Thermal-oxidative Degradation Of Single-use Paper Cups: PHA Bioplastics As An Alternative to traditional polyethylene film coatings

Hayden Mcgreal

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Abstract

Biopolymers, specifically Polyhydroxyalkanoates (PHAs) and PHA blend biopolymers are emerging as promising alternatives to traditional petroleum-based plastic packaging materials. Paper single-use cups present a challenge for current recycling systems; as a result, most end up in landfills. The conventional plastic films utilized by paper cups accumulate as debris and add to the global plastic waste problem. The desirable properties of PHA blend biopolymers can potentially provide practical solutions to many of the issues faced by traditional plastics. In this study, ThermoGravimetric Analysis (TGA) was performed to quantify the thermo-oxidative degradation characteristics of plastic samples currently present in the market and biopolymers suitable for their substitution. Samples were gathered from restaurants and purchased at a consumer level. PHA/Polylactic Acid (PLA) blend was found to degrade the fastest amongst pure PLA, Polypropylene (PP), and Polyethylene (PE) film found in paper cups. Ultimately, the use of PHA is a significant improvement in terms of both sustainability and degradability to existing market applications.

Introduction

The paper coffee cup has become ubiquitous in the lives of many North Americans; this simple container is touched by millions each day. Market research suggests that single-use hot beverage containers are being produced with an annual compounded growth rate of 1.27%. Currently sitting at 260 billion units per year, the market is expected to reach 280 billion units produced annually by 2027 (iMarc, 2022). Single-use beverage containers are a staple product segment of the food

Toronto Metropolitan University

and beverage industry; their use is and will continue to be widespread across the globe. The sustainability of single-use beverage containers, particularly hot-beverage containers, has been a point of contention for some time. Unlike typical cold-beverage containers, the hot-beverage counterparts are challenging to recycle correctly. The standard structural design of a paper cup consists of a paperboard outer wall with a plastic-coated inner surface. The plastic barrier is typically made of petroleum-based Polyethylene (PE). The nature of this laminated construction demands special processing procedures by both the end user and recycling facilities. As a result, many single-use beverage containers go directly to landfills, where the PE coating cannot be recycled or adequately degraded.

Bioplastics – polymeric materials produced from renewable biomass sources, are emerging as a sustainable alternative to the traditional PE coatings of single-use beverage containers. Polylactic acid (PLA) has been adopted as an alternative material, with substantial improvement in sustainable disposal, and suitable implementation of container properties like insulative capacity and structural stability. Polyhydroxyalkanoate (PHA) type bio-polymers are another alluring alternative due to their biocompatibility and superior biodegradability. However, no market options are currently available for PHA-based single-use beverage containers (Triantafillopoulos et al., 2020). There is insufficient data for the use of PHA, and PHA blend biopolymers as an effective packaging film compared to traditional plastics. Consumers often perceive paper cups as more eco-friendly than plastic-based constructions; this can be attributed to extensive marketing practices. Greenwashing is a term used to identify the deliberate approach of marketing and product positioning as environmentally positive or eco- friendly, regardless of the actual status of the product itself (de Freitas Netto et al., 2020). Greenwashing can be a deliberate act by an individual company or in the case of paper cups, a colloquial understanding established by larger marketing trends. As PHA and PHA blend bioplastics emerge in the packaging industry, do the properties of these polymers provide a significant advancement in degradability and sustainability when used in packaging applications, specifically single-use beverage containers?

Thermogravimetric Analysis (TGA) can be utilized to quantify the degradation process. TGA is a technique used to produce data on the thermal stability and presence of volatile components in a material by observing the changes in mass as the sample is exposed to controlled heat treatment (Loganathan et al., 2017). TGA is a powerful method for understanding the composition of materials for the rate at which thermal decomposition occurs. TGA alone is insufficient for determining the characterization and identity of individual components present in a material. However, the analysis can give valuable insight into a material's total overall degradation curve. This paper seeks to identify the differences in the degradation process between traditional single-use beverage packaging materials and PHA/PLA blend polymer substitutes.

Literature review

The use of biopolymer substitutes for petroleum-based plastics as a packaging material is still in its infancy. Bioplastics have many attractive properties in both their material qualities and their marketing advantages. Bioplastics also have a greater perception of eco-friendliness than traditional plastics, though production can still be taxing on the environment (Álvarez- Chávez et al., 2012). The sustainability of bioplastics needs to be examined with full consideration of the material's entire lifecycle. There is little literature exploring the use of PHA bioplastics in packaging applications. Typically, research conducted on degradation in plastics involves several advanced testing procedures or those which allow for the precise identification of individual components that make up the complete chemical profile of the polymer. Insight into the overall sustainability of bioplastics, in general, is substantial. There is significant evidence pointing towards biopolymers like PLA and PHA as potentially desirable replacements for the PE coatings found in single-use beverage containers; however, focused research on PHA is still currently being established.

Research from Changwichan and Gheewala's (2019) comparative life cycle assessment for petroleum-based plastic, bioplastic, and reusable stainless steel containers was conducted. Overall it was found that reusable stainless steel containers presented the most effective option in terms of reducing environmental impacts from single-use beverage containers. Bioplastic (PLA) cups were found to have a notably better environmental profile than traditional plastics derived from non-sustainable sources. However, significant economic and infrastructural influences currently make bioplastics undesirable from a cost perspective. The production cost of PLA cups was found to be 2-3 times higher than traditional means, making the economic decision to adopt the alternative material not clearly as advantageous. Additionally, the inappropriate processing and disposal of PLA were found to potentially release methane into the atmosphere, an environmentally hazardous consequence that contributes to climate change. Changwichan and Gheewala (2019) concluded that bioplastics are a better choice for beverage packaging than current market options. However, the implementation of said alternatives requires significant oversight regarding the proper management and disposal of the material.

PLA especially can release significant levels of methane without appropriate processing. It was also noted that recycling procedures for traditional plastics, when operating as intended, can significantly reduce the pressure on the creation of new feedstock for conventional plastics. (Changwichan and Gheewala, 2019). These findings are significant as they present relative evidence of bioplastics, specifically PLA, as being of notable improvement to overall sustainability compared to petroleum-based plastics like PP or polyethylene terephthalate (PET). This research also highlights the setbacks of bioplastic applications in today's market. The current infrastructure for recycling and waste management is curated to petroleum-based plastics. Proper processing procedures must be established if bioplastics are to produce a favorable environmental impact.

Álvarez-Chávez et al. (2012) examined the overall sustainability of bioplastics across their entire lifecycle, emphasizing the flow of materials in the production of biopolymer substitutes. Bioplastics were found to be overall more environmentally friendly than their petroleum-based counterparts; however, when examined across their entire lifecycle, none of the contemporary bioplastics were found to be fully sustainable. The production processes of biopolymers were commonly found to either produce toxic chemical byproducts or utilize them throughout their production. Many biopolymers also integrated non-renewable co-polymers in their manufacturing process, further limiting sustainability. It should be noted that a consensus definition for fully sustainable bio-based plastics does not exist. Instead, collective notions of sustainability are agreed upon among stakeholders. Typically, removing all non-renewable and environmentally degrading processes and inputs from the manufacture of bio-based plastic will qualify it as sustainable. Álvarez-Chávez et al. (2012) ultimately finds that there are several barriers to the sustainability of bioplastics as they currently exist. The unsustainable and environmentally taxing means of producing feedstock for plastic production, the use of hazardous chemicals and additives throughout the manufacturing process, and the general lack of infrastructure for the industrial compost or disposal of biopolymers, are all key concerns. The use of hazardous chemicals in PLA production is of note; those used to refine or regulate the production processes of PLA can affect the environmental impact of the end product. Though the resulting material is more environmentally friendly than petroleum-based plastics, parts of the production process can still be optimized. The novelty of PLA/PHA blend substitutes lies in the composition of the material as a fully bio-composed polymer. Rather than a partial mixture of PLA and some other petroleum-based polymer, PLA/PHA is fully biodegradable and composed solely of biomaterial. Its resulting sustainability profile differs from those studied in Álvarez-Chávez et al. (2012).

Choi et al. (2018) investigated the carbon footprint of PLA and PLA blend biopolymers, with particular attention to the resulting Greenhouse Gas (GHG) emissions and Global Warming Potential (GWP) when compared to traditional petroleum-based plastics. The research found that PLA blend plastics have a high index of GWP when disposed of via incineration. PLA films sent to landfills were favorable over incineration, though inadequacies in local waste management streams make the improper disposal of PLA and PLA-blend packaging likely. PLA was shown to have a comparable GWP to some traditional plastics in waste management processes, but the composition of the polymer blend greatly affected the resulting index. Choi et al. (2018) focused their research on three waste management streams: landfill, Incineration, and recycling; with the PLA and PLA blend packaging unable to be recycled, they are considered for incineration and landfill only. As seen in

Changwichan and Gheewala (2019) and Álvarez- Chávez et al. (2012), it seems that bioplastics may be favorable in terms of their overall environmental impact when special care is taken to adapt existing waste management streams. However, existing processes render some potential environmental gains mute, with the improper handling and disposal of biopolymer and biopolymer blend plastics. Choi et al. (2018) ultimately find that in some cases biopolymer blends can be more inimical to the environment in terms of emissions. These findings are notable, with the distinction that different polymer combinations have vastly different resulting environmental profiles. PLA/PHA is a full bioplastic blend with no traditional filler; this composition would likely deviate from the results of Choi et al. (2018).

In research by Lomonaco et al. (2020), the release of Volatile Organic Compounds (VOCs) from discarded plastic debris is examined, specifically via the photodegradation process. This study only examines the characterization of VOCs emitted from traditional petroleum-based plastics, which make up most waste debris today. It is found that plastic debris releases harmful VOCs as it degrades, varying across different polymer classifications and blends. PE and PP were both found to release toxic VOCs, confirming that the large masses of plastic debris accumulating constitutes a valid air quality concern apart from the more obvious concern of litter. This issue is prominent for traditional plastics across their respective lifecycles; however, substantial research remains to be conducted as to the degree to which these findings apply to bioplastics. In a review by Cabanes et al. (2020) the different VOC profiles of various plastics were examined. It was found that post-consumer recycled and discarded plastics often contain a large amount of absorbed VOCs as opposed to virgin samples of the same polymer. The focus of this review and many other studies on VOC emission from plastic waste is concerning the stripping of these supplemental VOCs in the recycling process. Insight such as that from Lomonaco et al. (2020) is scarce in literature for traditional plastics and even more scarce for alternatives. Pollutants emitted from landfill waste is a crucial point of interest when examining bioplastics which are more than likely to end up in landfills. Understanding the comprehensive environmental impact of these emerging bio-based alternatives can determine the most appropriate focus allocation for the market and municipal waste management systems. Contingent on the overall implications of bioplastic replacements, efforts to improve recycling systems for existing plastic debris may prove more fruitful in terms of overall environmental impact.

There is a gap in the literature regarding the degree to which biopolymers, specifically PHA-type polymers, improve upon traditional PE coatings in terms of degradability and overall sustainable impact. PLA and PLA/Petro-based blend substitutes have made it to market, though the lack of catered infrastructure presents challenges to the actual sustainability of the products. PE and PP plastics have been observed to negatively impact VOC emissions, though the same can be said of some PLA blend plastics. The current PE films used in single-use beverage containers cannot be recycled correctly in many waste management systems; the focus of bioplastic alternatives is to reduce debris left as the paperboard exterior degrades. A comprehensive understanding of the total impact of bioplastics will either make their use in containers more desirable or, conversely, the focus on enhanced recycling systems more advantageous; the research remains to be seen. Concerning the comparison of existing single - use beverage containers and potential bioplastic alternatives, this paper seeks to determ ine the degree to which emerging PHA alternatives may improve these materials' sustainability.

Tested Materials

Methodology & Materials

The bioplastic samples selected for this experiment are derived from 3D-printing filament; each is available on the North-American market and is commonly used in additive manufacturing. Two types of bioplastic have been selected. The first was a pure PLA sample manually cut from a spool of 3D-printing filament. The sample was suitable for extrusion and intended for use in additive manufacturing applications. Relevant technical data was listed as (Melt Temperature - >157ºC, Density - 1.24 g/cm^3). The second was a PHA/PLA blend sample, which was also manually cut from a spool of 3D-printing filament. The applications of this sample are similar to those of the pure PLA; relevant technical data was listed as (Melt Temperature - >155ºC, Density - 1.24 g/cm^3). The precise chemical composition of each of the bioplastic samples is unknown, though they are both assumed to represent typical formulations. See Table 1 for sample tabulations.

The samples of single-use beverage containers for this experiment are assumed to be representative of market standards. The first sample consisted of one unused hot-beverage paperboard cup obtained from a Tim Hortons restaurant in March of 2022. "Tim Hortons'' is Tim Hortons Inc., the Canadian multinational food chain. The second sample was a solid PP cold- beverage container obtained from a Tim Hortons restaurant in March of 2022. The plastic lini ng of the hot beverage sample was assumed to be PE, with the outer layer being made up of paperboard. According to industry standards, the plastic barrier was typically made of petroleum-based Polyethylene (PE). The paperboard component of the cup was typically of high basis weight, usually 150 - 350 g/m^2, while the PE film typically measured at 8 - 20 g/m^2 with a thickness of approximately 50µm (Triantafillopoulos et al., 2020). These assumptions are visually consistent with the sample. *See Figure 1; Table 1*

***Figure 1.** Photographs of sample materials submitted for thermogravimetric analysis.*

Sample	Thickness (mm)	Unit weight (mg)
PLA	1.75	10.469092
PLA/PHA	1.75	8.330923
Paper Cup	0.45	5.532684
Plastic Cup	0.3	6.327195

***Table 1.** Sample dimensions.*

Experimental Set-Up

The experimentation was carried out with quantitative analysis as the objective. Small sample units were extracted from the initial sample materials, weighing ~>10 mg and measured approximately 1 cm in length. Each sample was tested individually through TGA, and the resulting data was analyzed to determine the % weight thermal-oxidative degradation curve. The samples were then compared against each other to determine an index for the thermal- oxidative degradation characteristics amongst the selected bioplastics and gathered petroleum - based plastics that are currently in use across the market. Since TGA is a destructive analysis method, the samples have no physical condition after testing. Inputs for the experiment are the plastic and bioplastic samples, and the outputs are the TGA data. All testing was carried out in the Sustainable Packaging Research lab at Toronto Metropolitan University.

Key assumptions in this experiment are listed as follows.

- The PLA and PLA/PHA samples are representative of formulations suitable for film applications and, by extension, the current market applications.
- The gathered packaging samples are representative of packaging constructions in the larger market as a whole.
- The measured thermal-oxidative curve is assumed to be replicable and consistent with established findings.

The main limitation of this experiment was the scope of TGA. With TGA as the sole testing procedure carried out, it was only possible to determine the % weight thermal-oxidative degradation curve of a sample; there was no way to independently verify any potential filler materials or the chemical characteristics of the sample. This limitation made substantive analysis difficult as the degree to which each sample's fillers and unique chemical formulations impact the degradation curve was unknown.

Thermogravimetric Analysis

TGA was conducted using a Perkin Elmer TGA 4000 apparatus under an air atmosphere. Measurements were performed by increasing the temperature from 30°C up to 600°C at 20°C/min. Graphing and calculations were conducted knowing the weight (W) of the material before (W0), and after degradation (W); with constant data of weight against temperature, it was possible to obtain the weight loss (WL(%)).

$$WL(\%) = [(W)/W0) \times 100]$$

The TGA provided weight data for graphing, which was later examined. An understanding of the sample material's overall degradation time could be extrapolated from the curve. With this analysis, an understanding of the degradability and, therefore, suitability as landfill waste was determined. Due to the precision and stability of TGA, multiple samples need not be submitted to achieve replicable results. One single sample unit was submitted for each sample material for this experiment.

Results

Thermogravimetric Analysis provided a weight percentage thermal-oxidative degradation curve for each sample material; Figures 2-6.

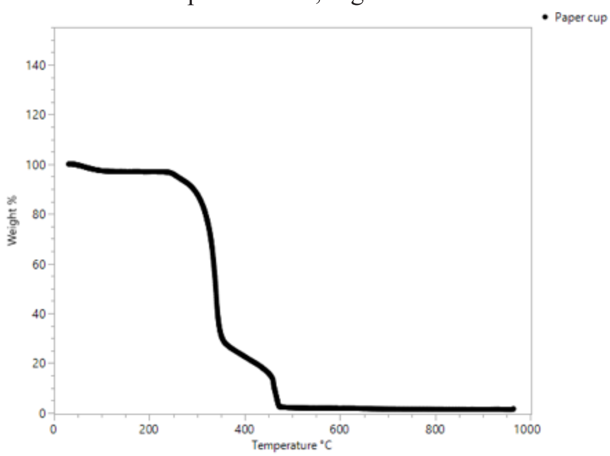

Figure 2. *Thermal-oxidative degradation for paper cup samples under air atmosphere. (20°C / min)*

The paper cup was first tested as a cut sample comprising the paperboard and PE layers. The data contains a steep initial drop in weight % starting around 250°C and plateauing around 380°C. The initial drop was presumed to be the incineration of the paper board layer, with subsequent drops making up the degradation of the PE film and any additives. The sample fully degraded at approximately 500°C.

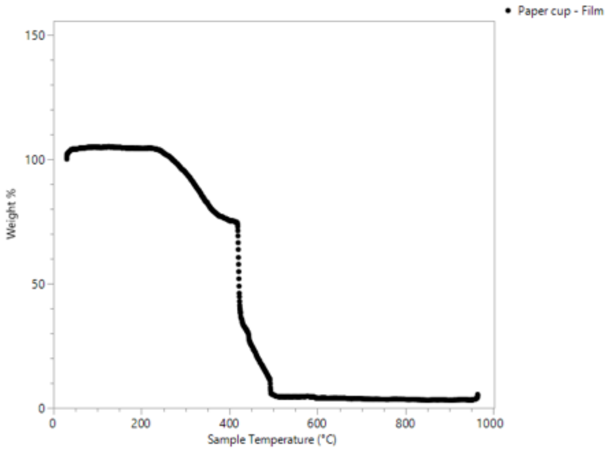

Figure 3. *The curve of thermal-oxidative degradation for film samples under air atmosphere. (20°C / min)*

The paper cup was tested again, with the paperboard portion of the sample removed. The resulting graph comprises the degradation curve of the isolated PE film. With the absence of the paperboard, the graphs of the isolated and non-isolated curves gave a clear distinction of the degradation curve for the PE film alone. This second test confirmed the preliminary analysis of the first test, with the second portion of the curve replicated. The isolated sample curve and the previous unisolated sample were conclusive with other TGA tests conducted on paper hot- beverage containers (Biswal et al., 2013).

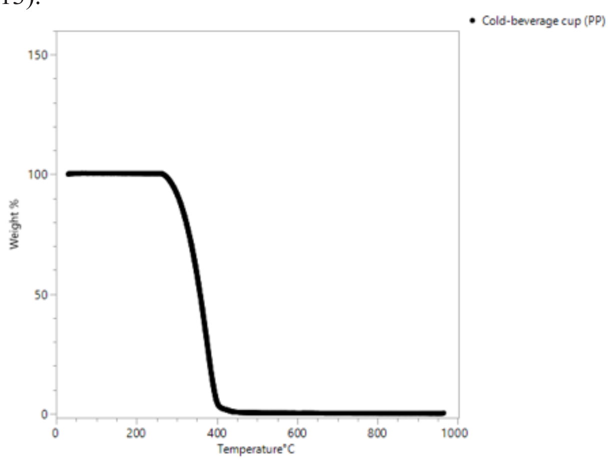

Figure 4. *Thermal-oxidative degradation for Cold-beverage cup samples under air atmosphere. (20°C /min)*

The cold-beverage plastic cup was tested. The graphed data contained a straightforward curve, which fell at approximately 280°C and completely degraded at approximatel y 400°C. This was consistent with a formulation that has very few fillers or additives.

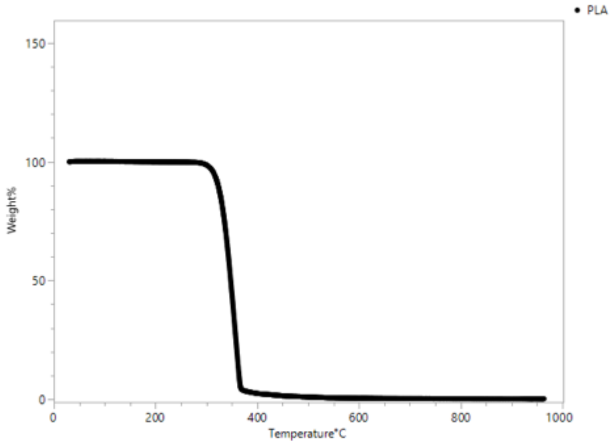

Figure 5. *Thermal-oxidative degradation for PLA sample under air atmosphere. (20°C / min)*

The PLA sample was tested. The graphed data was similar to that of a PP-type plastic, whose curve fell at approximately 300°C and bottomed out at approximately 360°C. This tendency towards lower temperature was indicative of a faster degradation time under environmental conditions.

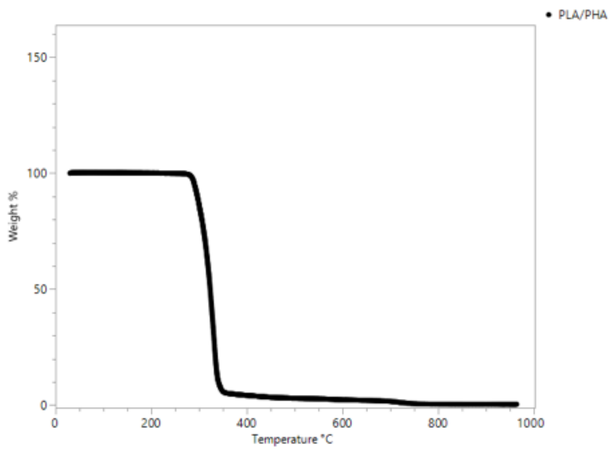

Figure 6. *Thermal oxidative degradation for PLA/PHA sample under air atmosphere. (20°C / min)*

The PLA/PHA blend sample was tested. The results were very similar to those of the pure PLA and the PP samples. There was a slight improvement upon pure PLA, with the curve falling at approximately 300°C and bottoming out at approximately 330°C.

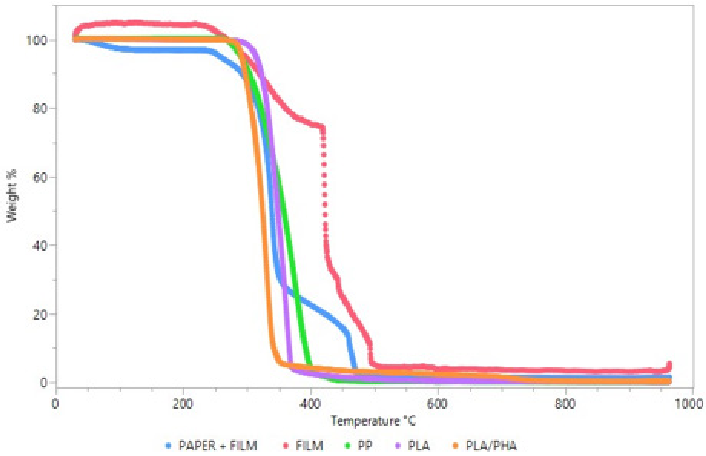

Figure 7. *Overlay plot of thermal-oxidative degradation for all samples under air atmosphere. (20°C / min)*

With all of the curves overlaid, it was possible to observe an index for degradation temperature. PLA/PHA degraded under the lowest temperature, followed by pure PLA, PP, and finally, the PE samples.

Discussion

The results of the TGA testing present evidence that the existing PE polymer films currently being used in single-use hot-beverage containers require a considerably longer time to degrade fully than both the two biopolymer samples and the PP cold-beverage container's sample.

According to the time-temperature superposition principle of polymer physics, the degradation curve of a polymeric material can be observed as identical at a higher temperature for a shorter period of time as at a low temperature for a longer period of time. Using this principle, the resulting TGA data can represent the duration of the various material's degradation processes. The PLA/PHA material performed the best of all the tested samples. These findings are consistent with existing literature, as the general biodegradable properties of bioplastics have been well established. It should be noted that the PLA/PHA blend performed better than all of the tested samples by a noticeable margin. This finding conclusively answers the research question of this experiment; in terms of degradation, PHA polymer substitutes provide an exceedingly large improvement over existing applications. In addition to degrading faster, PHA is also biocompatible, making it a favourable substitute from a food safety perspective. An interesting aspect of these results is the vast discrepancy between the PE film and the other samples. The PE film was found to degrade approximately 100°C hotter than the average of the other plastics. This difference is assumed to be due to fillers and additives present in the PE formulation. Per the time -temperature superposition principle, this ~100°C difference establishes the material as essentially inert within the environment for an immensely long period of time compared to all the other plastic samples tested. This finding is particularly troubling as landfill is the most widely accepted disposal stream for paper cups (Changwichan and Gheewala, 2019). Following the findings of this experiment, paper cups currently in landfills would likely have their plastic component remain intact for many years longer than even the PP cold beverage cups. The impact made by a bioplastic substitute for existing polymer coatings would be substantial. Previous studies such as Lomonaco et al. (2020), have focused their examination on photo-degradation which is most prominent when the debris is in the open air; the structure of this experiment examines thermal degradation, the process by which materials degrade at a relatively stable temperature range for a prolonged period of time. In photodegradation, PE plastics were shown to emit harmful VOCs into the atmosphere (Lomonaco et al., 2020), establishing long-lingering PE films as being more than just a concern of debris. As these plastic films remain in the environment, they pose a considerable health threat to both humans and surrounding biota. These paper cups, while seemingly less caustic than plastic waste, can account for far more devastating effects in the long term. PHA, and other biopolymer alternatives to PE, constitute a clear and present solution to an environmentally critical issue.

This experiment is relevant to this issue as the critical examination of emerging alternatives like PHA is currently limited. The data derived from TGA is robust enough to conclude concrete answers for the initial research question. However, this experiment is limited to the very broad understanding of thermal-oxidative degradation duration for each sample. Ideally, a detailed analysis of the individual chemical compositions of each submitted sample would be conducted. The sample collection could be expanded to incorporate several different paper cups currently available in restaurants. An understanding of the degree to which the findings of this experiment vary across similar products would be valuable. Additionally, an array of different PHA formulations could be tested further to narrow down an optimal substitute for PE film. More broadly, this experiment could be expanded upon to produce actionable results for use in packaging design. Overall the data collected is replicable and valuable, even if only by the instrumentation alone.

Conclusion

The current standard established for single-use hot-beverage containers leaves much to be desired in terms of environmental consciousness. Compared to bioplastic alternatives, specifically PHA-type biopolymers, the shortcomings are evident. PHA blend biopolymers provide a significant advancement in terms of degradability and sustainability when considered for use in packaging applications, especially hot-beverage containers. Thermogravimetric analysis (TGA) was effective in establishing an index for the thermal-oxidative degradation of PLA, PLA/PHA, PP, and PE plastics; the resulting data established PLA/PHA as the most suitable candidate for prompt degradation in landfill conditions. PE film was found to be excessively resistant to thermal-oxidative degradation. This understanding produces concerning questions about the state of plastic waste from paper cups. Much of the plastic waste produced by this type of packaging has and will continue to exist as debris for a significant period of time. The implications of this notion will continue to be relevant as global environmental concerns grow larger. PHA and other biopolymers present a viable solution to traditional petroleum -based plastics; the benefits are numerous, though significant infrastructural accommodation is necessary. Comprehensive research into biopolymers, specifically PHA, is still developing, and more robust characterization and application-based approaches are needed. Much can be remedied with a better understanding of biopolymers' qualities and characteristics.

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