Biopolymers for Paperboard Extrusion Coating and Converting

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ABSTRACT

Biopolymers have shown promising options for sustainable packaging applications. This article presents an overview of challenges and opportunities in biopolymers for paperboard extrusion coating and converting processes. Material properties, extrusion coating process and equipment requirements, regulatory compliance, and downstream converting are reviewed. The latest developments and emerging trends in biopolymer technology and innovation are discussed.

INTRODUCTION

In recent years, there has been an increasing demand in the marketplace by brand owners, consumers, Green movements and regulatory bodies for recycling, renewable sourcing and sustainability initiatives. Consumers have become more health and eco conscious. They have also expressed interest in products that are environmentally friendly while offering the same functions and performance as products made from synthetic materials. A move from the linear economy (take-make-waste) to the circular economy (make-use-return) is the key theme for sustainable packaging.

In response to the growing demand for more environmental protection and resource conservation, many companies have been investing in development and manufacture of their products on greater sustainability. Worldwide bioplastics production figures and capacities are difficult to estimate. A recent report published by European Bioplastics shows the rapid growth of the bioplastics industry in the next few years as illustrated in Figure 1 [1]. Bioplastics production capacity is projected to grow by more than 400% from 2014 to 2019, most notably is biobased PET resin, and much lesser in PBS, PBAT, PLA, PE and starch blends.

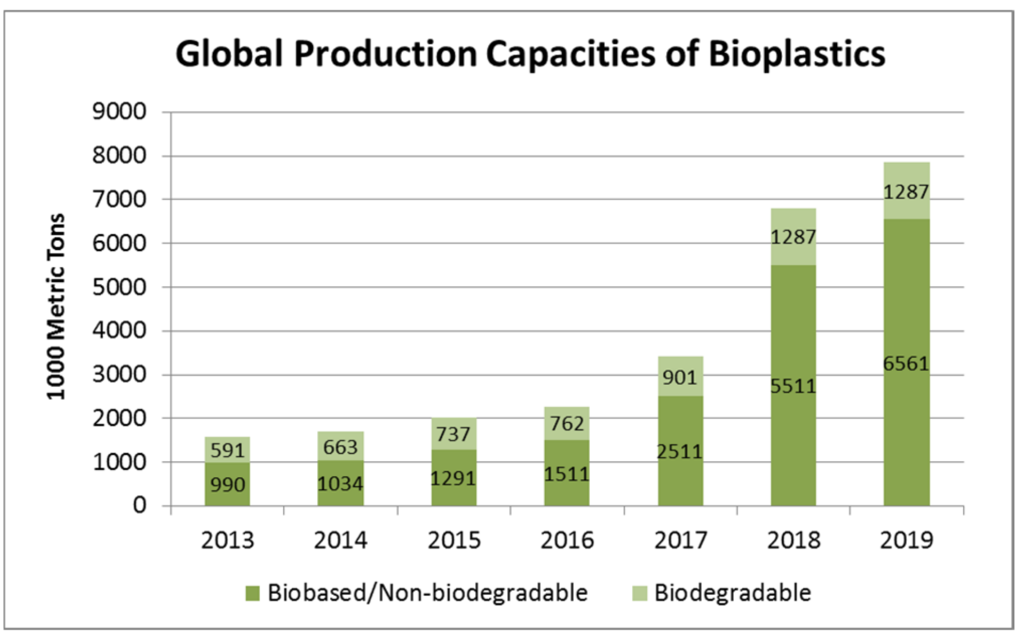

Figure 1: Global Production Capacities of Bioplastics [1]

Evaluation of renewable resource based bioplastics for improving sustainability has been one of the major efforts in green chemistry R&D and packaging development. Replacement of fossil based polymers with biopolymers is an alternate way of reducing carbon footprint and enhancing sustainability profile. Biopolymers are polymers derived from renewable resources as well as biological and fossil-based biodegradable polymers. The market drivers for biopolymers demand come from consumer acceptance, climate change, fossil material dependence, sustainable and new recycling options, properties and functionality.

Paperboard packaging is a very attractive option to address environmental concerns. Wood cellulose fibers are all natural, renewable, biodegradable, compostable, and recyclable. Therefore, utilizing biopolymers along with cellulose substrates in extrusion coated-paper and paperboard products offer an outstanding environmentally friendly and sustainable packaging opportunity in the marketplace. Biobased polymer coated-paper products offer an alternative high value proposition in renewal sourcing and carbon footprint reduction.

CLASSIFICATION OF BIOPOLYMERS

There is quite a lot of confusion and misconceptions in so called degradable, oxo-degradable, biodegradable, marine-degradable, compostable or biobased materials among the general public. United States Federal Trade Commission (FTC) has taken enforcement actions to crack down on certain false and misleading biodegradable plastic claims and unsubstantiated environmental marketing claims [2]. The FTC also released several business and consumer education resources designed to help users understand its Green Guides and environmental marketing.

Biodegradable polymers are certified according to any of the following international standards: ASTM D6400, EN 13432/EN 14995 and ISO 17088. A compostable polymer is a plastic capable of undergoing biological decomposition meeting any of the above standards that also can:

- pass chemical test and disclose all constituents, threshold values for heavy metals,
- disintegrate after 3 months' composting and subsequent sifting through a 2 mm sieve, no more than 10 percent residue may remain, as compared to the original mass,
- biodegrade in controlled composting conditions (oxygen consumption and production of CO_2): Proof must be made that at least 90 percent of the organic material is converted into CO_2 within 6 months,
- pass practical test of compostability in a semi-industrial (or industrial) composting facility with no negative influence on the composting process, and
- pass ecotoxicity test with minimal effect of resultant compost on plant growth (agronomic test).

All compostable polymers are biodegradable but not vice versa.

Biobased products are derived from plants and other renewable agricultural, marine, and forestry materials and provide an alternative to conventional petroleum derived products as defined in the United States Department of Agriculture Biopreferred program [3]. The biobased content of a biopolymer can be determined by the carbon-14 dating method. Carbon-14 has a half-life of 5700 years found in biobased materials but not in fossil fuels. ASTM D6866 and ASTM D7026 provide relevant standards. Table 1 shows the schematic for classification of conventional polymers and biopolymers [4].

Table 1: Classification of Conventional Polymers and Biopolymers

Biodegradable \ Biobased	**Non-Biobased**	**Partially Biobased**	**Biobased**
Biodegradable	PBS, PBSA, PCL, PGA, PVOH	Starch Blends, PLA Blends, PBS, PBAT	PLA, PBS, PHA, PHB, TPS, Starch, Cellulose Acetate
Non-Biodegradable	PE, PP, PET, PBT, PA6, PA66	PBT, PET, PTT, PA6.10	PE, PA11, PA12, PA1010, PEF, PET, PTT

Abbreviations list:

PA	Polyamide
PBAT	Poly(butylene adipate-co-terephthalate)
PBS	Poly(butylene succinate)
PBSA	Poly(butylene succinate-co-adipate)
PBT	Poly(butylene terephthalate)
PCL	Polycaprolactone
PE	Polyethylene
PEF	polyethylene(furanoate)
PET	Polyethylene terephthalate
PGA	Poly(glycolic acid)
PHA	Poly(hydroxyalkanoate)
PHB	Poly(hydroxybutyrate)
PLA	Poly(lactide)
PP	Polypropylene
PTT	Poly(trimethylene terephthalate)
PVOH	Polyvinyl alcohol
TPS	Thermoplastic starch

REQUIREMENTS FOR EXTRUSION, PACKAGING & CONVERTING

Extrusion-based processes are common plastic converting technologies to manufacture packaging materials and products. The three most common extrusion processes and process temperatures are shown in Table 2.

Table 2: Extrusion Process and Processing Temperature

Extrusion Process	Typical Processing Temperature Range
Blown Film	177-232^{o}C (350-450^{o}F)
Cast Film	232-288^{o}C (450-550^{o}F)
Extrusion Coating	288-332^{o}C (550-630^{o}F)

As one can see, the processing temperature varies in a wide range depending on the type of extrusion process, and also the polymers used and end-use applications. Not all biopolymers are available or suitable in every process. However, each extrusion process requires several common desired processing characteristics:

- Good melt strength, low, drawdown or low neck-in
- Wide processing window
- Stable melt film or curtain
- Good gauge or coat weight profile control
- Low or no die lip buildups
- Low or no defects (gels, voids, pinholes, bubbles, etc.)
- Low smoke
- Easy to purge, clean up and change over

Many packaging materials are used in agricultural, consumer, personal care, medical, foods and beverage applications. Functional or barrier properties such as water vapor transmission rate (WVTR), oxygen transmission rate (OTR), oil-and-grease resistance (OGR) and chemical resistance are required.

Since packaging and converting usually involves bending, folding, die-cutting, printing and form-fill-seal (FFS) operations, the biopolymers need to consider mechanical, physical and thermal properties such as tensile strength, elongation & modulus, flexural strength & modulus, impact resistance, abuse resistance, surface characteristics (friction, release and dyne energy level), and heat deflection temperature,

Low seal initiation temperature (SIT) and good hot-tack strength are two key heat sealing capabilities to ensure efficient package converting and secured package integrity. For paperboard extrusion coating process, the extruded molten polymers have to have strong adhesive bonding to the cellulose fibers to provide the necessary functional or barrier properties. Thermal properties are very important since many packaging materials are used in hot and/or cold temperatures.

Table 3 shows a comparison of select biopolymers to conventional extrusion coating polymers.

Table 3: Typical properties of biopolymers vs. conventional extrusion coating polymers [5-10]

Properties \ Polymer	**PLA**	**PHB**	**PBAT**	**PBS**	**PCL**	**LDPE**	**PET**
Density (g/cc.)	1.24	1.25	1.25-1.27	1.26	1.14	0.923	1.4
Melting point (^{o}C)	145-160	164	110-120	115	60	110	250
Glass transition temperature (^{o}C)	55-60	0.0	-30	-32	-60	-125	67-81
Tensile modulus (GPa)	-	1.4	0.08	-	0.4		2-4
Flexural modulus (GPa)	3.6	-	-	0.63	-	0.3	-
Tensile strength (MPa)	62	24-27	35-44	40	-	12	55-75
Elongation at break (%)	3.5	6-9	560-710	120	800-1000	550	60-110
Heat distortion temperature (^{o}C)	55	90	-	90	-		70

CHALLENGES AND OPPORTUNITES IN PAPERBOARD EXTRUSION COATING

Figure 2 shows a schematic of the extrusion coating process. Extrusion coating is a common continuous web process where a polymer melt film is extruded through a flat die and coated on a moving substrate. The discussions in this section relates to extrusion coating of biopolymers onto the paperboard substrate.

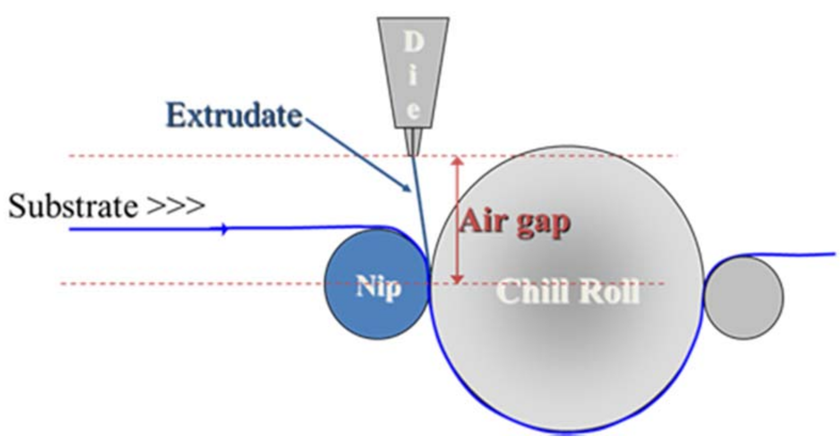

Figure 2: Schematic of extrusion coating process

Unlike plastic film used in flexible packaging, paperboard cannot be melted. Paperboard extrusion coating industry has employed various techniques to enhance adhesion and heat sealing between plastic and paper fiber including high-shear screw, high processing temperature (commonly 316 to 332°C or 600 to 630°F), flame treatment, high air gap, primer application, and ozone oxidation. However, these methods may work for certain polymers and sometimes have adverse processing and property effects. For example, high air gap may increase the time in the air gap to allow for oxidation of hot molten polymer. On the other hand, high air gap also increases the neck-in, reduces the useable web width and trim efficiency, and increases material usage and total manufacturing cost.

Materials

Table 4 shows commercially available polymers for extrusion coating in comparison to fossil based LDPE resin in cost, features, properties and benefits.

Table 4: Extrusion Coating Biopolymers vs. Fossil based LDPE

Features \ Polymer	Fossil LDPE	Bio-LDPE	PLA	(Bio) PBS
Biobased	N	Y	Y	Y*
Commercially Available	Y	Y	Y	Y*
Flexible	Y	Y	N	Y
Heat Sealability	Excellent	Excellent	Fair	Good
FDA Food Contact	Y	Y	Y	Y
Liquid Barrier	Y	Y	Y	Y
Oil Grease Barrier	Y	Y	Y	Y
Industrial Composting	N	N	Y	Y
Home Composting	N	N	N	Y**
Marine Degradable	N	N	N	?

*Bio version in scale-up to commercial supply   ** Limited grade

Polylactide (PLA) has been the most dominant biopolymer in a wide range of innovations, processes, products and applications. Nangeroni et al. [11] provided a very thorough review and teaching about PLA materials for coating application. The inherent brittleness, poor melt strength, low heat deflection temperature (HDT), narrow processing window and low thermal stability present significant technical challenges and limit large-scale applications [10].

High glass transition temperature and high melting point of the semi-crystalline PLA resin (poly-L-lactide or PLLA) limit the heat sealability of PLA based film or coating for packaging and converting. Intensive R&D efforts have been made by ways of copolymerization, plasticization or blending to modify the properties of PLA bioplastics. Some of the useful methods are blending or reactive modification of a base PLA with other biopolymers such as PBAT, PCL, PHA, PBS/PBSA or acrylate copolymer [12-18].

Biobased and biodegradable resins usually command higher levels of premium pricing to fossil based LDPE resin. This is a hurdle for biopolymers and extrusion coated paperboard products to overcome in order to be more competitive, receive wider product adoption and market acceptance.

The end-use applications, brand and marketing messages may determine the type of biopolymer used for the end-of-life sustainability whether it's biodegradable, biobased, repulpable, industrial compostable, home compostable, or marine compostable.

Process

Biopolymers for paperboard extrusion coating are facing the same challenges as fossil based polymers in high processing temperature and related processing characteristics requiring good melt strength, low neck-in, good web curtain and edge stability, uniform cross-machine-direction coat weight distribution.

Linear semi-crystalline PLA resins tend to have poor melt strength and very narrow operating window. Increasing molecular chain branching or altering rheology by reactor polymerization, blending or reactive modification to enhance melt strength and web curtain stability would overcome the majority of processing issues mentioned above.

Poor melt strength exhibited as web break, neck-in, draw resonance and edge weave can limit productivity and increase manufacturing cost as the machine has to operate at slower speed than designed. Small extrusion coating lines run at a speed of 152 to 305 meters per minute (M/min) or 500 to 1000 feet per minute (ft/min). Most extrusion coating lines can run 305 to 457 M/min (1000 to 1500 ft/min). High-speed lines can run 457 to 610 M/min (1500 to 2000 ft/min) or higher. With more neck-in and other extrusion process issues, the extruder may have to run at higher rate to wider deckle and at lower line speed to account for the lost edge trim efficiency and minimize process stability issues. These all translate to additional material usage, reduced efficiency and higher manufacturing cost.

Equipment

Since PLA and some other biopolymers are hydroscopic, pre-drying prior to use is essentially. Good desiccant dryer capability is required especially for the resins used from already-opened package. In-line drying is preferred for continuous operation.

As mentioned earlier, PLA resins have narrow processing window and shear-sensitive rheology. Proper screw design and die design are critical for extrusion efficiency and stability. Excessive shear heating, localized overheating, resin degradation and residual resin hang-up are potential problems. Screw cooling capability in the feed section is desirable to prevent the resin from sticking to the screw root. Smooth barrels are recommended to minimize excess shear heat generation.

Food Contact Compliance

Many plastic based packaging materials are used in food packaging and food service products. Some are used in reheating or cooking conditions. Bioplastics in compliance with relevant food contact regulations, the types of foods and conditions of use among a list of environmental, chemical and substance limitation requirements are critical in order to expand their products and applications.

Under relevant section codes of US FDA Compliance (21 CFR 2013), the resin should be certified to be safely used in articles or components of articles intended. EU, Canada, China and other countries have similar food contact regulations.

US FDA 21 CFR176.170 describes the components of paper and paperboard in contact with aqueous and fatty foods. "Conditions of Use" are referred to the time-temperature conditions used in the test procedure for \determining the amount of extractives from the food-contact surface of uncoated or coated paper and paperboard. Biopolymer supplier has to test and obtain proper conditions of use recommendation for the end-use applications.

Conditions of Use
A. High temperature heat-sterilized (e.g., over 212^{o}F or 100^{o}C)
B. Boiling water sterilized.
C. Hot filled or pasteurized above 150^{o}F (65.5^{o}C)
D. Hot filled or pasteurized below 150^{o}F (65.5^{o}C)
E. Room temperature filled and stored (no thermal treatment in the container).
F. Refrigerated storage (no thermal treatment in the container).
G. Frozen storage (no thermal treatment in the container).
H. Frozen or refrigerated storage: Ready-prepared foods intended to be reheated in container at time of use:
1. Aqueous or oil-in-water emulsion of high- or low-fat.
2. Aqueous, high- or low-free oil or fat.

Development of high heat-resistant biopolymers for ovenable applications has been a big challenge and is one of the emerging opportunities.

Applications

Many poly coated paper products require poly-one-side (P1S) heat sealing to the paper in the converting process while some others have poly-two-side (P2S) face-to-face heat sealing. Similar to form-fill-seal (FFS) operation in flexible packaging converting, a biopolymer having low heat sealing temperature and good hot-tack strength that can provide strong adhesive bonding in poly-to-paper and poly-to-poly interfaces at fast converting speed (i.e., low dwell time) is the ultimate winner.

Some examples of biopolymer-coated paper product applications are shown below Figure 3. Each packaging product has its unique fit-for-use requirements for end-use properties and performance that must be fully evaluated.

- Cups

- Gable Top Carton

- Folding Carton

- Trays/Plates

The shift in trends has created different wants and needs as shown in Figure 4. With urbanization, lifestyle and demographic change as well as health and environmental consciousness, consumers are demanding more convenience on-the-go, fresh, or ready meals in smaller package sizes. Innovation of biobased and biodegradable polymers need to focus on end-use packaging performance in shelf-life extension, freeze-thaw cycle, microwave and ovenable applications.

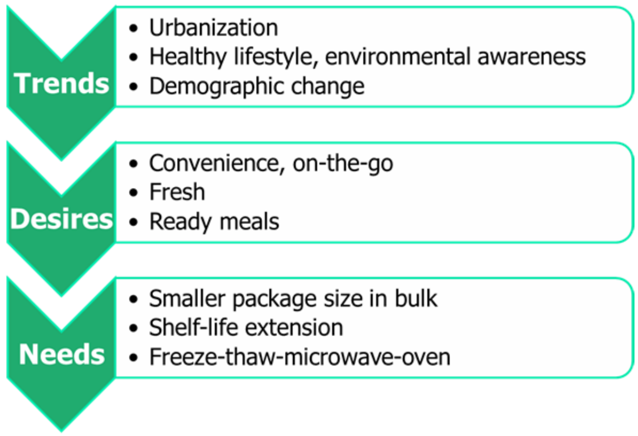

Figure 4: The Shift in Trends.

Additional opportunities in biopolymers for packaging are compostable biopolymers for liquid packaging, moisture and oxygen barrier performance for shelf-life extension, and soil, fresh water or marine biodegrdable.

Sustainability is the new way of living. The bio-eco economy requires collaboration in technology, innovation and markets. The circle of sustainability needs to involve various aspects in:

- Material supply: availability, prices
- Market research: trends, volumes, competitive analysis, feasibility studies
- Economic evaluation: process economics, product cost analysis
- Environmental evaluation: life cycle assessment (LCA), environmental impact
- Policy and strategy: government, non-government organization (NGO), system & framework
- Communication and marketing: information dissemination, conferences, workshops

Figure 5: Eco Economy and Packaging Value Chain

CONCLUSIONS

Biobased and biodegradable polymers for paper extrusion coating and converting are reviewed in this article. Biopolymer extrusion coated-paper and paperboard-based packaging products demonstrate the best viable eco-friendly sustainable packaging solutions from natural, renewable and compostable sources.

The challenges and opportunities in biopolymer material properties, extrusion coating process, equipment and operation, food contact compliance and conditions of use, heat sealing and adhesive bonding in package converting are discussed. Several biopolymer extrusion coated-paperboard products are demonstrated.

Cost of biobased or biodegradable polymers has premium pricing to fossil based materials. With more participants, more consumer demand and capacity expansion, the manufacturing structure is hopefully to enable more cost-efficient production.

Increasing investment in biobased versions of biodegradable materials is moving toward right sustainable trend. Biobased and/or biodegradable polymers having high moisture and oxygen barrier properties to keep foods fresh and extend shelf-life are the new frontiers. Soil, fresh water and marine biodegradable plastics offer solutions to end-of-life disposal options. Development of high heat-resistant biopolymer suitable for ovenable application is a big challenge and an opportunity.

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