Sustainability performance of polyethylene terephthalate, clarifying challenges and opportunities

Parikshit Sarda1 | Jay C. Hanan2 | Joseph G. Lawrence1 | Masoud Allahkarami2

1Polymer Institute, Department of Chemical Engineering, University of Toledo, Toledo, Ohio, USA
2Mechanical and Aerospace Engineering, Oklahoma State University, Tulsa, Oklahoma, USA

Correspondence
Jay Hanan, Mechanical and Aerospace Engineering, Oklahoma State University, Tulsa, OK 74106, USA.
Email: jay.hanan@okstate.edu

Abstract
Publications on polyethylene terephthalate (PET) continue to increase including the number of publications on recycling. PET is a versatile material with the ability to be remade from its polymer state through mechanical recycling and even back to its original monomer through advanced recycling. The scale of PET's use affords continued research and applications in improved recycling. Publications on new uses of discarded PET and the ability to clean and convert it into many forms including alternative materials are expanding with an attempt to complete circular use or improve the end of life. As indicated in life cycle assessment studies, increases in recycling lower the energy required to manufacture products. The future for PET will reduce energy demands further with the largest breakthroughs in recycling technologies and bio-sourced resins trending toward zero energy and carbon negative solutions. Opportunities remain for improvement in the use of PET with lightweight. The testing of new resins, development of bio-feedstocks, improvements in engineering, processing, recycling, and design continue to provide benefits. This review provides context for these developments.

KEYWORDS
bio-based, life cycle analysis, lightweight, polyethylene terephthalate, recycling, sustainable, upcycling

1 | INTRODUCTION

1.1 | Background and context for the review

Plastics have been extensively used for many decades. The material category that plastic encompasses continues to change. It is increasingly obvious and well documented how plastics are used in everyday life and find many special uses including personal protective equipment (PPE) that save lives and enables clinical advantages, food production, safety, storage, delivery, medical treatment, and entertainment. Global production of polymers was reported at 322 million metric tons in 2015 (steel was 1623 million metric tons in 2015) with the largest producers from Asia, Europe, and the United States.1 The vast majority of polymers are commodity thermoplastics.1 Thermoplastic polymers are endowed with properties that make them ideal for applications such as insulation and packaging.1,2 They are durable, water-resistant, and versatile. For example, in 2017 the contribution of plastics to the European economy reached a market size of EUR 355 billion while employing 1.5 million people2 with similar advantages across the globe. Plastics are lightweight and have significantly contributed to
decreasing transportation costs and extending the shelf life of perishable goods. One such plastic, polyethylene terephthalate (PET) is also one of the most stable materials for a variety of environments or uses and even long considered safe for medical applications. According to the same study, 90% of thermoplastics in use are made up of, polyethylene (HDPE and LDPE) (34.4%), polypropylene (PP) (24.2%), polyvinyl chloride (PVC) (16.5%), with smaller percentages from polystyrene (PS), polyethylene terephthalate (PET), engineered plastics, and high-performance polymers. The properties and value of plastics cover a wide range. It is important to carefully consider the life cycle of individual types of plastics, their potential, and possible contribution to sustainability. The main objective of this review is to consider the breadth of research of the plastic PET in context of the potential opportunities for improving sustainability across different domains of its life cycle.

Responsible manufacturing and end of life considerations for designs is an increasingly key area of focus around the globe. 600 million tons of plastics have been recycled. PET is recyclable and the most recycled plastic worldwide. Furthermore, the PET that is not recycled today is valuable enough that R&D efforts are underway to increase collection and recovery through advanced recycling techniques or maximizing features that aid in recycling, such as improved labeling and the reduction of secondary materials.

For multiple reasons, polyethylene terephthalate (PET) has become the most popular packaging material worldwide for beverages. With its clarity, food safety, cleanliness, toughness, competitive cost, and barrier properties; PET is unmatched as a material for use in soft drinks, mineral water, energy drinks, ice teas; and is often used for more sensitive beverages like beer, wine, juices, and medicines. This is also applicable for recycled PET (rPET). The first approval of post-consumer PET in direct food contact applications was in 1991 in the United States.

1.2 Hypothesis

This review paper considers recent updates in polymer research, manufacturing methods, life cycle assessments (LCA), and emerging observations focused on the specific plastic polyethylene terephthalate (PET) regarding its use in packaging applications, and its increasing potential for sustainable application over alternatives.

1.3 Definitions

Here we will follow the convention that plastics are polymers including any additives used for manufacturing goods.

Cradle-to-grave—an LCA or LCI covering all life cycle stages of a product system from raw material extraction through end-of-life and recycling when applicable.

Cradle-to-resin—an LCA or LCI covering life cycle stages from raw material extraction through raw material production (i.e., does not cover the entire life cycle of a product system).

Cradle-to-EG—an LCA or LCI covering life cycle stages from raw material extraction through Ethylene Glycol production.

Life cycle assessment—compilation and evaluation of the inputs, outputs, and the potential environmental impacts of a product system throughout its life cycle.

Life cycle inventory—phase of life cycle assessment involving the compilation and quantification of inputs and outputs for a product throughout its life cycle.

Life cycle impact assessment—phase of life cycle assessment aimed at understanding and evaluating the magnitude and significance of the potential environmental impacts for a product system throughout the life cycle of the product.

Biodegradable polymer—a polymer that can be decomposed by the action of living organisms, usually microbes, into water, carbon dioxide, and biomass. The conditions for biodegradation should be clearly indicated. For example, industrial composting conditions or elevated temperatures.

Bio-PET—a PET plastic that is made partly or wholly from polymers derived from renewable sources. Bio-PET is not normally bio-degradable. The fraction of polymer from a renewable source should be indicated. For example, 20%, 60%, or 99% bio-PET.

Advanced recycling—includes chemical recycling, a process by which a polymer is reduced at the chemical level to its original or intermediate monomer form so that it can eventually be re-polymerized and remade into the same or new plastic materials that go on to be new products.

Upcycling—a reuse of discarded objects or material in such a way as to create a product of higher quality or value than the original use. The comparison is not to the discarded material, but to what use it originally came from. An example is upcycling a shirt to a syringe. Downcycling would include recycling bottle material into carpet.

2 METHODS

2.1 Explains the literature search methods used for literature review

We systematically reviewed the literature regarding sustainability of PET associated with its recycling practices, upcycling capabilities, ability to be carbon neutral and
life cycle assessment. Web of Knowledge and Science Direct databases were used, and different key words such as recycling of polyethylene terephthalate, life cycle analysis of polyethylene terephthalate and bio based polyethylene terephthalate based on the section being reviewed were used. The literature review was performed in the first 2 weeks of February 2021. There is no dearth of publications in this field and the following histogram in Figure 1 represents the total number of publications and publications since 2015 for given keywords.

Breakdown of literature on recycling of polyethylene terephthalate into different recycling routes is presented below in Figure 2.

It was important to filter the publications based on quality and relevance. Also, our aim was to show the history and development progress for each topic. For advanced recycling of PET, we present literature covering each recycling method and their sub methods.

Readers are also referenced to existing recent review papers on advanced or chemical recycling of PET. PET has two major applications in the packaging sector and textiles sector. Even though, the primary focus was on bottle recycling, a study on PET based textile recycling is also presented. This covers the entire spectra of advanced recycling of PET. PET upcycling is an emerging topic and there are comparatively less studies published in comparison to advanced recycling of PET. Not all these cases represent a conversion to a higher added value product. In this paper, we sequentially presented PET upcycling studies which converts PET into alternative useful products including polyhydroxyalkanoates (PHA), hydroxyanthranilic acid (HHA), activated carbon, fiber reinforced plastics (FRP’s), N-doped carbon, fuel cell prerequisites and similar applications. A variety of conversion processes such as hydrolytic pyrolysis, physical activation, fermentation, carbonization, biological valorization, crosslinking, and saponification that facilitate PET upcycling are covered. In some cases, these conversion products have a lower value end of life than PET, however, if the process can tolerate a lower purity source, there can still be value in the conversion.

Among, 45,920 published life cycle analysis studies, we present comparative life cycle analysis of PET with other materials commonly used for packaging like aluminum and glass. We also included publications which discussed comparative life cycle analysis of PET with other polymers such as HDPE, PP and polylactic acid (PLA). To get insights into improved sustainability of PET over the years, we included a study that compared PET life cycle analysis over the years. Similarly, looking into PET only, we included studies composed of “cradle to resin” and “cradle to grave” life cycle analysis of PET. Also, readers are referenced to a recent review paper which discussed different life cycle analysis of PET. In the case of bio-based PET, we chose to cite the work that exhibited successful integration of green monomers obtained from commercially available renewable sources and showed comparable mechanical and barrier properties to PET. In this regard, monomers obtained from tartaric acid, sugar-based monomers, bio derived raw material (including waste byproducts) which have commercial availability were considered in this review. There are other sources of renewable monomers, but they were not included in this review.

3 | RESULTS

As a foundation for section 4, discussion, the results provide a summary and sampling from observations in the literature on the topics of sustainability in PET.
3.1 | Life cycle analysis

3.1.1 | Provides LCA examples from the literature for PET including some comparisons to alternative materials such as glass

Life cycle assessment (LCA) is a scientific method for making comprehensive, quantified evaluations of the environmental benefits and tradeoffs for the life cycle of a product system throughout its life. LCAs begin with raw material extraction and continue through disposition at the end of their useful life. Life cycle analysis (LCA) should be conducted following internationally accepted standards for LCI (Life cycle inventory) and LCA methodology as outlined in the ISO 14040 and 14,044 standard documents.11,12 In this section, we provide our review on current state of art of Life cycle analysis for polyethylene terephthalate (PET). We have also included, LCA of PET compared with alternative packaging materials, biodegradable plastics, and other commodity polymers such as polyolefins.

Marathe et al., reviewed the different LCA studies performed on PET bottles as a functional unit to enable a holistic understanding of the complete life cycle.13 During comparative studies, the entities under consideration are commonly the units used as single serving of alternative materials for packaging. This study provides better insights for decision-making to optimize the material and energy flow for each stage of life cycle of PET bottle by comparing various system dynamics of the PET bottle at micro and macro levels at distinct stages, regions in the life cycle.

As discussed earlier, LCAs begin with raw material extraction, production, manufacturing, recycling, and continue through end-of-life disposal. Typically, an LCA can be subdivided into two major stages, namely production and disposal. Hence it becomes important to study the effect of various disposal methods on the life cycle analysis of PET. Ncube and Borodin performed a LCA of PET bottles in Tomsk, Siberia, Russia to evaluate environmental burden in terms of energy required and waste generated with PET bottled products.14 They studied the effect of different end-of-life management scenarios including landfilling, incineration, and recycling. The environmental profile for 1 kg of PET bottle as reported in their study is tabulated in Table 1.

With different disposal methods, it is also important to have a detailed analysis of the PET production stage. In a recent study by Franklin Associates, they performed a “cradle to resin” life cycle analysis of polyethylene terephthalate (PET) resin. This analysis included raw material extraction and acquisition (terephthalic acid and ethylene glycol) followed by resin manufacturing process within its system boundaries.15 The TRACI 2.1 method, developed by the United States Environmental Protection Agency (EPA) specific to U.S. conditions and updated in 2012, is employed for most of the impact categories examined.16 The life cycle inventory results and life cycle impact assessment results are presented based on 1000 kg of PET resin and categorized into (1) cradle-to-PTA (terephthalic acid) production (2) cradle-to-EG (ethylene glycol) production and (3) PET resin production. The results are summarized in Figure 3 and Figure 4.

The report also compares LCI and LCIA results of this study (2018) with a previously published study (2011)17 prepared by Franklin Associates. This provides us with the effect of technological advancements, which improves the sustainability of the PET production process

|                      | Energy requirement (MJ) | Emission (g) | Greenhouse gases (kg CO2-eq) |
|----------------------|-------------------------|--------------|------------------------------|
| PET production       | 83.80                   | 2433.20      | 2.35                         |
| Waste recycled       | 27.10                   | 170.25       | 0.98                         |
| Waste landfilled     | 60.00                   | 2177.78      | 44.65                        |
| Waste incinerated    | 32.50                   | 2019.65      | 1.95                         |

TABLE 1 | Environmental profile for 1 kg of PET bottle14

FIGURE 3 | LCA analysis of 1000 kg of PET (cradle to resin)15
over the last decade. Table 2 shows the comparable LCI and LCIA categories for the year 2011 and year 2018 for 1000 kg PET resin production basis and includes the percent change from the original results for each category.

Along with the LCA of PET, it is vital to perform comparative life cycle analysis of alternate packaging materials like glass and aluminum. This provides us with the relative sustainability of various packaging materials. Sahel employed an LCA methodology to perform a comparative evaluation of beverage packaging materials such as glass, aluminum (Al), and PET.18 The study was based on a functional unit of 1000 L of beverage to be carried by 300 ml glass bottles, 330 ml Al-cans, and 2000 ml PET bottles which are manufactured, consumed, and disposed within Palestine. LCI data regarding the three packaging materials (glass, Al, PET) were collected from different local beverages industries working in Palestine. Results of LCI study performed is summarized in Table 3.

The life cycle impact assessment was followed to correlate the products life cycle and its potential environmental impact. Impact categories were classified, characterized, normalized, and weighed based on their significance in Palestinian context. The sensitivity analyses of the three packaging systems revealed that the total weighted environmental score of all packaging systems decreases as the recycling rate of packaging materials increase.

A similar comparative analysis was conducted by Franklin Associates, a division of the Eastern Research

![Figure 4](image)

**FIGURE 4** LCIA analysis of 100 kg of PET (cradle to resin)15

|                          | Total energy (GJ) | Total solid waste (kg) | Global warming (kg CO2-eq) |
|--------------------------|-------------------|------------------------|---------------------------|
| **PET 2018**             | 61.4              | 95.1                   | 2233                      |
| **PET 2011**             | 70.4              | 141                    | 2733                      |
| Percent decrease (%)     | 13                | 33                     | 18                        |

**TABLE 2** Comparison of LCI and LCIA analysis for 1000 kg PET in 2011 and 201815,17

| 1000 L of beverage       | Energy consumed (MJ) | Water consumed (m³) | Solid waste (kg) | Greenhouse gases (kg CO2-eq) |
|--------------------------|----------------------|---------------------|-----------------|-----------------------------|
| 300 ml glass bottle      | 9266.3               | 9.228               | 721.4           | 640.66                      |
| 330 ml Al can            | 12230.6              | 28.80               | 43.62           | 80.32                       |
| 2000 ml PET bottle       | 334.4                | 2.236               | 24.16           | 16.91                       |

**TABLE 3** Comparative LCI analysis of packaging materials for 1000 L of beverage capacity18
Group. They evaluated the environmental footprint of the PET bottle and alternative containers used for non-refillable single-serving soft drinks with the help of LCI. The analysis considered 12-ounce aluminum can (8333 units), 8-ounce glass bottle (12,500 unit), and 20-ounce PET bottle (5000 unit) to suffice functional unit of this analysis of 100,000 ounces of soft drink. The weights of each container system were determined by purchasing samples at local (Kansas City) retailers in 2008 and listed on the Table 4.

The container system included primary containers, closures, and labels. The scope and boundaries of the system included material production, fabrication, disposal, and recycling. The LCI results include energy consumption, solid waste generation, and greenhouse gas emissions for different containers as shown in Table 5.

This Table 5 shows us that environmental impact in terms of energy consumed, solid waste generated, and greenhouse gases emitted is maximum for glass which is heaviest based on weight per functional unit among all three packaging materials considered. However, this correlation does not hold up for aluminum can, which are the lightest, but still requires more energy and produces more solid waste and greenhouse gases than PET bottles, which had the least detrimental impact on the environment. We can see in Table 4 the weights of aluminum cans and PET bottles per functional unit are comparable with each other whereas glass bottles have at weight per functional unit around 20 times higher than Aluminum cans and PET bottles which along with the higher processing temperature increases energy requirements, solid waste generation, and greenhouse gas emissions.

Franklin Associates, a division of Eastern Research Group, have performed different studies on life cycle analysis (LCA) of standalone PET resin and PET resin in comparison with other polymers. In one of their studies, a virgin 12-ounce PET bottle was analyzed and compared with similar bottles produced using poly-lactic acid (PLA). The end-of-life scenario considered for these bottles is 20% incineration with energy recovery and 80% landfilling. In the same study, an additional end-of-life scenario for PET was considered where PET bottles were recycled with a 23.5% open loop recycling rate. The complete LCI results include energy consumption, solid waste generation, and environmental emissions for 10,000 water bottles and are summarized Table 6.

When compared with other packaging materials, as we can see from Tables 3, 2 L PET beverage bottles have the least environmental impact, while 330 ml Al beverage cans come second after PET bottles, whereas 300 ml glass bottles were found to have the highest environmental impact. Table 6 shows that the net energy requirement for PLA water bottles is 15%–25% higher compared to PET water bottles. Postconsumer solid waste generated in PLA water bottles is higher compared to that of recycled PET. Total greenhouse gas emission for PET and PLA water bottles was within 5% and hence not considered significantly different.

In another study, Franklin Associates performed Life Cycle Analyses (LCA) on post-consumer recycled polypropylene (PP), high density polyethylene (HDPE) and polyethylene terephthalate (PET) resins to quantify the total energy requirements, energy sources, atmospheric pollutants, waterborne pollutants, and solid waste during production of recycled resins. In addition to this analysis, they also reported life cycle impact assessment (LCIA) results including acidification potential, eutrophication potential, and smog formation potential. This was a “cradle to gate” analysis which begins with collection of postconsumer plastic resins and includes sorting and separation processes as well as reclaimed processing and transportation between process steps. In this study, postconsumer recycling methodologies were categorized as cut-off methods or open-loop methods. In the cut-off method, all virgin material production burdens are assigned to
the first use of the material, and all burdens for material recovery, transport, separation and sorting, and reprocessing are assigned to the recycled material. In the open-loop allocation method, the burdens for virgin material production, recovery and recycling, and ultimate disposal of recycled material are shared among all the subsequent applications throughout lifecycle. Table 7 summarizes % reduction for different parameters as compared to virgin resins.

As can be seen from the table, the analysis concludes that recycled resin has approximately 50% lower environmental impact as compared to virgin counterparts. Their results also indicate PET uses 12+/−1% less total energy, generates less global warming, and has lower water consumption than the other polymers in the study.

More recent LCA analysis of single-use cups indicates that the efficiency of recycling and resin production is so high, it is difficult to achieve a lower environmental footprint with reusable containers. The researchers followed a two-step method where traditional LCA was followed by a second step which aggregated a single impact for impact categories such as acidification, eutrophication, and water scarcity index into production, use, and end of life phases. This analysis led to estimation of an environmental breakeven point for reusable products to be more sustainable than single use products. This environmental breakeven point for reusable cups is dependent on washing location and transportation distance. In this study both PP and PET showed the best environmental performance.22

### 3.2 Reducing material use through efficient design

3.2.1 Provides some examples of published cases where sustainability was improved through design changes often categorized as lightweighting

Lightweighting is a concept for designing and manufacturing plastic parts or containers with a focus on efficient material use. The intention is to use less material without compromising functional properties of a part or the packaging. Accompanied by all the efforts for recycling plastics, one of the main aspects of sustainability is to take advantage of advances in material science and processing technology to optimize properties of plastics in order to use less material and energy. A good example of this approach is nanocomposite plastics or self-reinforced composite plastics.

Reinforcement with the same base chemistry (self-reinforcing) has advantages in compatibility, load transfer, and processing ability. As an example, polyethylene terephthalate (PET), a semi-crystalline polymer, possess a micro-structure that can be engineered through process history. Bi-axial stretching near the glass transition temperature yields a semi-crystalline microstructure in PET controlled by a function of temperature and strain rate where, in many cases, the crystalline phase, while even at 60% volume, can be kept to small domains that act as nano-scale reinforcements. Depending on process history, the material can also remain clear at such high crystalline fractions.

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**Table 6** Comparative LCI analysis of PET with PLA²⁰

|          | Net energy (GJ) | Post-consumer solid waste (kg) | Greenhouse gases (kg CO2-eq) |
|----------|----------------|-------------------------------|-----------------------------|
| PLA      | 19             | 168                           | 744                         |
| PET      | 16.6           | 163                           | 757                         |
| PET with 23.5% RR | 15.2       | 144                           | 710                         |

**Table 7** % Reduction for different parameters as compared to virgin resins²¹

| Parameter          | Cut-off | Open-loop |
|--------------------|---------|-----------|
|                    | PET     | HDPE      | PP       |
|                    | PET     | HDPE      | PP       |
| Total energy       | 79      | 88        | 88       | 39      | 44        | 44       |
| Water consumption  | −4      | 59        | 46       | −2      | 29        | 23       |
| Solid waste        | 58      | −1        | 23       | 29      | 0         | 12       |
| Global warming     | 67      | 71        | 71       | 34      | 35        | 36       |
| Acidification      | 70      | 47        | 58       | 35      | 23        | 29       |
| Eutrophication     | 46      | −2        | 43       | 23      | −1        | 21       |
| Smog               | 75      | 37        | 50       | 37      | 18        | 25       |

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Bi-axial stretching near the glass transition temperature yields a semi-crystalline microstructure in PET with outstanding mechanical properties,\textsuperscript{25,26} illustrated in the Figure 5A, enabling a process window for light packaging and container applications. Understanding the microstructure is often done in the context of micro-mechanical models and experimental methods sensitive to the 3-D orientation of crystal domains. X-ray diffraction conveniently probes the polymer at a scale of the reinforcement’s lattice parameter. Using X-ray texture analysis, molecular alignment was also shown favorable for improving mechanical stiffness as shown in Figure 5B.

Understanding the mechanical behavior of plastics and resulting implications in the design of packaging containers has advanced significantly in the past four decades.\textsuperscript{27–30} Successful design initiatives across the industry have reduced the weight of PET containers by more than 30\% while maintaining the performance of the container.\textsuperscript{31} Improving the efficiency of targeted material distribution through design and process optimization is crucial for future light-weighting efforts.\textsuperscript{32}

This necessitates the adaptation of advanced characterization tools as part of the design and development process. In conjunction with improvements in understanding the role the crystalline phase plays in material property enhancement, new techniques in material characterization and process optimization have helped lightweight products. X-ray tomography, a non-destructive 3D imaging technique based on the X-ray absorbance of materials has been implemented for package design.\textsuperscript{33–35} It is effective in capturing details of complex 3D geometries with micrometer resolution.\textsuperscript{33,36}

Stretch crystallization is an important material characteristic of PET. Understanding it is useful in the design of containers. During blowing, PET is stretched bi-axially in both radial and axial directions as shown in Figure 7. Bi-axial stretching orients PET chains which induce a form of crystallinity, making the film stiffer and stronger,\textsuperscript{30} see Figure 5. During blow molding, the wall thickness and diameter of the bottles change along their length, making it a non-uniform correlation with the initial preform thickness profile. A method to determine the biaxial stretch ratio and thickness at each segment of the stretched film is available\textsuperscript{37} which introduces an optical microscopy-based technique to track material translation and deformation from a preform segment to a bi-axially stretched segment on a bottle, as in Figure 6. Percent crystallinity at different stretch ratios was measured using micro X-ray diffraction.\textsuperscript{25}

When reviewing the multiple LCAs referencing PET containers and packaging, it is apparent that optimal design of the containers was not always considered. This means the carbon footprint and overall environmental impact of the containers in those studies can be further reduced. Even when a material appears a best choice with respect to many alternatives, this one engineering

FIGURE 5 (A) Experimental modulus compared to Halpin-Tsai model, based on percentage of crystallinity; (B) X-ray diffraction pole figure showing PET crystal orientation

FIGURE 6 Material tracking from preform to bottle
Improvement amplifies its advantages. As will be discussed momentarily, when that same material can be obtained via carbon negative or otherwise more beneficial environmental pathways, the advantages toward that material further increase.

3.3 | Mechanical recycling

3.3.1 | A brief introduction to mechanical recycling with reference to the literature

The majority of recycling today is done mechanically. It is so common, most mechanical recycling is simply called “recycling”. The contrast to other types of recycling will be more apparent when considering advanced recycling in the next section. Mechanical recycling has been commercially successful for PET. Particularly in some countries in Europe such as Germany where mechanical recycling recovers over 90% of PET from packaging.\textsuperscript{38} The most successful programs have high recovery of consumer products. Often, successful PET bottle collection programs with high collection rates incorporate deposit return systems. There is enough material available around the world that industry has developed methods to physically clean and reuse PET for food grade applications. Plastics in general still make up a minority of the material going to material recovery facilities, where paper-based products make up the majority, often greater than 90%. The latest developments are around sorting and cleaning technologies, particularly using new sensors and automation.\textsuperscript{39,40} Though the recently published article, published in 2019, mistakenly concludes that non-food contact PET cannot be recycled into food contact applications, when the technologies already exist to mechanically upcycle PET to food contact applications. Generally, this depends on multiple layers of washing and cleaning with an important dependence on melt filtration.\textsuperscript{41} Some regulations may not account for this capability. Globally, the current state of mechanical recycling is around 12%.\textsuperscript{42} There has also been research on improving polymer performance using chain extenders, for example,\textsuperscript{43} advances in melt filtration, and other tools. Early on, there was difficulty to minimize thermal degradation.\textsuperscript{44} The latest technologies minimize this issue with some options for processing further reducing energy consumption as well.\textsuperscript{45–47} As stated above in the LCA section, using recycled content significantly reduces the energy required to remake a product. Another advantage of recycling is the freedom to remake products into the variety of shapes and styles that consumers demand.

Mechanical sorting technologies have difficulty removing some substances such as dyes used to color PET. Therefore, colored PET is often used for applications that do not need clear polymer. Along with a fiber reinforced plastic (FRP) application referenced here, there are several others in development which can consume otherwise unwanted material.\textsuperscript{48} Such applications are often a “down-cycling” with reference to the material’s use in its original application. Even with off color material, there are standard methods in industry today to improve the color of mechanically recycled PET. The typical issue is PET can become dark or yellow when melt mixed with other materials. There is a well-developed industry of additives to help brighten the color and often yellow is masked with blue dye.\textsuperscript{49} However, clear or natural PET is the best choice for continued mechanical recycling.

As another example, umber color beer PET bottles or pharmaceutical containers are considered problematic for mechanical PET recycling. Such containers also often have additives used to prevent oxygen ingress,\textsuperscript{50} and they are also often singled out by recyclers\textsuperscript{51} as problematic.
over a certain total volume fraction. On the other side, bottles that are known to cause excessive yellowing during the reprocessing steps can be blended to r-PET to reproduce new amber colored bottles or containers without concern of yellow effects. While mechanical recycling can be used to at least maintain the same application, research is maturing on and advanced form of recycling also called chemical recycling where the monomer is recovered and able to be purified for use in any application be it medical, food grade, optical, or otherwise.

3.4 Advanced recycling of PET

3.4.1 An introduction to the topics and literature on advanced recycling

Advanced Recycling includes Chemical Recycling, also known as Chemolysis, which breaks down PET into its raw materials by means of various methodologies namely alcoholysis, aminolysis, ammonolysis, glycolysis, and hydrolysis based on the use of a reagent to decompose the PET structure. The reaction mechanism and end products of Chemolysis are summarized in Figure 8. Selecting a particular mechanism is an important component of Advanced Recycling.

Alcoholysis comprises the depolymerization of PET in an alcohol medium under high temperature and pressure circumstances. Alcoholysis is a reliable recycling method due to its ability to attain materials with diverse properties by optimizing vital reaction parameters such as temperature, pressure, catalyst, and alcohol type. Although over recent years methanolysis has gathered more attention, the use of various types of alcohols including ethanol, butanol, isooctyl alcohol, and trimethylolpropane has also been reported. Methanolysis of PET can be carried out with three different forms of methanol namely liquid methanol (conventional), super-heated (vapor) or supercritical methanol. Higher reactivity of the amine-group as compared to the hydroxyl-group in glycol or alcohol led to the development of Aminolysis and Ammonolysis of PET. Further, drawbacks of other conventional chemolysis methods (hydrolysis, methanolysis) like elevated temperature and high-pressure conditions were potentially reduced by aminolysis and ammonolysis. Glycolysis is the simplest and oldest method of PET depolymerization. Glycolysis is solvolytic degradation of PET by which the ester bonds are split and substituted by hydroxyl functionalities. Therefore, PET is required to react with proper glycols including ethylene glycol (EG), diethylene glycol (DEG) or propylene glycol (PG), dipropylene glycol (DPG), etc. to attain bis (2-hydroxyethyl) terephthalate (BHET) and oligomers. It was reported that glycolysis, as the oldest approach to PET degradation, was employed by prominent companies including: DuPont, Goodyear, Shell Polyester, Zimmer, and Eastman Kodak. Recently Eastman announced a new methanolysis plant in the United States. In another study, Khoonkari et al. show waste PET glycolysis was carried out using different catalysts namely metal salts, ionic liquid, enzymes and hydrotalcites. They conclude that the use of hydrotalcites as catalysts will enable a higher catalyst recovery rate and better purity of the final product. Concerning hydrolysis depolymerization, impure PET chains can be transformed into value-added products, particularly purified TPA and EG monomers. Existing commercial plants which use TPA and EG directly to produce PET have been the driving force for the progress of this approach. Hydrolytic depolymerization of PET waste can be performed under three conditions, namely neutral, basic, or acidic. Regardless of the reaction medium, hydrolysis involves chain scission of an esteric bond to form a carboxylic functionality (as an end group) by attacking a molecule of water.

Many researchers have been working on advanced recycling of PET. This can be evident with 17,088 publications on advanced recycling of PET out of which 1818 were published in the year 2020. Geyer et al. reviewed existing literature and summarized the reaction temperature, reaction time, reaction pressure, reaction products, yield, catalyst, and reagent used for various advanced recycling methods. They also suggest an alternative chemical recycling of PET by controlled depolymerization using blocking chain scission with defined amounts of the depolymerization agent. This method produced PET-oligomers of well-defined molecular weights in a greater range. These building-blocks enabled the synthesis of tailored block-co-polymers as potential compatibilizers to produce transparent PET-PEN-blends, which are suitable as an encapsulation material for photovoltaic cells or for hot refillable and pasteurizable food packaging. In a similar study, Sinha et al. reviews publication focusing on different advanced recycling techniques with an emphasis on industrial application. They conclude that PET waste recycling is economically viable if operated on a large scale and especially applicable to industrial waste. Bartolome et al. reviews methods for the advanced recycling of PET with special emphasis on glycolytic depolymerization with ethylene glycol. It covers research including the work by the authors, on various processes including sub- and supercritical, catalytic, and microwave-assisted depolymerization and introduces innovative technologies such as nanotechnology on the future developments in advanced recycling of PET. A comprehensive review of all advanced recycling methods is presented by Shojael et al. It supplies detailed information about the reaction condition of temperature and pressure, catalyst influence, catalyst dosage, catalyst
types, synthesized eutectic solvents, the assistance of heating source, PET/reagent ratios, characteristics of the product. They also discuss diverse depolymerization protocols like ionic liquid catalyst, phase transfer catalyst, ultrasound energy and microwave assistance in chemical recycling of PET at moderate operating conditions.69

From Zimmermann, “plastics containing ester bonds such as PET and polyester PUR can be considered as realistic targets for enzymatic degradation and recycling”.70 Biocatalytic degradation of PET with polyester hydrolase resulting in TPA and EG recovery at 60 to 70°C was achieved by Zimmermann. This process has easy recovery and purification of monomers and this process can also be targeted to produce value added polyurethane.

The studies presented above mostly dealt with PET bottles. In their published work, Barot et al. successfully recycled post-consumer cloth using castor oil as a replacement for diol in the presence of a Zinc Acetate catalyst. From the numbers of experimental sets, the depolymerization process in the presence of 1% of catalyst and ratio of PET: CO (1:5–1:7) at 230–240°C temperature provided the highest conversion and yield. Castor Oil in comparison with diols is renewable, easily available, environmentally friendly, economically cheaper, and hence sustainable.71 Success in the development of these technologies will lead to additional waste recovery efforts for products that were previously difficult to recycle. Initial implementations have already started in the United States and Europe.

3.5 | Upcycling of PET as a compliment to recycling

3.5.1 | Given the above introduction to recycling, this section introduces upcycling of PET and selected publications

Upcycling is a reuse of discarded objects or material in such a way as to create a product of higher quality or value than the original use. It involves the creative reuse of and transformation of waste materials into new materials of improved quality and environmental value. In addition to existing disposal, end of life management practices, and advanced recycling, several commendable initiatives have started to upcycle PET waste into higher cost products.72 For example, a study performed by Kenny et al. on the conversion of PET to a biodegradable
plastic polyhydroxalkanoate (PHA) is one of the earliest examples of PET conversion into PHA. In their study PET waste was hydrolytically pyrolyzed to obtain terephthalic acid (TPA) solid residue. Recovery was achieved by using carbon rich TPA as a substrate for a biotechnological process. Soil samples were collected from PET exposed soil at an industrial site as a TPA degrader. Three strains were identified from the soil and PHA accumulation was achieved at maximal rate of approximately 8.4 mg PHA/l/h for 12 h before reduced activity. The economics of PHA production through fermentation was linked to the cost of the starting substrate, which was replaced by PET waste in this study. In another study Esfandiar et al. developed activated carbon from PET waste. This was achieved by a physical activation method where PET waste was carbonized at 800°C and activated using CO₂ at 975°C to obtain activated carbon. Characterization of prepared activated carbon was performed to calculate physical and chemical properties. Activated carbon prepared using this method has a surface area of 790 m²/g and iodine number 630 I₂/g C which is comparable to commercial activated carbon. In another effort to reclaim PET waste, researchers at National Renewable Energy Laboratory combined reclaimed PET (rPET) and renewably sourced monomers to produce high-value fiber reinforced plastics (FRPs). To accomplish this, PET was first deconstructed and glycolyzed with linear diols that can be obtained from renewable sources and subsequently reacted with renewably sourced monomers to produce a series of UPEs or diacrylic polymers. These polymers were then dissolved in a reactive diluent with a free radical initiator to form a resin, which is applied to a woven fiberglass mat and reacted to produce a series of rPET-FRPs. rPET-FRPs produced with renewably sourced monomers outperform the standard petroleum-based styrene and maleic anhydride based on their experimentally observed higher glass transition temperature. rPET-FRPs materials result in 57% lower production energy and 40% lower GHG emissions. Tiso et al. presented the sequential conversion of PET into two types of bioplastics: a medium chain-length polyhydroxalkanoate (PHA) and a bio-based poly(amide urethane) (bio-PU). PET films were hydrolyzed by a thermostable polyester hydrolase yielding 100% terephthalate and ethylene glycol. Monomers from PET hydrolysis were used for conversion into PHA using Pseudomonas sp. GO16 K53. P. putida KT2440 was engineered to synthesize HAA (hydroxy acid) that led to the formation of Poly (amide urethane) by copolymerization with a diisocyanate and butanediol.

In the textile industry, interwoven cotton polyester blends represent one of the most prominent mixtures on the market, which are difficult to recycle because of their heterogeneity. In an effort to create a circular economy for cellulose based textiles, Haslinger et al. used [DBNH][OAc], a superbase based ionic liquid to selectively dissolve the cellulose component and separate cotton and PET polyester. The dissolved cotton component was dry jet wet spun to textile grade fibers with properties comparable to that of Lyocell with linear densities between 0.75–2.95 dtex, breaking tenacities of 27–48 cN/tex, and elongations of 7%–9%. They observed degradation of PET during this process which can be minimized by optimizing dissolution process conditions. Aiming toward the rectification of fiber grade PET waste, Barot and Sinha recycled post consumer polyester clothes into bis (2-hydroxyethyl) terephthalate (BHET) monomer in the presence of ethylene glycol as depolymerizing agent and zinc acetate as catalyst. The depolymerization yielded 90% BHET with no mechanical stirring and at operating temperature of 180°C with 1% catalyst loading. The recovered BHET can be utilized in industry for numerous applications including to synthesize PET. Sharma et al. proposed a method to utilize fiber waste obtained from spinning and weaving industry to manufacture a composite which has high degree of resistance to washing and abrasion. This composite fabric possessed hydrophobic character as evidenced by its lower surface energy and low moisture content. The crystallinity of the composite as compared to the cotton fabric increased to 59.29% from 55.81%. The thermal stability of the composite sample proved it was more stable having the weight loss of 3.0% for 5% PET composite as against 3.5% in cotton. The mechanical properties of this fabric were almost doubled in stiffness. The proposed composite would act as a substitute for a commercially available PET cotton blend of low percentages because of its composition and improved properties.

An additional recent example of upcycling is described in J. Qi’s paper discussing reprocessable vitrimers. Therein they describe a method to melt blend PET with 2,2-bis(hydroxymethyl)-2′, 2′-nitrilotriethanol, a polyol with a tertiary amine structure and used diepox (D.E.R.) during reactive extrusion to cross-link the activated PET chains. The resulting cross-linked PET showed improved creep resistance and was repressible by several methods including injection molding.

In another effort to recover and potentially upcycle PET waste, Kim et al. evaluated the feasibility of biological valorization of PET. In this study, PET was hydrolyzed in the presence of microwave radiation. The PET hydrolysate was separated into solid and liquid fractions by filtration. First, to obtain terephthalate (TPA) and EG. They demonstrated the in vivo protocatechuate enzyme (PCA) production from TPA. TphAabc 0a1s TPA 1,2-dioxygenase and TphB as DCD dehydrogenase, both
from Comamonas sp. E6, were used for the biosynthetic route from TPA to PCA in E. coli. PCA is also a key intermediate chemical in lignin refineries and the key precursor of aromatic or aromatic-derived chemicals. In the same study, TPA was biologically converted to various higher-value products compared to the value of PET, such as gallic acid, pyrogallol, catechol, muconic acid, and vanillic acid via PCA, using single or combined reactions of hydroxylation, decarboxylation, oxidative ring cleavage, and methylation. The other monomer from terephthalic acid (TPA) and vanillic acid was potential to use other monomers such as gallic acid, pyrogallol, catechol, muconic acid, and vanillic acid to produce higher-value products compared to the value of PET.

In the first step, PET was mixed with designated amounts of melamine and ZnCl₂/NaCl to obtain robust crosslinking structure of PET-melamine-ZnCl₂/NaCl as a mixture. Subsequently ZnCl₂/NaCl eliminate the weak bonds of the crosslinking structure with dehydration and Decarboxylation of the crosslinked structure to yield a more thermally stable carbon framework. NPC bears a large BET of 1173 m²/g and rich nitrogen dopants (13.5 wt%), and abundant micropores (0.4–2 nm) with many micron-scale pores (ca. 1–4 μm) and displays good performance in CO₂ capture and solar-driven steam generation. Another attempt to upcycle PET waste was performed by Ko et al. where they presented an innovative route wherein synthetic graphite is prepared from a waste polyethylene-terephthalate (PET) bottle. Carbonization of PET was performed at 900°C in inert atmosphere to obtain a carbonaceous material with carbon yield of 19%. Subsequently catalytic graphitization of previously obtained carbonaceous material by mixing with amorphous boron and thermal treatment at 2400°C under flowing helium gas. This technique overcame the intrinsic non-graphitizable property of PET and yielded graphite showing high crystallinity. It showed a much higher degree of graphitization (80.6%) than that (68.9%) derived from a well-known AR mesophase pitch. In addition, via a microwave-assisted liquid-phase exfoliation, the PET-derived graphite was exfoliated as graphene sheets with the average lateral size of 410 nm. Synthetic graphite has many potential applications such as anode materials in secondary batteries and fillers for carbon composites and may serve as an alternative source for graphene production. Ghosh et al. used density functional theory to upcycle of PET waste to synthesize disodium terephthalate for Li/Na storage. They carried out saponification of PET waste into disodium terephthalate in a microwave oven. After saponification, the white-color powder materials were collected using centrifugation and repeatedly washed with ethanol. The electrochemical behavior of this low-cost, environmentally benign organic molecular compound was studied in Li- and Na-ion cells. The delithiation potential of a disodium terephthalate anode was found to be approximately 0.65 V higher than the desodiation potential. They conclude that, the better C-rate performance of the composite anode for a Li-ion cell, as compared to a Na-ion cell, was due to inferior mobility of Na-ions in the electrode material, which is largely defined by ion size. In another study, Mirjalili et al. successfully upcycled PET waste bottles into electrochemical active carbon material by electrospinning fiber production. The process simply consists of dissolution of the plastic, fiberization through electrospinning, and carbonization in a furnace to obtain electrochemical carbon material capable of functioning as double layer supercapacitor substance. Characterization techniques confirmed that the developed material is a combination of amorphous carbon and reduced graphene oxide with high surface area. The results showed that the active material performed as a supercapacitor with added pseudo-capacitance and has a scope to further enhance the electrochemical performance of the prepared active material.

CF₄ is greenhouse gas with high global warming potential. Yuan et al. developed cost-effective methodology to synthesize porous carbon which was subsequently used to capture CF₄. They first carbonized the PET waste samples at 600°C followed by thermal activation in the presence of KOH at 600–100°C. Different KOH/carbon mass ratios from 1:1 to 3:1 had remarkable effects on the textural properties of the prepared porous carbons. CF₄ uptake was significantly affected by the specific surface area and pore volume of narrow micropores less than 0.9 nm in diameter. The recovered PET derived porous carbons exhibited a high CF₄ uptake, good CF₄/N₂ selectivity at relatively low CF₄ pressures, easy regeneration, rapid adsorption/desorption kinetics, and excellent recyclability.

### 3.6 | Bio-Based PET

#### 3.6.1 | This section reviews recent published developments and further benefits for sustainability from bio-based PET

Most of the PET produced globally is derived from petroleum feedstock. Since PET is a clean, safe, reusable material; dependence on petroleum for the raw materials of PET has fueled efforts to develop renewably sourced options. PET synthesized using renewable sources of raw material is called Bio-PET because of the living organism that is used to produce the material from carbon in the environment. This opens a possibility to achieve carbon neutrality or even sequester carbon in carbon negative...
pathways. It is important to note that PET synthesized from petroleum feedstocks is not normally biodegradable and integration of renewable monomers in Bio-PET will help in reducing carbon emission, but would not normally affect the biodegradability of the PET. Synthesis of terephthalic acid (TPA), ethylene glycol (EG) and in some cases dimethyl terephthalate (DMT) monomers for PET from renewable feedstock is under development and has produced some promising results.89 Industry application of at least partially bio-based bio-PET has been underway for over a decade.90 Much of this was focused on selecting a biological source for the EG portion of the PET molecule. Development of renewably sourced alternative polyesters to PET has also gained interest and to that end efforts to synthesize 100% renewably sourced PET have been realized.91 There are many monomers used in the production of most important polyesters that are accessible from renewable feedstock.92,93 Some of them are currently produced at ton/year levels and many other are still in an incipient state of development.

Tartaric acid (TA) is a low-cost naturally occurring compound present in many fruits and tis also formed in grape juice fermentation, making it a surplus byproduct of the wine industry. 2,3-O-methylene-L-threitol (Thx-diol) and dimethyl 2,3-O-methylene-L-tartrate (Thxdiester) can be synthesized from commercially available dimethyl L-tartrate. Japu et al. synthesized and performed characterization of PET co-polyester containing 2,3-O-methylene-L-threitol (Thx-diol) and dimethyl 2,3-O-methylene-L-tartrate (Thx-diester), two monocyclic acetal compounds obtained from L-tartaric acid.94 Homopolymers (PET, PThxT, PETHx) as well as PEXThxT and PETHxy copolyesters were synthesized through a two-stage polymerization process carried out in bulk. PET copolyesters made from acetalized L-threitol displayed thermal stability and \( T_g \) comparable to PET, whereas these two properties were suppressed in the copolyesters containing threurate units. Both types of copolyesters were able to crystallize for contents in tartaric acid derived units up to around 30% by adopting the crystal structure of PET. They conclude the suitability of tartaric acid as source of monomers, both diol and diacid, for preparing bio-based PET copolyesters.

In a similar study, Japu et al. copolymerized 2,4;3,5-di-O-methylene-D-glucitol (Glux-diol) and dimethyl 2,4;3,5-di-O-methylene-D-glucarate (Glux-diester) with ethylene glycol and dimethyl terephthalate by polycondensation in the bulk to produce PET copolyesters as well as their respective homopolymers.95 These sugar-based bicyclic monomers were synthesized from 1,5-Dgluconolactone, a commercially accessible compound derived from D-glucose. The PET copolyesters with either the diol or the diacid counterpart partially replaced by Glux had molecular weights in the 20,000–40,000 range and a random microstructure, and they were stable well above 300°C. The PET copolyesters containing more than 10%–15% of sugar-based units were amorphous while those others displaying crystallinity were observed to crystallize from the melt at much lower rates than PET. The glass transition temperature of PET dramatically increased with the incorporation of Glux whichever unit, diol or diacid, was replaced but the enhancing effect was stronger in the former case. A preliminary evaluation of the mechanical behavior of these copolyesters indicated that the genuine properties of PET were not substantially impoverished by the insertion of Glux. Compared to PET, the copolyesters exhibited a higher hydrolysis rate and an appreciable susceptibility toward biodegradation. The homopolymers made of these sugar-based monomers could not be obtained with high enough molecular weights as to be comparatively evaluated with copolyesters.

Damian Salazar synthesized PET with comparable quality using both petro based and bio derived raw materials.96 This research examined the critical areas in the separation and production of polymer precursors from bio derived samples, their use in the polymerization process to create PET, the effect of remaining low concentrations of impurities and the characterization of the polymer produced. Bio EG was provided with a high purity grade and PTA precursor (p-xylene) was separated using distillation and crystallization from two bio based aromatic samples. Separated p-xylene was oxidized to PTA following a modified Amoco process obtaining a material with quality close to a commercial PTA. The synthesis of PET with bio and petro monomers using melt phase polymerization was followed by a multivariable characterization of the polymers produced. Through the knowledge obtained from these analyses, the suitability of using a bio derived monomer to produce PET polymer was evaluated. The cumulative analysis of results determined that there was no difference in the use of bio or petro EG to produce PET and it concluded that it was possible to synthesize PET polymer with comparable quality using petro and bio derived monomers with similar purity.

2,5-Furandicarboxylic acid (FDCA) is a furan analog of TPA with carboxylic acid groups present at the 2 and 5 position on the furan ring. The 2 and 5 substitution on the five membered furan ring is similar to the meta connected benzene ring and introduces non-collinearity between the acid groups. FDCA can be synthesized by catalytic oxidation of hydroxy-methyl furfural (HMF). FDCA has been identified by the U.S. Department of Energy (DOE) as one of the platform chemicals for development of renewably sourced materials.97

Recently, Joshi et al. published a detailed study on the effect of inclusion of furan units on the chain
dynamics of PET and its parallels with the observed chain dynamics in PEF. They revealed that the solubility of FDCA is an order of magnitude higher in EG compared to that in TPA at the process temperatures. The enhanced solubility of FDCA in EG results in improved esterification kinetics especially at temperatures from 180 to 210°C to yield complete end group conversion. Lower esterification temperatures resulted in reduced DEG incorporation in the chains, and it also enables the direct esterification route of PEF. In continuation to this work, Joshi et al. published a fundamental study of orientation and strain induced crystallization of the PETF copolyesters and the subsequent effect on properties relevant for packaging applications. To avail the advantages exhibited by PEF, copolymerization of PET with small quantities of FDCA enabled a fundamental study of the effect of biaxial orientation, a critical microscopic phenomenon for packaging polymers, on the microstructure and properties of copolyesters of PET with FDCA (PETF). DSC studies revealed that the rigid furan units in the backbone disrupted the thermal crystallization mechanisms and reduced the crystallization rate significantly. The incorporation of the furan units in the PET backbone enhanced the oxygen barrier performance and mechanical properties in unoriented and biaxially orientated states.

4 | DISCUSSION

4.1 | The topics reviewed above are discussed in context of the reviewed PET literature and the author’s experience

It now only takes a few decades for new materials to become commonplace. The ability to purify, source, and modify new materials has never been so grand or broad. There are textbook examples in electronic materials based on purifying single crystal Si, the discovery of new alloys, and the recent promise of new applications from wonder materials such as graphene. Useful new materials that can be manufactured at low cost and high quantity find the greatest number of applications. Some of the best examples are polymers or plastics. Principally made from petroleum or natural gas, the cost to manufacture polymers has continued to decrease with new varieties and properties enabling invention, discovery, and the flourishing of industries and technologies from communication and transportation to agriculture and healthcare. The value for human health and medicine from polymers is alone a review worth considering, but outside the present scope. While one can quote broadly that worldwide production of plastics exceeds 360 million metric tons per year or that 5 billion metric tons of plastics have been discarded across the globe in the past decades, this overlooks the individual specifics of each polymer type.

While not the primary focus here, a review on PET is in the context of the global annual fraction used to produce fiber. Generally, 60% of global PET production goes to fiber applications. Each application has its own benefits and broader ramifications. Material selection plays a key role in benefiting the environmental and economic impact from a variety of applications, but design for optimum manufacturing and design choices for end of life also play important roles. The LCA of a product depends heavily on the material selected. The material influences the manufacturing and supply chain. Generally, when a material can improve sustainability in one application, it suggests it has advantages in other applications. For example, the production of a product with a polymer can have a low or even have a negative carbon footprint and therefore be an advantage in all situations. However, if there is one application instance where an LCA suggests undesirable environmental performance, such as energy consumption from an empty container shipped a long distance, it is not the general case for all applications of that material, as the associated manufacturing infrastructure is application dependent. In this specific case, shipping a container to a neighboring building has almost no impact on greenhouse gas emissions, but today some empty packaging is shipped great distances. Several materials such as glass, aluminum, multilayer composites, and others have off-site manufacturing steps that require transportation to the place of use. Furthermore, as we look to the future, new technologies for recovery of discarded materials in general and PET specifically can turn less favorable applications into favorable ones. More recent examples include upcycling or advanced recycling, where the application it came from such as fiber-based or multi-layered products is no longer a constraint. In addition, continued advances in mechanical recycling are broadening the capabilities to maintain a circular economy for the reuse of PET as a clean recycled raw material.

4.2 | Additives for PET

4.2.1 | The blow section discusses the additional beneficial properties of some additives for improving sustainability

Along with the variety of copolymers, research remains active in additives that modify PET to improve sustainability. Typical additive examples include antioxidants and compatibilizers as well as barrier enhancements and mechanical reinforcements. Recent research on chain extenders is another example that has been developing over
the last two decades. The authors have used such additives with industry applications and seen the marked improvements. In some cases, new additives perform multiple useful roles simultaneously, such as increasing barrier properties and acting as a reinforcement. Barrier enhancement from platelet like particles has been reported with examples in nano-clay and graphene. Increasing barrier results in longer shelf life for package contents and reduced waste in both the supply chain and for end users. Enhancement such as scratch resistance has also been reported in PET.

Particularly when the reinforcement is substantial, designers can reduce the weight of material needed to perform a load bearing function. This material reduction improves carbon use and overall environmental footprint without requiring significant changes in the installed infrastructure. Such improvements are only starting to be realized and another reason to continue the path forward in development for sustainability in polymers.

Presently, many mechanical recycling methods are not compatible with some additives. Limitations have been outlined by the Association of Plastic Recycler. When the rPET supply is substantial, dilution of additives is a practical solution. Dilution is not always necessary as many additives are effective in the range of 10 to 100 ppm. The ability to accept a broader range of PET based materials is being developed in sorting equipment and other compatibility focused technologies. This highlights another advantage to advanced recycling methods discussed above in their tolerance to additives. Additives are easier to purify and remove when polymers are broken down to their precursor state. Such tolerance to a wider range of enhancements to PET will allow the transition from higher carbon generating materials such as glass and aluminum. For example, advanced PET packaging solutions coupled with additives that improve shelf life with a lower overall LCA than alternatives will be a positive outcome of these efforts. Some competitive enhancements to barrier properties, for example, can also be accomplished with multi-layer solutions. Some of these barrier layers are also designed to be compatible with existing mechanical recycling technology but can require retraining of the systems that sort incoming containers for optimal application.

4.3 | Life cycle analysis

4.3.1 | Discussion of the PET life cycle analysis including results from section 3.1 in the context of sustainability

Life cycle assessments of PET and specifically PET bottles have generated great interest in the last decade. Most LCA studies are conducted to find the environmental impact associated with a certain quantity of PET production associated with specific packaging applications, for example: soft drinks, alcoholic drinks, cosmetics, pharmaceutical, and medicines; and also to assess the environmental impact associated with distinct stages of a PET package’s life cycle such as production, transportation, and recycling. Various recycling systems, protocols, and policies can be designed from the insight gained at each stage.

From Table 1, we saw that there is a significant advantage of recycling PET in terms of energy savings and emission reduction. Also, incineration performed under environmentally sound conditions such as energy recovery and flue gas scrubbing should be the preferred option over landfilling.

Based on LCI and LCIA analysis of 1000 kg of PET in Figure 3 and Figure 4 in Section 3 of this paper, PET production has comparatively lower environmental impact as compared to raw material extraction and purification. In Table 2, we saw that energy consumption and waste generation for PET production has decreased in the last few years by improving process efficiencies and operating practices. Such decreases are expected yet sometimes difficult to capture as they are not generally published by manufacturers.

Furthermore, it is notable that the energy cost per package as well as the carbon footprint has continued in a favorable reduction of 50% over the last decade with it possible for industry to produce more than 150 0.5 L bottles with 1 kg of PET where in 2010 70 to 80 was typical. While the studies referenced here are recent, PET was generally analyzed at container weights that are historically heavy or otherwise leave room for improvement, meaning the analysis was a conservative under prediction, whether of single-use cup performance, or the various bottle studies. Lightweighting has less room for improvement in aluminum or glass. “Glass has also negative effects on the environment and it is not clear that its use is beneficial in comparison to plastic.” Glass has advantages for barrier properties today, so continuing research on improving barrier in PET or other polymer packaging is beneficial to reduce the environmental impacts by enabling the ability to switch from glass to plastic without a loss in shelf life. Further analysis of the break-even point is also an attractive way to analyze the latest developments in efficient manufacturing and design. This type of analysis is needed for clearer understanding of modern package performance.

Furthermore, most LCAs do not include the generation of ocean plastic or microplastics.
This can be in part due to how difficult and recent the observation of microplastics is and the remaining uncertainty of their impact on the environment. Furthermore, most studies in these new areas do not differentiate between the types of resins observed and are even less likely to indicate the resin's source. In the recent 2020 review from Rahman, the primary focus was on the effects and presence of polystyrene (mentioned more than ten instances) and polycarbonate (three instances) with no indication of negative effects from PET. The studies in general do acknowledge the need for recycling and that some materials are easier to recycle than others. For example, microplastics could be hypothesized as existing in a similar percentage to what was manufactured, with polyethylenes being the most common polymer, they could also be the most common to find in a waste stream, but this ignores the details of how microplastics are generated. Among others, Andrady provides a useful review. It is also likely that the more often collected and recycled resins are correspondingly or proportionally absent from the waste streams and environment. One more reason to increase recycling rates. Of course, if the resin is present as a fiber as well as other forms (film, spheres, or films), the source of the fiber would be different than other forms—even if of the same polymer. Therefore, confirming success of a recycling program at reducing waste must be connected to measurements of the targeted polymer type and its production form. This is admittedly difficult to do for several reasons. The analysis is slow. It is also often conducted by teams that do not have expertise in the manufacture process. Some studies suggest that the majority of ocean plastic and even microplastics tends to originate from fishing gear or fibers. Such details can be crucial to navigating solutions to environmental issues, while not overreacting or exaggerating policy to the detriment of communities, often exacerbating the detriment further for disadvantaged populations. Also, some have suggested a focus should be on proper waste water treatment. In one recent publication related to wastewater treatment from PET, a critical detail was overlooked. The type of plastic used to simulate what might occur in the environment was contaminated with an additive not used in PET in most of the world. Unless this detail is clear, the conclusions can be misleading. However, it is still important that proper engineering controls be put in place to minimize environmental exposure. Additional research is needed to indicate what fraction of the variety of products including packaging are lost to the environment and what of those products remain inert over time.

4.4 | Recycled PET

4.4.1 | Discussion focused on recycled PET including both mechanical and advanced methods including results from sections 3.3 and 3.4 in the context of sustainability

Packaging and manufacturing of products using PET has seen multiple iterations of life cycle assessment due to it being common and the most recycled polymer. Recycling adds value and provides an obvious mechanism for increased environmental benefit, where the ultimate goal is to reduce environmental impact and support a circular economy. However, the complexity of that benefit requires LCA analysis such as Gomes who states, “multi-recycling trips, which relate the number of times the same packaging will return for recycling, significantly reduce all environmental impacts” and “PET is better for human health and ecosystem quality.” Gomes showed PET exhibited better environmental performance than glass bottles and aluminum cans, but even though relatively recent, under predicted the value for improved advanced recycling methods as well as the significant environmental contribution possible with package lightweighting. More recently, Kouloumpis stated, “the substitution of PET with glass as the material for bottling under the current waste infrastructure and management practices could lead to significant increases in GWP and hinder efforts to tackle climate change.” Sinha indicates, “PET recycling represents one of the most successful and widespread examples of polymer recycling. The main driving force responsible for this increased recycling of post-consumer PET is its widespread use, particularly in the beverage industry.” Sinha goes on to state, “it does not have any side effects on the human beings.” and, “PET does not create a direct hazard to the environment.” Finally, Sinha concludes, “the recycling of PET does not only serve as a partial solution to the solid waste problem but also contributes to the conservation of raw petrochemical products and energy. Products made from recycled plastics can result in 50–60% energy saving as compared to making the same product from virgin resin.” Since this publication, recycling technologies have continued to improve. Both mechanical and advanced recycling technologies have improved to the extent that bottle to bottle recycling is accepted in the industry with many methods obtaining letters of non-objection or equivalent from organizations such as the FDA or EFSA.

Recycling can be performed indefinitely or sometimes termed “infinitely” in many material systems. Infinite recycling refers to a process where the material used to manufacture a product can be broken down and reused
again to remake the same or similar product and then go through this process of remanufacturing for a subsequent cycle repeatedly without significant changes in properties or performance. Some losses are expected from cleaning and reprocessing, so a fraction of new material must be added to keep the recycling loop going. PET is a polycondensation polymer with an equilibrium polymerization state that can be shifted back up to longer chains after scission. This capability sets it aside from many other polymers. The main concern is to limit contamination in order to keep the process at a lowest energy cost. More contamination requires more energy to clean and remove.

One way to evaluate the efficiency of the recycling system is to evaluate the fraction of new material required to remake the product. In the case of PET, bottles can be remade completely from rPET. However, some material is lost in real processes. For example, cleaning can create small losses. There is also a possibility of a shift in the average molecular weight from Solid State Polymerization (SSP) or the presence of additives such that a small amount of new polymer can improve or stabilize successive manufacturing cycles. The practical limiting boundaries in industry are not fully characterized. It is likely that adding a small fraction (<5%) of new material to subsequent manufacturing steps is sufficient to keep recycling indefinitely, but on the industrial scale where PCR quality and production methods vary, some variation in this limit would be expected. Being able to use bio-sourced resin for the new material is an ideal scenario for minimizing environmental impact. Some have assumed a relatively low fraction of 35% for use of rPET in products. This is a reasonable practical value when considering the availability of rPET but should not be considered a limit for evaluating the long-term value of recycling PET or improving the carbon footprint of manufacturing as fractions above 50% are common including some manufactures producing using only rPET. As material becomes increasingly available due to improved collection, sorting, and cleaning methods, the practical value will increase. It is anticipated stricter state policy as considered in Europe, California, and elsewhere will help increase the available rPET. Careful consideration of the relevant infrastructure and how quickly it can react to new policy is necessary for this to be successful.

It should not be surprising that PET can be infinitely recycled. There is however little information in the scientific literature. For example, the recent book on Recycling of Polyethylene Terephthalate, Forrest, Martin J., 2019 has little to say on the topic. Forrest did reference one paper on cycles of recycling, but the methods in that paper did not include an SSP step to rebuild the IV. SSP is a common industry practice. It recovers the original chain length of the molecule and provides an opportunity to decontaminate recycled material. There are also common filtration and cleaning methods that may have been overlooked in the literature. Even in 2004 R. Franz indicated, “PCR PET materials and articles produced by modern superclean technologies can be considered to be safe in direct food applications in the same way as virgin food-grade PET”. Industry continues to improve on the ability to clean and use rPET at higher fractions. There are unpublished industry studies that show no limit on PET recycling, at least when there is a fraction of vPET included. For example, in one earlier study 50% rPET was judged infinitely recyclable, but 100% showed some difficulties in later cycles. As this was performed several years ago, before some of the more modern methods of processing rPET was available, an even greater performance at high fractions is to be expected. vPET fraction is an important detail for any study, as this can be tested in the lab and prove that fractions exceeding 50% can be sustained indefinitely. Researchers are working on new developments that will help improve the stability of PET during recycling as the industry builds up enough supply to increase the rPET fraction. As one example, an early version of chain extender technology was patented in 2002, and the work has substantially improved since then, even as existing cleaning, melt filtration, and related technologies are being refined increasing the overall quality and quantity of rPET.

Much of the additional published work is from before 2004. From this literature it is at least clear that without solid state polymerization (SSP) and without mixing virgin PET (vPET), recycled (remelted) rPET is still acceptable at 100% after 7 cycles. The authors indicate that the first injection molding step shows the greatest change in properties. This is also well known in industry and supports that recycling should have no limit based on chain length when using modern recycling equipment that repairs the polymer chain by SSP and filters out contaminants. For example, “The study of all properties showed a significant loss after the application of the first reprocessing step, thus showing a sort of threshold for PET to be recovered by further mechanical recycling”, but this was without SSP and at 100%. It is unremarkable that earlier investigators did not find a driving force to explore the already satisfactory performance of >50% rPET fractions when modern recycling methods including SSP are involved in reprocessing of the rPET.

Mechanical PET recycling technologies have evolved to the point that bottle-to-bottle recycling is now possible with mechanically recycled PET (rPET). What appears often in popular media are photos of PET, not because it is the most offensive material of a class, but it becomes easy to photograph as it is a commonly collected and recycled polymer. Bottles are also the typical product
thought of as recyclable, but recyclers are adding capabilities to recycle many products—even carpet. What was once material only useful for recycling into less demanding applications is now often useful for reuse for its original application, even with the lower energy recycling methods. However, depending on the quality of the collection system and source material, some fraction of rPET can exhibit degraded physicochemical and optical properties as a result of random chain cleavage, oxidation, or cross-linking. In addition, current mechanical recycling technologies must sort out mixed, colored, and contaminated PET waste.

A significant fraction of the PET recycling literature reflects on ways to consume discarded PET in down-cycled applications. These may be useful in the short term, but a best use is to recover PET that cannot be immediately mechanically recycled and restore it for use with advanced recycling. To complete a circular life cycle for more difficult to recycle waste, integrated and selective advanced recycling has been acknowledged as a promising solution to augment and overcome the limits associated with mechanical recycling. Advanced recycling can enable conversion of discarded PET—often colored flakes or fabric—into valuable products including monomeric precursors that are used for repolymerization into high-quality rPET or the production of different products of higher value for other supply chains. This ability to fully close the material reuse loop with PET is unusual among materials and alleviates many environmental concerns.

As referenced earlier, Zimmerman also underlines the value of enzymatic degradation for mixed waste and composite or multi-layered materials that are otherwise difficult to recycle. This is a key future technology to aid in the circular economy. Advanced recycling will become increasingly more efficient than it is today. Polymerizing directly from BHET, for example, is lower energy than TPA and EG.

Frank Welle provided an overview of the global progress of bottle-to-bottle PET recycling. He concludes that the use of PCR PET into new packaging applications has not yet led to a significant cost reduction of the PET bottle manufacturing process. Recently, supply is predicted to fall behind demand and the price of rPET remains close to PET even though the energy cost to produce it is significantly less. Before 2019, there were cases when rPET was favorable in price to PET, but recent regulation and tightening of supply has erased those economic advantages. Welle indicates recycling is driven by the publics’ concerns about the environment coupled with the business sector’s embracing of sustainability ethics. Cost reduction is therefore not yet a main driving force for PET bottle-to-bottle recycling. However, the recent increase in demand has a potential to help the recycling industry scale such that economic advantages of recycling will also follow. When new producers can switch into PET due to its favorable properties, then that also increases the supply of material to be recycled as well as begins to reduce the variety of materials competing to be sorted at material recovery facilities.

Along with packaging applications, polyethylene terephthalate (PET) is relevant as a fiber forming polyester. It is extensively used in textile and clothing sectors due to its high strength, good wear resistance, excellent dimensional stability, chemical stability, abrasion resistant and wrinkle-free characteristics. Therefore, the challenge lies in finding a suitable way of reusing or recycling fabric waste. Advanced recycling area of research has reached a new level of maturity. It offers improved pathways to recover PET from contaminated waste streams. Early methods were energy intensive and later abandoned due to costs. Presently several advanced recycling technologies under intense development. Some have had limited success. These technologies often require expensive catalysts, use harsh conditions, or produce deleterious waste streams. However, some recent research shows promise for a more efficient and cost-effective method for advanced recycling. For instance, some processors are developing a solvent-based process using KOH as the catalyst with improved product selectivity. However, this technology may require additional development to be effective. Continued research is addressing challenges with expectations of commercially attractive solutions within this decade. It will follow that material collection technologies would also need to improve as materials that were previously discarded, such as mixed fibers, some thermoformed articles, and heavily mixed or dark colored plastic would be valuable for recovery and upcycling of PET. Until the proper recycling infrastructure is in place, converting PET into a resin that degrades potentially reduces its value as the carbon that was available to be recycled back to PET is, after conversion, lost to a form that eventually converts into CO₂. Once the recycling capability is available, the environmental value of such biodegradable resins would increase. Therefore, efforts on their development as well as consideration for their detection at material recovery facilities and recovery in general must be considered.

4.5 | Bio-PET

4.5.1 | Discussion relevant to bio-PET with context of sustainability including the role of recycling and results from section 3.6 in the context of sustainability

Two primary challenges with replacing current comonomers with renewable alternatives are (i) the renewably sourced comonomers should be available at
industrial scale with reasonable cost (ii) the comonomers should exhibit improved or at least equivalent performance compared to existing comonomers. When comparing PET to other materials that are bio-degradable, one of the major challenges that stands out even in the most recent studies is complications in recycling of the biodegradable food packaging material. As we have seen above, recycling is the leading common factor in reducing the environmental impact of all packaging materials. This has many contributing factors, but the existing infrastructure that is optimized for recycling commodity polymers is significant and valuable. Obviously, bio-based PET would be recyclable in the same existing infrastructure.

Applications that have a long service life, while important to optimize, are less time critical as their environmental impact is delayed and industry can learn from solutions developed for other shorter lifetime products. Fiber for clothing and sheet and film for packaging are two shorter lifetime example areas. These should both be treated with a full context in mind. The focus here is on polymers used in packaging.

Plastic packaging can be further divided into flexible and rigid types with overlap becoming more common between these areas as the rigid types become thinner. With the rise of polymer packaging, glass has been displaced by several polymers, PC, PE, PET. In several studies, the assumption of transportation of the container skews the analysis such that all the polymers including PET are over penalized. Along with recycling, a significant decrease in energy is possible with self-manufacture and filling of PET containers, avoiding the transportation of empty containers. Some manufacturers ship empty containers, but making containers at the site of use is becoming increasingly common, yet not accounted for in most studies. In some recent cases, PE has been displaced by PET. As this increases, one advantage is more uniform waste material for recycling. This displacement generally occurs when the costs are favorable and often depends on large scales to drive change.

Both polymers have also seen technical improvements and equipment manufacturers have continuously improved processes making them more efficient and reducing costs. An example is injection molding giving a 60% improvement in efficiency with recent improvements in machines, motors, and process methods. Improvements in blow molding efficiencies have also seen opportunities for up to a 30% savings over the last decade. Being able to manufacture packaging on site leads to environmental and cost savings, but is difficult to do with some materials such as glass and aluminum, so that continued displacement of these packaging materials is likely for economic and sustainability reasons. Several other displacements for alternative material selection have been proposed. Applications using bio-based polymers is also a broad category with some materials already showing less value than originally anticipated, but many new opportunities are still in development.

Future displacement of some packing materials for environmental benefits should continue, and careful analysis is required to select a best path forward keeping regard for a range of geographic and other factors. Furthermore, it is beneficial for such an analysis to take into account the trajectory of innovations and improvements rather than rely on the limitations of existing technologies or even outdated methods of production or manufacture. As has been discussed elsewhere, there is also an environmental cost to retooling an industry. Being able to use existing production equipment for lower carbon footprint or even new or future carbon negative material solutions, is another value afforded by options such as Bio-PET.

This review shows context for developments in the versatile polymer, PET. PET can be remade through mechanical and advanced recycling. New designs and manufacturing methods allow lighter weight containers. As illustrated in Figure 9, combining this with recycling, bio-PET, and developing beneficial additives further lowers the CO2 required to manufacture products along with benefiting other categories considered in LCAs.
5 | CONCLUSION

Publications on PET have continued to increase exponentially over the last five decades. More recently, the number of publications has increased on recycling with advanced recycling taking the majority, corresponding to development in academic and research laboratories. PET is a versatile material with some peculiar traits including the ability to be remade from its polymer state through mechanical recycling combined with solid state polymerization and even back to its original monomer through advanced recycling. The scale of PET’s use has afforded continued research and practical applications in improved forms of recycling both in the lab and industry. Publications on new uses of discarded PET and the ability to clean and convert it into many forms including alternative materials are expanding. Some of these can be classified as upcycling. Others should be labeled as downcycling. However, both are an attempt to extend or improve the end of life for polymer materials. Being able to clean downcycled PET for upcycled use in medical and food grade applications is particularly useful for conservation of resources.

Literature review confirms, PET is the lowest environmental impact alternative over glass. With on-site manufacturing, where transportation of the empty packaging is not needed, energy use is much lower than alternatives. PET also uses less energy than aluminum and is comparable to many other polymers for most manufactured part and packaging applications. While well accounted for in industry, it is important to correctly manage oxygen and contaminants when recycling PET also in the laboratory. Comparatively less research is published on mechanical versus advanced recycling. This may be due to the straightforward nature of mechanical recycling as a solution already implemented in industry. A continued increase in recycling from all types inevitably lowers the environmental impact required to manufacture products.

The future state for PET has potential to reduce energy demands even further with the largest breakthroughs in recycling technologies and bio-sourced resins trending toward carbon negative solutions. Once bio-PET is competitively priced and carbon negative, it becomes the material of choice for packaging applications alongside rPET. Increasing the scale of use for PET improves the fraction of material in the recycling stream, which further improves the ability to sort and reuse the material. This is itself beneficial for the environment even with fossil fuel-based polymers due to the aforementioned lower carbon footprint. When adding in the possibility to sequester carbon with bio-sourced resins, acceleration of the environmental benefit for selecting PET is significant. The two categories, recycling and bio-sourced PET, can be further broken down into sub-categories. For recycling, there is mechanical and advanced recycling as well as new sorting and collection innovations. For bio-sourced resins, there are many new developments for raw materials and existing patents for technologies already beginning to scale with new ones not far behind. The first commercial example of bio-PET used bio-sourced ethanol as one of the raw materials.

Opportunities remain for improvement in the use of PET as light weighting is not fully implemented in many applications. The rPET fraction for some applications has been demonstrated at 100%, yet the availability of material is limited, so a focus on increased rPET availability is germane. Advanced recycling and increasing the collection percentage are two areas for continued development. In addition, for some applications, developments in additives or other methods to improve barrier properties are necessary for PET to outperform alternative materials such as aluminum, yet remain optimal for mechanical recycling. Advanced recycling will free up more additive options for continued application development and help them remain recyclable.

Research and industry developments continue with a focus on improving material performance, cost, and environmental impact. Often this is driven by raw material costs and supply factors. Consumer sentiment and public policy can also be driving factors. The testing of new resins, development of bio-feedstocks, improvements in engineering, processing, recycling, and design will continue. The above review provides some context for these developments. Improving the full life cycle, reducing usage without sacrificing performance through improved design, taking full advantage of recycling methods while supporting their further development, moving forward with carbon negative bio-materials, and new additives all work together to provide increased sustainably performance.

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CONFLICT OF INTEREST
The authors state no direct conflict of interest. The corresponding author does have patents and has performed consulting or other work for hire in industry related to polymers including PET and their composites.

DATA AVAILABILITY STATEMENT
Data sharing is not applicable to this article as no new data were created or analyzed in this study.

ORCID
Jay C. Hanan https://orcid.org/0000-0002-2583-2079

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AUTHOR BIOGRAPHIES

Parikshit Sanjay Sarda is a doctoral candidate at the Polymer Institute in the University of Toledo. His research is focused on development of sustainable technologies to separate and upcycle plastic waste and synthesize bio-based monomers for plastic manufacturing. Parikshit has a Master’s degree from West Virginia University where he worked in Process Simulation, Control, and Modeling. Parikshit has also worked in India’s largest refinery as a Process Engineer where he looked after capacity enhancement, heat integration, and other debottlenecking projects.

Jay C. Hanan is an Associate Professor of Mechanical and Aerospace Engineering at Oklahoma State University running a laboratory out of the Helmerich Research Center in Tulsa, OK. He has also spent much of the last 17 years as a consultant to industry including manufacturers of Polyethylene Terephthalate parts primarily for packaging. He has over 200 patents and journal publications with an emphasis on structural materials and composites including polymer nanocomposites.

Joseph G. Lawrence is the Sr. Director of the Polymer Institute and Center for Materials and Sensor Characterization (CMSC) at the University of Toledo, Toledo, Ohio. He is also a Research Professor in the Department of Chemical Engineering. Dr. Lawrence has 15 years’ experience in research in the fields of polymers, nanomaterials, composites. He has 3 years’ experience in process industry managing operations, planning and workforce. Dr. Lawrence has served as principal/co-principal investigator for...
research projects funded by various agencies. He advises/co-advises graduate students and has authored 1 book chapter, 30 publications and 35+ conference presentations.

**Masoud Allahkarami**, received his bachelor and Master degrees in solid state physics and worked as researcher at the Material and Energy Research Center (MERC). His Ph.D. at Oklahoma State University was focused on new method development for material characterization. His interest is in development of new material characterization techniques. He has worked as postdoc at OSU and research scientist at Niagara bottling. His current research has been related to diffraction and tomography of PET polymers with applications in the bottling industry.

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