

# Film Performance of Poly(lactic acid) Blends for Packaging Applications

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## ABSTRACT

*Poly(lactic acid) (PLA), a biodegradable thermoplastic derived from renewable resources, stands out as a substitute to petroleum-based plastics. PLA based films for food packaging has been an area of both commercial and research interest within the context of sustainability. In spite of its high strength, packaging applications have been limited because PLA is more brittle than traditional oil-based plastics. Because of this, films display low tear and impact resistance and produce a loud crackling sound when manipulated. Although many studies address the toughening of PLA in the bulk, little attention has been placed on the film performance. The present study is aimed at providing a survey of binary PLA based blends with other biodegradable and non-biodegradable plastics. Acrylic impact modifier (AIM, 5 wt. %), ethylene vinyl acetate (EVA, 20 wt.%), polyhydroxyalkanoate (PHA, 10 wt.%), polycaprolactone (PCL, 30 wt.%), polybutylene succinate (PBS, 20 wt.%) and polypropylene carbonate (PPC, 30 wt.%) were each blended with PLA through single-screw extrusion and converted into films via the blown-film process. Tear and impact resistance, heat seal strength, and noise level were measured. EVA, PHA, PCL, and PBS improved the tear resistance with EVA having the highest effect (>2x). Similarly AIM, EVA and PPC improved the resistance of the film to impact-puncture penetration. Heat seal strength was significantly improved by the PHA and moderately increased by AIM (2x) and EVA. Additionally, we proposed a method to quantify the annoyance of the noise made by the films upon manipulation. PCL and PBS significantly reduced the annoyance level of the films.*

**KEY WORDS:** *polylactic acid, films, packaging, noise measurement, blends*

## 1.0 INTRODUCTION

The past decade has witnessed a steady growth for the market of bio-based and biodegradable resins [1], [2]. Plastics are being approached in a more responsible and holistic way, taking into account the sources from which they are derived as well as the ultimate fate of the materials. As the technology evolves, suppliers are able to tailor the polymer to specific applications by controlling the molecular weight and molecular architecture. As a result, new and improved grades of bioplastics resins are continuously being introduced in the market.

Poly(lactic acid) (PLA) is at the forefront of commercially available bioplastics. PLA is derived from renewable resources and biodegradable. PLA based films for food packaging has been an area of both industrial and research interest within the context of sustainability [3]. In spite of its excellent properties, packaging applications have been limited because PLA's brittle behaviour. Because of this, films display low tear and impact resistance, low elongation, and produce a loud crackling sound when manipulated.

Different approaches have been proposed to overcome the brittleness of PLA including copolymerization, plasticization and blending with other polymers [4]. Copolymerization is not economically viable unless produced in a large industrial scale. Plasticization requires large amounts of plasticizers to achieve flexibility that can leach out after the material is manufactured [5]. On the other hand, blending with other polymers is a convenient way to modify the properties. Polyhydroxybutyrate (PHB) [6], Polycaprolactone [7], poly(ethylene-co-vinyl acetate) (EVA) [8], among other resins have been blended with PLA to improve toughness. Ma and coworkers found that the compatibility between PLA and EVA could be adjusted by varying the vinyl acetate (VA) content on EVA. Copolymers with 50 wt.% VA content showed the highest improvement

in elongation at break, over 300% compared with <10% for pure PLA [8].

Although some studies have addressed the toughening of PLA through blending, the sample manufacturing and characterization has focused on thick samples such as dog-bone shape and thick bars for flexural and impact test. Little attention has been placed on the film performance. The present study is aimed at providing a survey of binary PLA based blends with other biodegradable and non-biodegradable plastics with the ultimate goal of identifying blend systems with the potential for property improvement targeting specific applications. The material selection focused on newer resins with high molecular weight intended for thermoplastic applications. The manufactured films were evaluated in terms of tensile properties, tear and impact resistance and heat seal strength. Additionally, noise level produced by the film upon manipulation was recorded and analyzed.

## 2.0 EXPERIMENTAL

### 2.1 Materials

Table 1 lists the materials used in this study as well as the relative blend compositions (Ratio by mass). Blend compositions were selected based on a literature review and suppliers recommendations for the case of AIM. Poly(lactic acid), Ingeo 4043 D, was purchased from Natureworks, LLC. Acrylic impact modifier (AIM), Paraloid BPM 515, was obtained from DOW Chemical Company. Ethylene-vinyl acetate copolymer, Levamelt 500, was supplied by Lanxess. The copolymer contains 50 wt % vinyl acetate. Polyhydroxyalkanoate, M-vera B5008, was supplied by Metabolix. Polycaprolactone (PCL), CAPA FB 100, was kindly donated by Perstorp. CAPA FB 100 has a slightly cross-linked structure with a molecular weight of 100,000 Daltons. Polybutylene succinate (PBS) Bionelle

Table 1: Blend formulations information. MFI measured at 210°C and 2.16 kg

Blends	MFI (g/10min)	Ratio by Mass	Additive Full Name	Target Level
PLA	5.2	100:00		Natureworks 4043D
PLA / AIM	6.1	95:05	Acrylic Impact Modifier	Paraloid BPM-515
PLA / EVA	6.4	80:20	Ethylene co-vinyl acetate	Lanxess Levamelt 500
PLA / PHA	7.5	90:10	Polyhydroxyalkanoate	Metabolix Mirel P5001
PLA / PCL	9.3	70:30	Polycaprolactone	Perstrop CAPA FB 100
PLA / PBS	21.0	80:20	Polybutylene succinate	Showa Bionelle 1001MD
PLA / PPC	5.7	70:30	Polypropylene carbonate	Novomer

1001MD was supplied by Showa Denko America, Inc. Polypropylene carbonate (PPC) was kindly donated by Novomer.

## 2.2 Melt mixing and film manufacturing

Melt mixing of the components was carried out on a single-screw extruder (Wayne Machine & Die Co.) followed by pelletizing. The temperature profile used, from die to feed throat, was 390, 390, 380, 375, 375, 355, and 320 °F. Only for the PLA/EVA blend the profile was adjusted to 335, 335, 330, 310, 280, 250, and 120 °F.

Films were manufactured in a LabTech blown film laboratory line (390 to 360 °F). Blown film conditions (i.e., blow up ratio, rotational screw speed) was adjusted targeting a nominal thickness of 1.5 mils. All the materials were pre-dried before mixing and blown film process.

## 2.3 Testing

Tensile testing of all samples was performed on an Instron 4301, 43K1 tensile tester with 5 kN load cell and pneumatic grips. The initial gauge length was set to five inches and a crosshead speed of 0.5 inch/min was used according to ASTM D 882-02.

Specimens were carefully selected and cut to 1" x 6" strips with the machine direction.

Tear resistance was determined according to ASTM D 1922 standard using an Elmendorf-type tester (200g pendulum). Test specimens in both machine direction and cross direction were cut to 2.5" x 3" with a template such that the direction of the tear would be parallel to the 2.5" side. A machine direction specimen is defined such that the direction of the tear is parallel to the machine direction. Spencer Impact resistance of the films was measured according to ASTM D3420 procedure B on a Thwing-Albert's Protear Elmendorf Tear & Spencer. A 1600 grams pendulum was used for the test.

The potential of using PLA and PLA blends as heat seal layers was investigated by determining the heat seal strength (ASTM F88-06). Fin seals were produced on a Sencorp Bar Heat Sealer 24 AS/1 using temperatures ranging from 120 to 140 °C for 1 second.

PLA films are known for producing a loud noise upon manipulation, which has been proved an undesirable characteristic in packaging applications. Film samples of 9.5 by 9.5 inch squares were

cut for noise characterization. At least four replicates of each sample were tested. Manual manipulation of the films was done in a recording studio at RIT (reverberation time is about 0.3 sec at 500 Hz). The noises from the films have been recorded using a Brüel & Kjær head-and-torso-simulator (HATS) model 4100. This simulator is a special microphone that captures precise psychoacoustic information as a human hears. Among many psychoacoustic attributes, perceived “annoyance” should be the most important attribute to be considered in manufacturing a film since high annoyance may degrade usability of a film by imposing negative impression. In the current analysis, the authors used the annoyance prediction model proposed by Carletti et al. [9]. The annoyance model takes three parameters: peak level, sharpness, and loudness and the MIR (Music Information Retrieval) ToolBox by Lartillot et al. [10] was used to calculate the parameters. The noises were recorded to a digital audio workstation (Pro Tools HD) with sampling frequency of 48 kHz and 24 quantization bit. We chose a 10-seconds long segment having a relatively constant noise pattern from each recorded signal.

## 2.4 Results and Discussion

Table 2 shows the modulus of elasticity, tensile strength, elongation at break and impact resistance of the samples. The thickness in the last column is the average of 10 samples. All the blends decreased the modulus of elasticity but only the changes produced by EVA, PCL and PBS were statistically significant. The results were anticipated because EVA PCL and PBS are flexible and rubberlike (low modulus) imparting flexibility to the blends as the content increase. Similarly, a decrease in tensile strength was observed in the PLA/PCL and PLA/PBS blends.

Only some of the blends showed a marginal increase in the elongation at break (i.e., PLA/PBS and PLA/PPC), which is usually an indicator of toughening. Ma and coworkers prepared PLA/EVA blends with the same composition and VA content and found a significant decrease in the tensile strength and a dramatic increase in the elongation at break (>300%) [8]. The thickness of the samples may have had an effect on the observed elongation when comparing the two studies. The cross section of Ma’s samples was 2 x 0.8 mm compared to 25.4 x 0.03 mm in this study. The results in Table 2

Table 2: Tensile properties of PLA and PLA blends. Groups that have the same letters are not statistically significant ( $p > 0.05$ )

	<b>Tensile Strength (MPa)</b>	<b>Modulus of Elasticity (GPa)</b>	<b>Elongation at Break</b>	<b>Average thickness (mil)</b>
PLA	41.6±5.0 <sup>A</sup>	2.28±0.23 <sup>A</sup>	2.5-3.2%	1.45
PLA / AIM	38.7±6.5 <sup>A</sup>	2.11±0.34 <sup>AB</sup>	3.1-6.4%	1.52
PLA / EVA	37.3±3.3 <sup>AB</sup>	1.87±0.17 <sup>BC</sup>	3.0-12%	1.52
PLA / PHA	42.3±3.7 <sup>A</sup>	2.17±0.12 <sup>A</sup>	3.1-5.2%	1.58
PLA / PCL	32.3±4.4 <sup>B</sup>	1.76±0.17 <sup>C</sup>	4.5-20%	1.89
PLA / PBS	22.5±4.3 <sup>C</sup>	1.09±0.20 <sup>D</sup>	6.4-17.2%	1.27
PLA / PPC	37.5±2.9 <sup>AB</sup>	2.25±0.18 <sup>A</sup>	8.2-24%	2.13

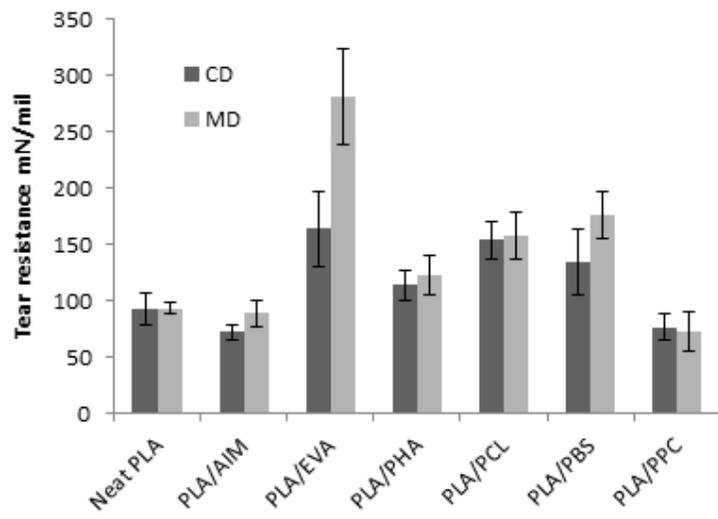


Figure 1: Tear resistance of film samples

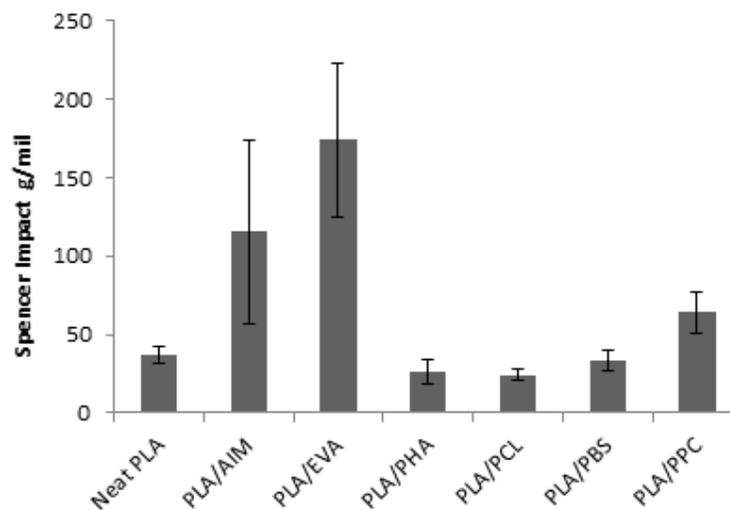


Figure 2: Spencer impact strength of film samples

suggest that the thickness of the samples could play an important role in determining the deformation mechanisms, since the films only had a moderate increase in elongation. One of the objectives of this study is to compare the results obtained for film samples to reported results in thick solid samples. The comparison suggest that the bulk properties usually measured in thick samples may not necessarily reflect the film performance.

Tear resistance measures the energy to propagate a crack in plastics films. Figure 1 shows the tear resistance of the samples. AlM and PPC had a negative effect on the tear resistance while the other samples showed an improvement compared to neat PLA. PLA/EVA showed the highest improvement. This may be due to the dispersed EVA phase because as the crack propagates it encounters the more flexible EVA domains. The elastomeric nature

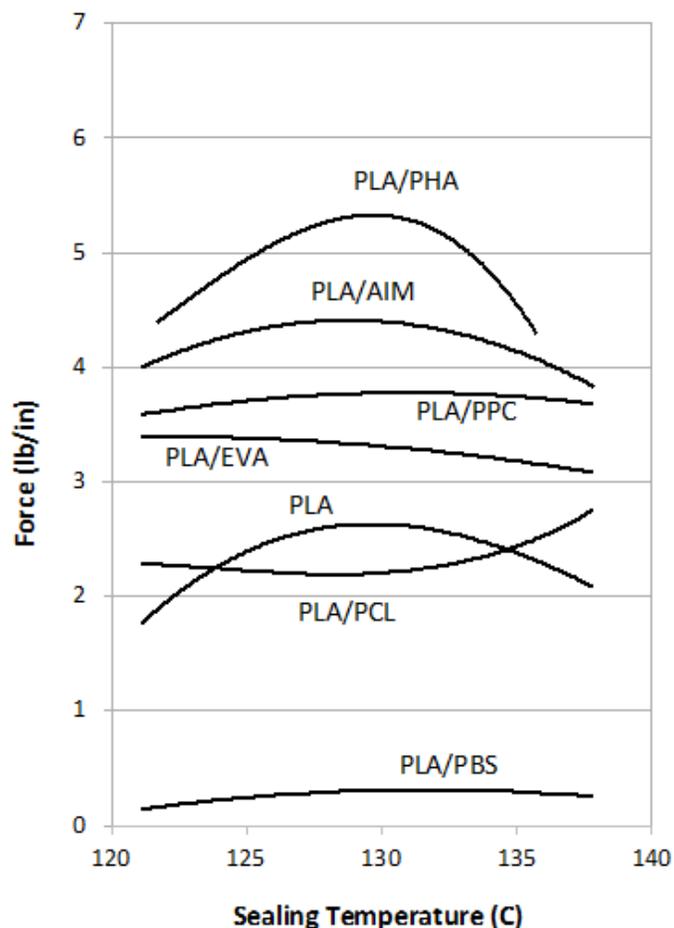


Figure 3: Heat Seal strength of PLA and PLA blends

of the EVA can dissipate more energy, thus increasing the overall tear resistance.

Results shown in Figure 2 display a three and four fold increase in impact resistance for the PLA/EVA and PLA/AIM samples respectively and a slight increase for PPC. Rubber toughening effect was expected in the PCL blend increasing the impact strength. However, the observed decrease in impact resistance in the PLA/PCL blends could be an indication of poor interaction between the PCL and PLA phases. Semba and coworkers found an improvement of 3.5 times in Izod impact strength when using peroxide as compatibilizer imparting the ductile nature of PCL to PLA [11].

The heat sealing ability of the blends was investigated in the range of 120 to 140 °C (see Figure 3). EVA showed a moderate increase in the heat seal strength while PHA and AIM showed a two-fold increase. In PVC formulations, acrylic impact modifiers, similar to AIM used in this study, can lower the melt temperature of the resin and increase fusion times [12]. A similar effect could have taken place when the additive was dispersed in the PLA matrix allowing the interface to melt during heat sealing resulting in higher seal strengths. The blend with PPC showed an increase in heat seal strength slightly higher than EVA exhibiting fused seals in the range studied.

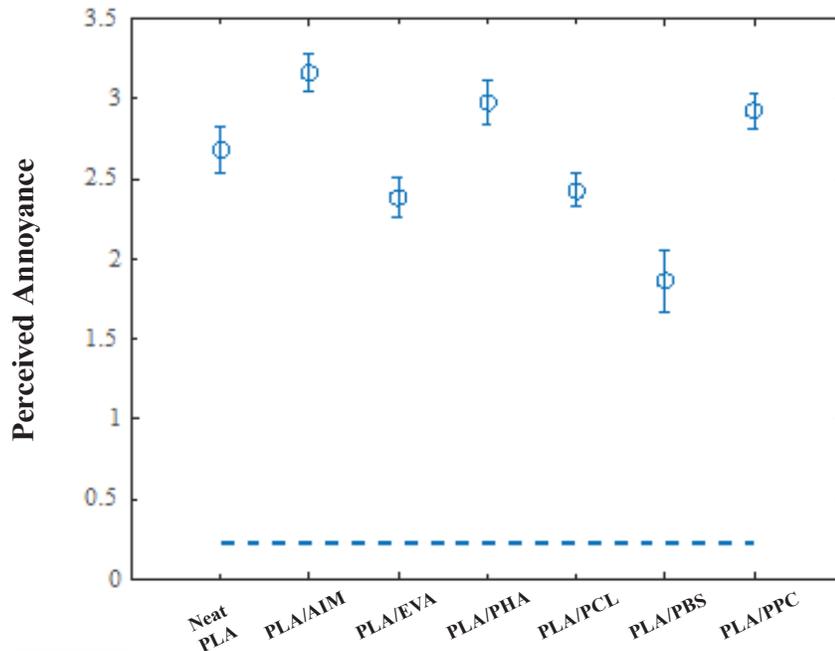


Figure 4: Annoyance values for PLA and PLA blend films. The dashed line represents the RMS level of a reference LDPE bag.

In the case of the PLA/PHA blend, PHA disperses in small domains which remained amorphous at low concentrations [13] facilitating the softening of the material upon heat sealing. The observed increase in seal strength could also be an indication of less crystalline PLA in the blend compared to the films of neat PLA. Potential in using PHA as a dispersed phase to tailor the heat sealing properties of PLA exist.

Since the heat seal was studied in a fixed range, optimum heat seal temperatures of some of the blends may fall out of this range. This may be the case for the blend with EVA. Another interesting observation is that PLA/EVA and PLA/PCL created peelable seals whereas PLA formed fused seals. Therefore, blends with EVA and PLC could be used in packaging applications with easy-open peelable features which have a seal strength of 1 to 2.5 lb/in [14].

One issue that came to the surface upon commercialization of PLA was the noise produced

from the manipulation of the films. Noise became a concern for PLA films since consumers complained how loud chip bags can be. In other words, consumer purchase decisions may be influenced by the noise produced by snack packaging. Perception in multi-sensory and little attention has been placed on noise. PLA films are loud. Part of the reason has to do with the glass transition temperature of PLA which is just above room temperature. The sound has been described as “crispy and crunchy” [15]. Here, we proposed a method to quantify the annoyance of the noise made by films during manipulation.

Figure 4 shows the perceived annoyance values of the films. PBS produced significantly less annoying noise when manually crumpled. The results suggest that the rubbery nature of the dispersed phase, as in the case of EVA, PCL and PBS, had an attenuating effect in the noise produced by the films, with a lower perceived annoyance.

### 3.0 CONCLUSION

This study investigated film properties of PLA and PLA binary blends. Property improvement was observed for the different blends compared to neat PLA in terms of tensile properties, tear and impact resistance, heat seal strength, and noise/annoyance level. Selection of the best blend system will be dependent on specific properties. For instance, the acrylic impact modifier (AIM) improved the impact resistance and heat seal strength but reduced the tear resistance.

PLA blended with EVA showed significant improvement in both tear and impact strength. Potential in using PHA as a dispersed phase to tailor the heat sealing properties of PLA exist. Significant reduction in the annoyance level was achieved in films containing EVA, PCL, and PBS.

The results show the potential of melt blending to improve the film properties of PLA. Further studies should focus on modeling and optimization of particular blend systems. For blends such as PLA/PCL, property improvement could be hindered by poor interaction among the components, thus a compatibilizer should be considered.

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### REFERENCES

- [1] TechNavio, "Global Bioplastics Market 2011-2015," 2012. [Online]. Available: <http://www.technavio.com/report/global-bioplastics-market-2011-2015>. [Accessed: 17-Mar-2014].
- [2] Plastics News, "World bioplastics demand (2005-15)," 2012. [Online]. Available: <http://www.plasticsnews.com/article/20120203/FYI/302039972>. [Accessed: 27-Mar-2014].
- [3] I. Armentano, N. Bitinis, E. Fortunati, S. Mattioli, N. Rescignano, R. Verdejo, M. A. Lopez-Manchado, and J. M. Kenny, "Multifunctional nanostructured PLA materials for packaging and tissue engineering," *Prog. Polym. Sci.*, vol. 38, no. 10–11, pp. 1720–1747, Oct. 2013.
- [4] H. Liu and J. Zhang, "Research progress in toughening modification of poly(lactic acid)," *J. Polym. Sci. Part B Polym. Phys.*, vol. 49, no. 15, pp. 1051–1083, Aug. 2011.
- [5] T. Mekonnen, P. Mussone, H. Khalil, and D. Bressler, "Progress in bio-based plastics and plasticizing modifications," *J. Mater. Chem. A*, vol. 1, no. 43, p. 13379, Oct. 2013.
- [6] M. Zhang and N. Thomas, "Blending polylactic acid with polyhydroxybutyrate: The effect on thermal, mechanical, and biodegradation properties," *Adv. Polym. Technol.*, vol. 30, no. 2, pp. 67–79, 2011.
- [7] C. L. Simões, J. C. Viana, and A. M. Cunha, "Mechanical properties of poly( $\epsilon$ -caprolactone) and poly(lactic acid) blends," *J. Appl. Polym. Sci.*, vol. 112, no. 1, pp. 345–352, Apr. 2009.

- [8] P. Ma, D. G. Hristova-Bogaerds, J. G. P. Goossens, A. B. Spoelstra, Y. Zhang, and P. J. Lemstra, "Toughening of poly(lactic acid) by ethylene-co-vinyl acetate copolymer with different vinyl acetate contents," *Eur. Polym. J.*, vol. 48, no. 1, pp. 146–154, Jan. 2012.
- [9] E. Carletti, F. Pedrielli, and C. Casazza, "Development and validation of a numerical prediction model to estimate the annoyance condition at the operation station of compact loaders.," *Int. J. Occup. Saf. Ergon.*, vol. 17, no. 3, pp. 233–40, Jan. 2011.
- [10] O. Lartillot, P. Toivainen, and T. Eerola, "A Matlab Toolbox for Music Information Retrieval," in *Data Analysis, Machine Learning and Applications SE - 31*, C. Preisach, H. Burkhardt, L. Schmidt-Thieme, and R. Decker, Eds. Springer Berlin Heidelberg, 2008, pp. 261–268.
- [11] T. Semba, K. Kitagawa, U. S. Ishiaku, M. Kotaki, and H. Hamada, "Effect of compounding procedure on mechanical properties and dispersed phase morphology of poly(lactic acid)/ polycaprolactone blends containing peroxide," *J. Appl. Polym. Sci.*, vol. 103, no. 2, pp. 1066–1074, Jan. 2007.
- [12] A. Abu Bakar, A. Hassan, and A. F. Mohd Yusof, "Study of processability of poly(vinyl chloride): Effects of acrylic and chlorinated polyethylene impact modifiers, oil palm empty fruit bunch fiber, and mixing temperature," *J. Vinyl Addit. Technol.*, vol. 14, no. 4, pp. 204–210, Dec. 2008.
- [13] P. Ma, A. B. Spoelstra, P. Schmit, and P. J. Lemstra, "Toughening of poly (lactic acid) by poly ( $\beta$ -hydroxybutyrate-co- $\beta$ -hydroxyvalerate) with high  $\beta$ -hydroxyvalerate content," *Eur. Polym. J.*, vol. 49, no. 6, pp. 1523–1531, Jun. 2013.
- [14] J. Zhang, E. Manias, G. Polizos, J.-Y. Huh, A. Ophir, P. Songtipya, and M. del M. Jimenez-Gasco, "Tailored Polyethylene Nanocomposite Sealants: Broad-Range Peelable Heat-Seals Through Designed Filler/Polymer Interfaces," *J. Adhes. Sci. Technol.*, vol. 23, no. 5, pp. 709–737, Jan. 2009.
- [15] D. Loepp, "The problem with PLA: chip bags that are really loud," *Plastic News*, 2010. [Online]. Available: [www.plasticsnews.com/article/20100818BLOG01/308189996/](http://www.plasticsnews.com/article/20100818BLOG01/308189996/). [Accessed: 01-Dec-2015].