as a potential candidate for the manufacture of biofuels, and is also used in other non-food industrial sectors such as the pharmaceutical industry, the textile industry, the paper industry and the cosmetics industry [16]. Cameroon is experiencing an annual increase in cassava production of 2.7×10^3 tonnes per year [17]. **Figure 1** shows the percentage distribution of raw materials and starches in the world [12].

The production of starch raw material is 89.9 million tons [12], and from **Figure 1(a)**, it can be seen that maize is very available in the world and represents 62.3% of the world production, which justifies why it is very used in terms of starch and represents the 80.5% of the 44.5 million tons produced in the world. **Figure 1** shows that corn, cassava, potato and pea are widely produced in the world as raw material and starch.

2.1. Composition and Structure of Starch

Homopolymer of D-glucose, starch with the chemical formula $(C_6H_{10}O_6)_n$ consists of amylose and amylopectin, which are two polymers with different primary structures, since one is quasi-linear and the other is branched. It also contains components (lipids, minerals, and proteins) whose content varies according to the extraction technology and the botanical origin as shown in **Table 1**.

This table shows that starch consists of 98% - 99% amylose and amylopectin, and 0.1% - 2% of other non-carbohydrate substances.

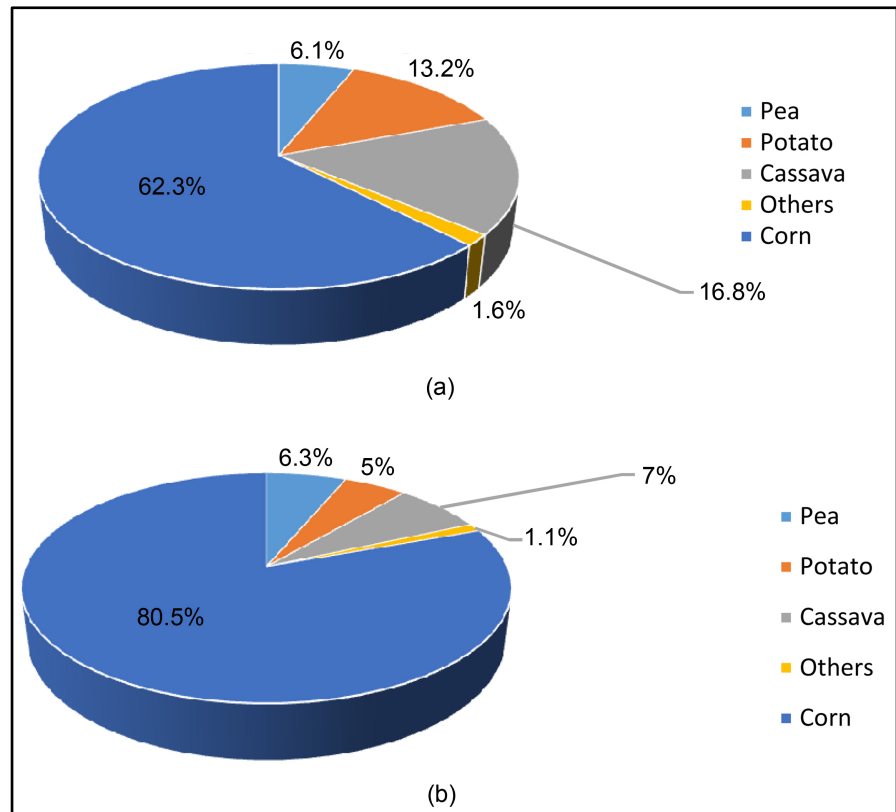


Figure 1. Percentage distribution of raw materials and starches in the world: (a) Segmentation by raw material; (b) Segmentation by starch.

Table 1. Composition of starch in some botanical sources in % of dry matter. [18]

Botanical source	Amyloidosis	Amylopectin	Lipids	Proteins	Minerals	Phosphorus
Standard corn	26 - 28	72	0.63	0.3	0.1	0.015
Wheat	26 - 27	72	0.24	0.33	0.3	0.05
Waxy corn	<1	100	0.23	0.1	0.1	0.003
Standard corn	26 - 28	72	0.63	0.3	0.1	0.015
Cassava	14 - 24	14 - 24	0.1	0.1	0.2	0.01
Pea	33 - 35	33 - 35	0.19	0.18	0.05 - 0.22	0.04
Potato	19 - 23	79	0.09	0.05	0.3	0.04

2.1.1. Amyloidosis

Amyloidosis (**Figure 2**) [12] is a linear polymer composed of D-glucose units, linked by α -(1.4) type bonds. Depending on the botanical origin, native amylose contains 500 to 600 carbohydrate units divided into several chains whose average degree of polymerization is around 500, corresponding to an average molar mass by weight of 10^5 to 10^6 g/mol.

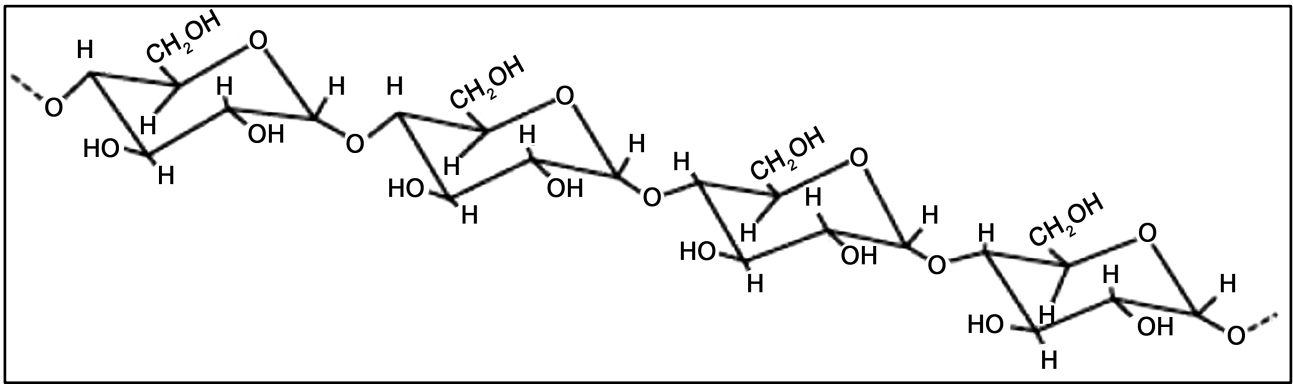


Figure 2. Structure of amylose.

2.1.2. Amylopectin

Amylopectin (**Figure 3**) [19] is a highly branched macromolecule, consisting of short chains of glucose units, linked mainly by α -(1.4) type bonds and 5% to 6% α -(1.6) bonds, responsible for ramifications. Amylopectin has a weight-average molar mass of 10^7 to 10^8 g/mol. Its ramifications and its large molar mass are responsible for the reduction of the polymer chain mobility.

2.2. Morphology of Starch Seeds

Starch size, morphology, amylose and amylopectin content and structural variation are associated with species difference, growth conditions, and environment and plant genetic mutations in plants. Native starch comes in the form of granules. **Figure 4** shows the morphologies of starch granules from a few botanical sources [12].

From the previous figure, it is clear to see that the size and shape of the granules vary depending on the nature of the starch.

2.3. Hydrothermal Behavior of Starch Seeds

Gelatinization or starching corresponds to the transition from a kinetically metastable state where the chains are reorganized. The irreversible swelling and partial solubilization of the starch grain at temperatures above 60°C and in the presence of excess water characterize it. During heating, water molecules penetrate into the amorphous areas of the starch grains causing the grains to swell. This swelling leads to the breaking of the hydrogen bonds in the crystalline zones of the grain and causes the destructuring of the grain. When the heating is prolonged, the hydrogen bonds in the crystalline zones are broken, the amylose diffused out of the grain dissolves in the medium and the grains gradually burst [20] [21]. Gradually the viscosity increases and becomes maximum when the majority of the granules are swollen. During this phase of the hydrothermal treatment, a loss of crystallinity occurs within the grain and is called starching; and the starch paste formed consists of grain ghosts and solubilized macromolecules. Gelatinization is therefore described as the diffusion of amylose chains out of the swollen grain, the amylopectin only appearing in solution if

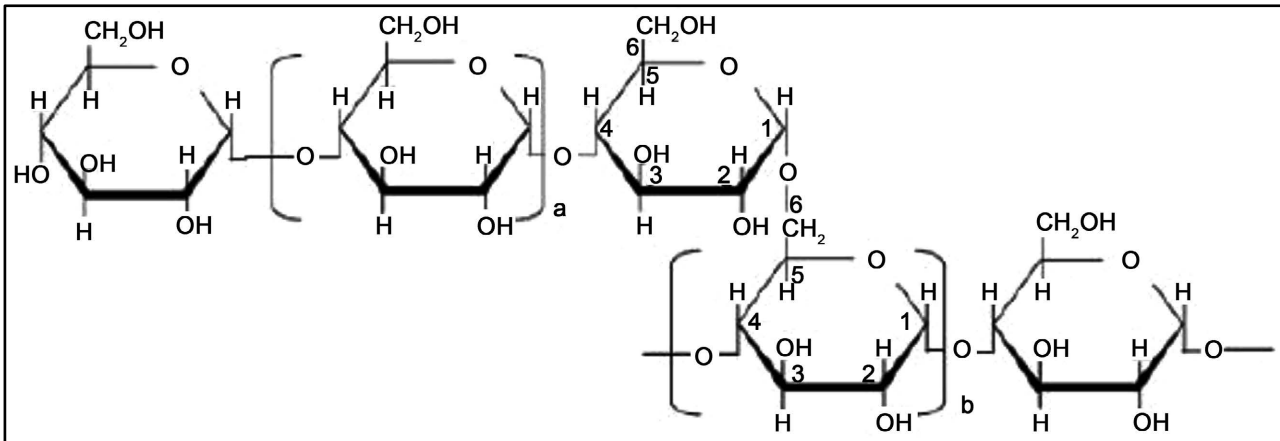


Figure 3. Structure of amylopectin.

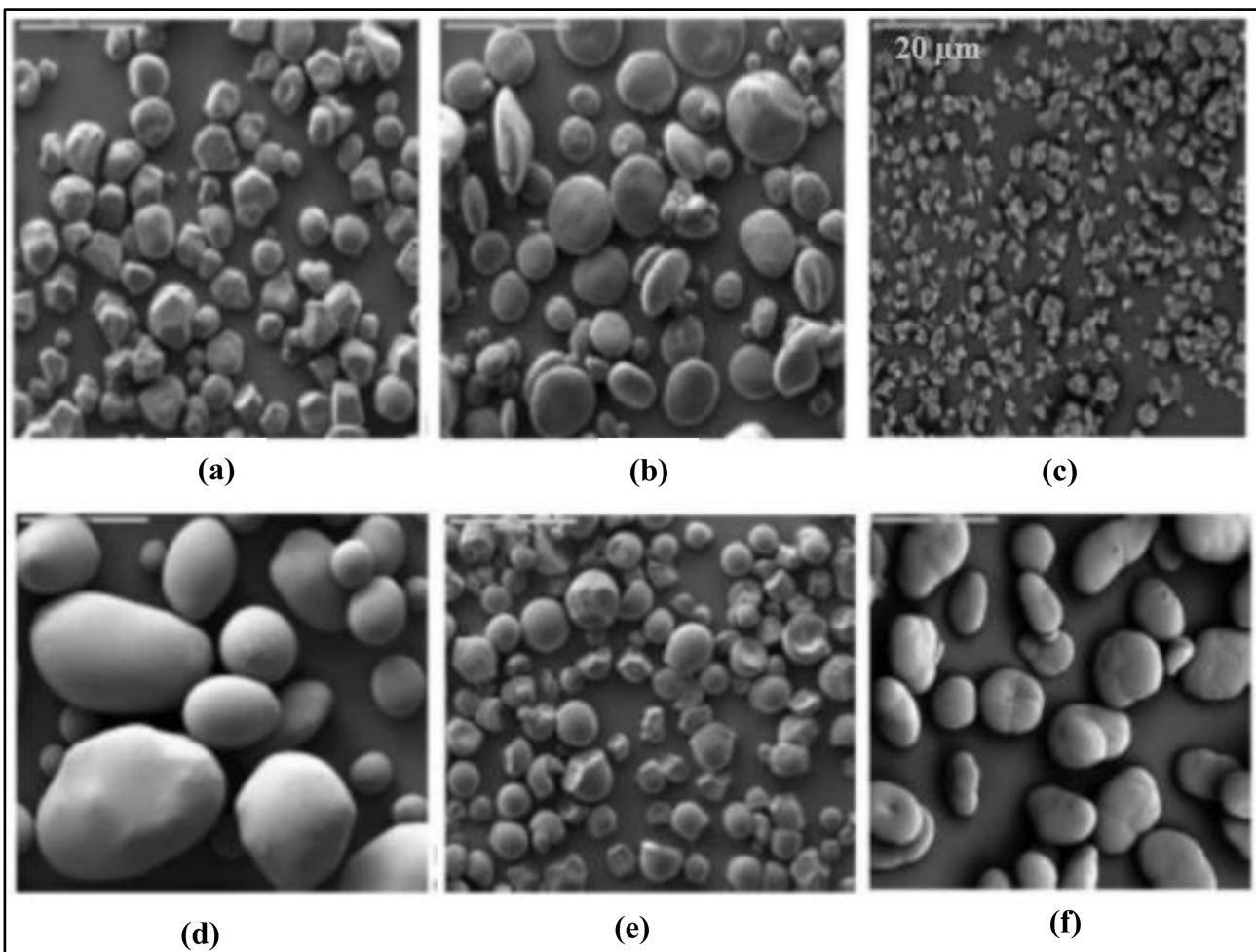


Figure 4. Grains of different starches observed by scanning electron microscope (SEM) (Magnification $\times 280$): (a) Corn; (b) Wheat; (c) Rice; (d) Potato starch; (e) Cassava; (f) Peas.

a mechanical treatment is applied to the swollen grain. This results from the difference in hydrothermal behavior of these two types of chains, linear and branched. [20] [21]

Retro gradation designates the structural reorganization or recrystallization, which takes place during the cooling of a suspension of destructured starch. When the temperature decreases, the gelatinized starch is in an unstable state. Amylose and amylopectin macromolecules naturally tend to recombine and recrystallization occurs step by step. [22]

The glass transition is a reversible phenomenon of state transition, between the hard and relatively brittle form and the molten or rubbery form of an amorphous material. [22]

2.4. Advantages of Starch

In the composites and nanocomposites industry, starch can be used as reinforcing filler and/or as a resin. Because of its biodegradability and renewability, starch is a solution to plastic packaging problems. In the food industry, starch is the main source of energy for food and animal feed; it is an abundant, renewable and inexpensive nutritional compound. With desired functional properties, starch could play a role in quality of various food products and could replace the chemically modified starch that is commonly used in many products as a thickener, gelling agent, or thickeners, gelling agents, binders, sweeteners. In the agronomic industry, the results of studies on starches allow the cultivation of species that produce starch with desired properties. Starch is also used in many other industrial sectors production: pharmaceutical, cosmetic and textile industries. It has also become a potential candidate for the manufacture of biofuels in recent years.

3. Thermoplastic Starch Matrix Composite (TPS)

In its natural state, starch is not a thermoplastic material so it must be plasticized. It is used in the field of biopolymers as a thermoplastic polymer [6] [7] [11] [18]. The transformation of starch into thermoplastic is a process that involves transforming semi-crystalline starch granules into homogeneous materials between macromolecules under the effect of shear and pressure. In this process, in the presence of a plasticizer (sorbitol, glycerol, urea) and under the action of temperature and the effect of shear, new hydrogen bonds between starch and plasticizers are formed by replacing the hydrogen bonds between the starch molecules, we obtain the plasticization of the starch as shown in **Figure 5** [23]. This promotes the flexibility of the starch. Glycerol, glycol, sorbitol, sugars and ethanol amine are the traditional plasticizers, but the main plasticizer used in thermoplastic starch is glycerol [1] [5] [6] [7] [11] [13] [15] [24] [25] [26]. The proportion of plasticizer and its chemical nature strongly influence the physical properties of thermoplastic starch.

Thermoplastic starch when reinforced becomes a composite film. A composite film can be defined as a combination of several immiscible materials of intrinsic characteristics, the mixing of which produces a material of improved performance of small thickness, which can be in the order of less than 3 mm.

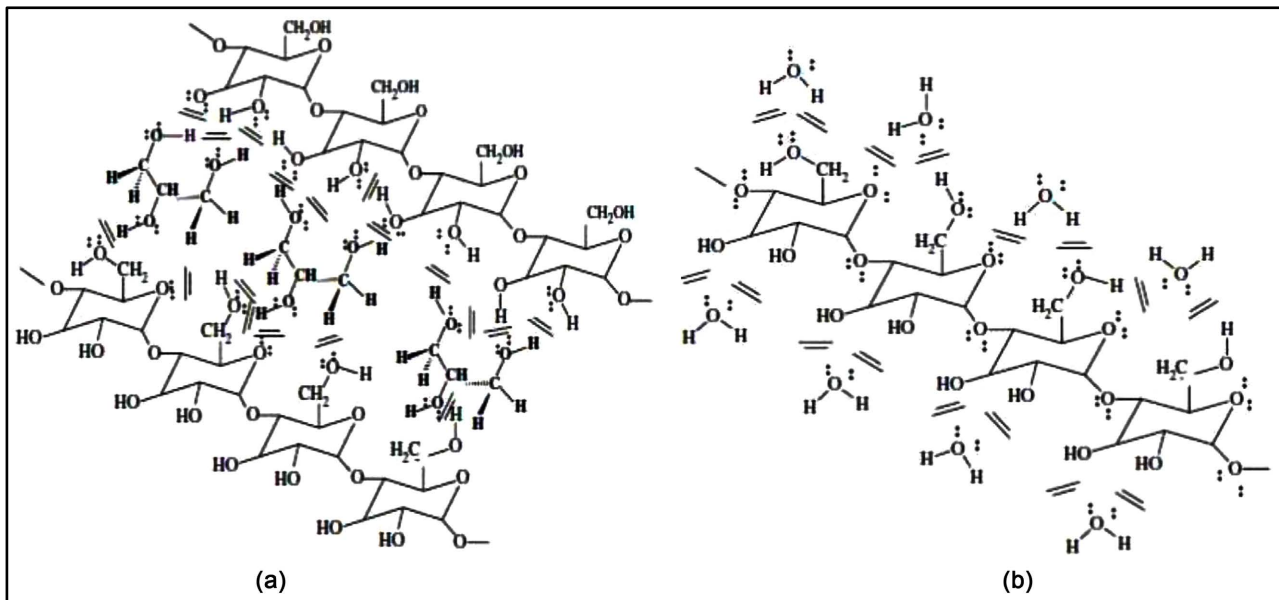


Figure 5. Starch and plasticizer interactions: (a) Starch and glycerol; (b) Starch and water.

Film composite is used nowadays in several industries such as food packaging, production of electrical and electronic equipment [1].

Thermoplastic starch compared to other commonly used plastics has two main disadvantages: poor mechanical properties and high solubility in water [6] [7] [9] [27]. To improve these properties, researchers have used reinforcements such as mineral fillers [7], fibers [23] [26] [27], and organic fillers [15].

Thereafter, it is a question of seeing the influence of plasticizers as well as fiber content, in a thermoplastic starch matrix.

3.1. Influence of the Addition of a Plasticizer

The most commonly used plasticizer in thermoplastic starch is glycerol [6] [7]. However, its addition relative to the weight of the native starch influences the properties of the material.

Belibi 2013 [6] elaborated the thermoplastic starch by adding 10%, 20%, 30% and 40% by weight of glycerol compared to the starch. The results show that, for an addition by weight of 20% relative to the starch, it is impossible to have the mechanical properties, because the material is brittle, and at more than 40%, the mechanical properties decrease.

The addition by weight of glycerol relative to the mass of starch must be between 30 and 40 percent to have good plasticization, this is justified through the percentage of glycerol used in the work [1] [8] [9] [25]-[29].

3.2. Influence of Fibers Addition

The starch matrix and the surface of the natural fibers in combination should lead to composites with good mechanical properties because, having an identical chemical nature (presence of OH group), hydrogen bonds between the matrix

and the fibers are possible.

Jumaindain *et al.* [5] Studied the composites made from the stems of banana leaves, the molding was done by thermo pressing to obtain biofilms 3 mm thick. The results show that for an addition by weight of fibers relative to the TPS matrix of 10%, 20%, 30%, 40%, in tensile testing; the stress at break and the Young's modulus increase with the addition of fibers, the strain at break as for it decreases. The results are similar in most publications, which observe an increase in breaking stress and tensile modulus, and a decrease in breaking strain with the addition of fibers [1] [15] [25] [26] [27] [28].

Edhirej *et al.* [26] Studied composites made from cassava bagasse, molding was done by *casting*. The results show that for an addition by weight of cassava bagasse relative to the mass of starch of 3%, 6% and 9%, the physical parameters such as thickness, water content and rate of water absorption of composites increases with the addition of fibers; on the other hand, the density and the degree of solubility decrease. These results are similar in most publications that make the same observation [1] [8] [25].

It is observed that in the works where the molding is done by thermo pressing, the size of the particles is between 1 mm and 3 mm; in the case of casting, the sizes are lower than 0.6 mm.

3.3. Forming Processes for Thermoplastic Starch Films

To transform starch, the solvent route (by *casting*) and the molten route by extrusion are the two main implementation techniques most used. They differ in particular by the quantity of water used [30].

Figure 6 shows a thermoplastic corn matrix composite reinforced with kenaf fibers obtained by *casting*.



Figure 6. Biocomposite based on maize starch reinforced with kenaf fibers [8].

3.3.1. Solvent Route

The solvent route (*casting*) as shown in **Figure 7** [30], is a technique used to obtain films thanks to the phenomenon of starch gelatinization.

The phenomenon of gelatinization occurs when the starch is subjected to a certain temperature, called the gelatinization temperature, and this is in the presence of an excess quantity of water. It has been shown by a number of authors that this temperature can be between 50°C and 90°C, for a water fraction greater than 0.7. However, it depends on many factors, such as the botanical origin of the starch. [30]

3.3.2. Molten Way

Extrusion, as shown in **Figure 8**, is the most widely used technique for shaping plastic films on an industrial scale, unlike the melting process. In this case, to transform (destructure) the starch into a molten phase, it is necessary to bring thermal energy (heating) but also mechanical energy (shear) under a low humidity rate in the presence of plasticizers. As for the solvent route, during extrusion, the starch grains undergo a certain number of transformation steps in the presence of plasticizers in order to obtain a homogeneous material.

To obtain plasticized starch, the native starch + plasticizers undergo fragmentation then destructuring, then fusion and finally depolymerization.

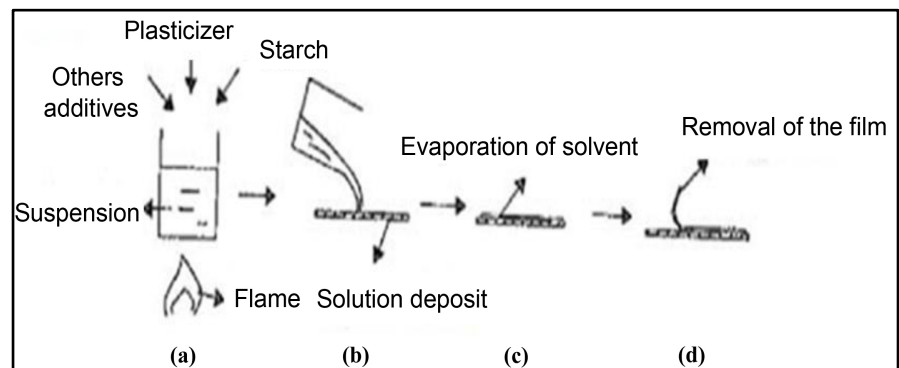


Figure 7. Thermoplastic starch (TPS) obtained by solvent process. (a) Formulation of starch in suspension subjected to thermal effect; (b) Deposition of the solution on a flat platform; (c) Cooling and evaporation of the solvent; (d) Detachment of the film from the support.

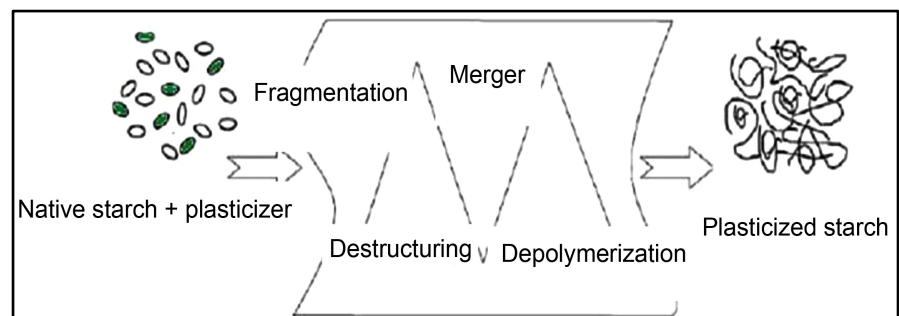


Figure 8. Starch processing steps by extrusion.

3.4. Characterization Techniques

Composites in general in the industrial field have already proven themselves [31] [32] [33] [34] [35]. This can be explained by the determination of the properties of plant fibers composites, by characterizations in compliance with standards as observed in the reference articles [36]-[43]. In general, the structure of bios composite is characterized by X-ray diffraction (XRD) and by transmission electron microscopy (TEM). In certain cases, it is advisable to use simultaneously the diffraction with the great angles and the diffraction with the small angles of the X-rays (SAXRD) in particular when the spacing of the platelets is of the order of 6 to 7 nanometers or when the platelets are rather messy in the case of exfoliated composite bios. Nuclear magnetic resonance (NMR) is also a tool that can help to know the level of exfoliation. Fourier transform infrared (FTIR) and Raman spectroscopy can be used to understand the formation of biocomposite structures through the analysis of interfacial interactions.

The properties of biocomposites are characterized by various techniques. Differential thermal scanning (DSC) and gravimetric (TGA) analysis are used to evaluate melting, crystallization, glass transition, thermal decomposition or barrier effects. In the case of composite bios of polymers, dynamic thermomechanical analysis (DMTA), is a tool of choice in particular for the evaluation of the glass transition temperature and allows accessing Young's modulus.

The following physical parameters are to be determined in the development of thermoplastic starch matrix composites [11] [26] [44] [45]:

- Density;
- The content in water;
- The water and moisture absorption rate;
- solubility in water;
- Water vapour permeability;
- Porosity;
- Biodegradability;
- The thickness.

The determination of the mechanical properties of thermoplastic starch matrix composites depends on the thickness. For thicknesses of less than one millimetre, tensile testing is only possible in accordance with ISO 527-3: 2018 [46]. For thicknesses greater than 1 mm, the bending test and impact resistance are possible.

The SEM will also allow observing the adhesion of the reinforcement matrix.

4. Conclusion

The objective was to present the literature on starch, and highlight the debate on the development of composite films to facilitate further research in this field. It emerges that starch is very available in the world and is the subject of several studies. In the development of bioplastics as solutions to packaging from petrochemicals, researchers are recommended to diversify the reinforcement to improve the properties of thermoplastic starch.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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