

Shear Assisted Processing and Extrusion (ShAPE) of Plastics: Recycling and Remolding

September 2023

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under Contract DE-AC05-76RL01830

Printed in the United States of America

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Prepared for
the U.S. Department of Energy
under Contract DE-AC05-76RL01830

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Abstract

Polyethylene (PE) and polypropylene (PP) are often disposed as mixed plastic wastes. The challenges in recycling and upcycling these mixed polyolefin wastes lie in the difficulty in separating individual constituents in a cost-effective and scalable manner. Direct recycling the mixed PE and PP wastes in conventional melt-phase extruders typically result in a product with poor properties and low added value, because of immiscibility, phase separation, and lack of crystallinity. Friction extrusion (FE), a solid phase processing technique that has successfully extruded metal matrix composites with desired end products, has never been utilized to address the issue of recycling mixed plastic wastes. In this study, FE was performed on single-stream low-density polyethylene (LDPE), single-stream PP and mixed-stream LDPE+PP. Consolidated filaments of 2.5 mm diameter were extruded from different precursors. The thermal, infrared, and microscopic properties of extruded filaments were measured to evaluate the effects of FE process on the structure of recycled polymers. Meanwhile, the energy efficiency of FE was estimated based on extrusion rates and compared with conventional melt extrusion processes.

Acknowledgments

This research was supported by the Strategic Investments I3T Commercialization Program, under the Laboratory Directed Research and Development (LDRD) Program at Pacific Northwest National Laboratory (PNNL). PNNL is a multi-program national laboratory operated for the U.S. Department of Energy (DOE) by Battelle Memorial Institute under Contract No. DE-AC05-76RL01830.

Acronyms and Abbreviations

ShAPE- Shear Assisted Processing and Extrusion

FE- Friction Extrusion

PE- Polyethylene

PP- Polypropylene

ABS- Acrylonitrile Butadiene Styrene

DSC- Differential Scanning Calorimeter

FTIR- Fourier Transform Infrared

ATR- Attenuated Total Reflectance

SEC- Specific Energy Consumption

PCL- Performance Characteristic Line

QS- Quickstarter

Contents

Abstract.....	ii
Acknowledgments.....	iii
Acronyms and Abbreviations.....	iv
1.0 Introduction.....	1
2.0 Experiments	3
3.0 Results and discussion.....	5
3.1 Friction extrusion filaments	5
3.2 Effects of FE on thermal properties of polymers.....	5
3.3 Energy analysis.....	8
4.0 Summary and conclusions.....	11
5.0 References.....	12

Figures

Figure 1. Conventional mechanical recycling processes	1
Figure 2. (a) Thermoplastic polymer precursors: polypropylene (PP) pellets and low-density polyethylene (LDPE) pellets, (b) Experimental setup of friction extrusion.....	3
Figure 3. (a) Friction extrusion thermoplastic PP+LDPE filaments; (b) mixed PP and LDPE pellets precursor; Zoom-in pictures: (c) PP FE filament, (d) LDPE FE filament, (e) PP+LDPE composite filament.....	5
Figure 4. Heat capacity of LDPE during reheating and cooling steps for raw pellets and single-stream friction extruded samples.....	6
Figure 5. Heat capacity of PP during reheating and cooling steps for raw pellets and single-stream friction extruded samples.....	6
Figure 6. (a) Melting behaviors, (b) Crystallization behaviors of friction extruded LDPE+PP composite filament and single-stream PP and LDPE filaments measured on DSC during the reheating step.....	8
Figure 7. Performance characteristic line (PCL) model of conventional extrusion.....	10

Tables

Table 1. Thermal characteristics of LDPE and PP before and after friction extrusion measured on DSC. Numbers in parenthesis are standard deviations.....	7
Table 2. Thermal characteristics of LDPE and PP phases in the LDPE+PP composite filament compared to single-stream extrudates measured on DSC. Numbers in parenthesis are standard deviations.....	8
Table 3. Specific energy consumption (SEC) of conventional plastic extrusion machine or extrusion site.....	9

Table 4. Machine SEC estimated for extruding different plastics at commercial scales	9
Table 5. SEC of ShAPE process (experimental data) and SEC of conventional plastic extruders (estimated from the PCL model).....	10

1.0 Introduction

Plastic waste is an urgent global issue for environmental protection and sustainable energy. There are three main issues of recycling mixed plastic: (1) it is energy-inefficient and inconvenient to sort and remelt end-of-user mixed plastics, (2) it is impossible to separate multi-phase plastics into single phases (e.g., separate a single-use multi-layer plastic bag). [1-3] (3) the current recycling processes degrades the properties of plastics. [4] These issues devalue the plastic and also result in an unfavorable cost of recycling and reuse plastic waste compared to using newly made plastics.

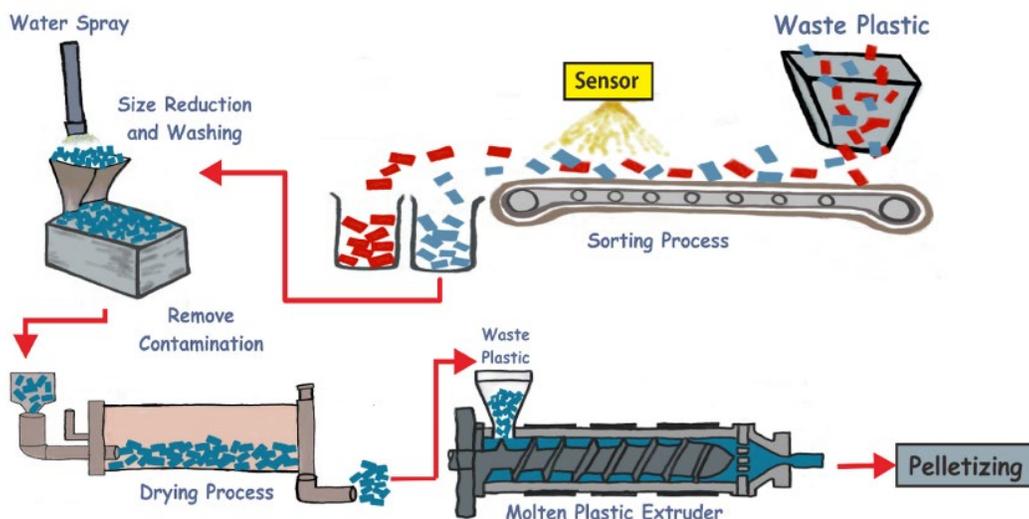


Figure 1. Conventional mechanical recycling processes

A potential new recycling avenue is to convert the mixed plastic, e.g., Polypropylene (PP) and Polyethylene (PE) into a “new” plastic directly. However, the conventional processes have limitations on mixing and homogenizing the multi-phase plastics without degrading the properties due to their temperature mismatches. ShAPE is a unique technology that consolidates and homogenizes plastics via shear thinning and mixing in the solid state. [5, 6] From a previous Quickstarter (QS) project have demonstrated that ShAPE can make solid rods and discs directly from mixed plastic wastes in a single step. [7] Preliminary calculations regarding energy cost from have shown that using ShAPE to recovery mixed plastics conserves at least 25% energy compared to making new plastics, and potentially saves energy compared to conventional recycling process involved with sorting and remelting because ShAPE avoids preheating, melting, and incinerating. Additionally, the QS ShAPed plastic rods have shown tensile strength was retained up to 80% of the “rule-of-mix” strength of virgin plastics at the same proportions. However, the existing toolings designed for processing metals have several limitations when applied to plastics. For example, a longer die landing is anticipated to make the extrudate straighter, and a higher extrusion ratio with a concave face die can enhance the mixing of plastics. Moreover, a longer billet will improve the energy efficiency by enabling a longer steady state, which needs less power. In addition, the molecular weight, heat flow curve and microstructure of the ShAPE recycled plastics have not been investigated due to limited funding in the QS project. It is essential to obtain this information to better understand the potential application of the ShAPE recycled material. Also, the mechanical properties have not been verified with an enough number of standardized samples.

Friction extrusion (FE) is a low-energy, solid phase processing technology [8, 9]. FE does not require an external heat source to heat the material as opposed to traditional melt-phase extrusion. Instead, it uses friction between the rotating tool and the precursor material to generate heat [10]. The benefits of FE, such as low processing temperature, low energy consumption, and rapid production of composite from blend powder, are extensively demonstrated in the consolidation and forming of metal materials [11-15]. Apart from metal materials, similar principles of “friction stir” have been applied by several researchers to weld and process polymers in prior studies [16]. For example, Clark developed tooling for joining thermoplastics [17]. Kiss studied the ability of friction stir welding on joining polypropylene and concluded that the material flow is critical to the weld strength [18]. After that, friction stir welding or processing has been applied to various types of polymers, including PP [19], PE [20], acrylonitrile butadiene styrene (ABS) [21], and fiber-reinforced polymer composites [22, 23]. However, the effect of friction-based solid phase extrusion on polymer properties on both micro and macro levels has not been fully investigated [24, 25]. Obtaining a thorough comprehension of the structural effects of the process is critical to tackle issues that are associated with product performance, to optimize solid-phase processing procedures, and to improve the processibility of mixed plastic wastes in their solid states.

The low processing temperature of FE can reduce the energy cost of polymer extrusion [26] and therefore reduce the risk of polymer degradation. Additionally, high shear-induced mixing and homogenization could be a potential avenue to recycle mixed stream polymers into new materials [27]. To the best of the authors' knowledge, no prior research has been conducted on the use of FE for polymer processing. This work aims to demonstrate FE as a novel method for processing thermoplastics, specifically PP and LDPE. Firstly, single stream PP and LDPE filaments were extruded from compacted pellets separately. The thermal properties were characterized and compared with the raw materials to assess the effect of the FE process. Next, a mixture of PP and LDPE pellets was compacted and extruded by FE into a composite filament. Thermal and microscopic testing were performed with a focus on miscibility of LDPE and PP. Lastly, the energy consumption of FE was determined from machine data obtained during a single-stream extrusion of LDPE as a representative batch. At industrial scale, the energy consumption difference caused by material type is minimal compared to that caused by process load. The potential energy saving for LDPE is demonstrated by comparing the theoretical scaled value for FE with literature data for conventional extrusion in an industrial environment.

2.0 Experiments

Two types of thermoplastic materials were used in this study, see Figure. 2(a). One is PP blue pellet (TOTAL Polypropylene 4944CWZ, made by PROSPECTOR®). The density (0.905 g/cm³) and melt flow rate (50g/10min at 230°C and 2.16kg load) of PP pellets are provided by the manufacturer. The other constituent is LDPE pellet appearing transparent and white (AT Plastics 194 10-201321). The melt flow rate is measured on a melt flow indexer (Hanatek Model 4050) to be 16.26g/10min at 98 °C, following ASTM D1238. The density is 0.892 g/cm³ measured on Mettler Toledo XPR205 by comparing the mass of sample immersed in ethanol to the mass in air, referring to ASTM D792. Single stream PP, LDPE and a mixture of PP and LDPE pellets with 1:1 volume ratio were employed as precursor in this study. Figure. 2 (a) Thermoplastic polymer precursors: polypropylene (PP) pellets and low-density polyethylene (LDPE) pellets, (b) Experimental setup of friction extrusion.

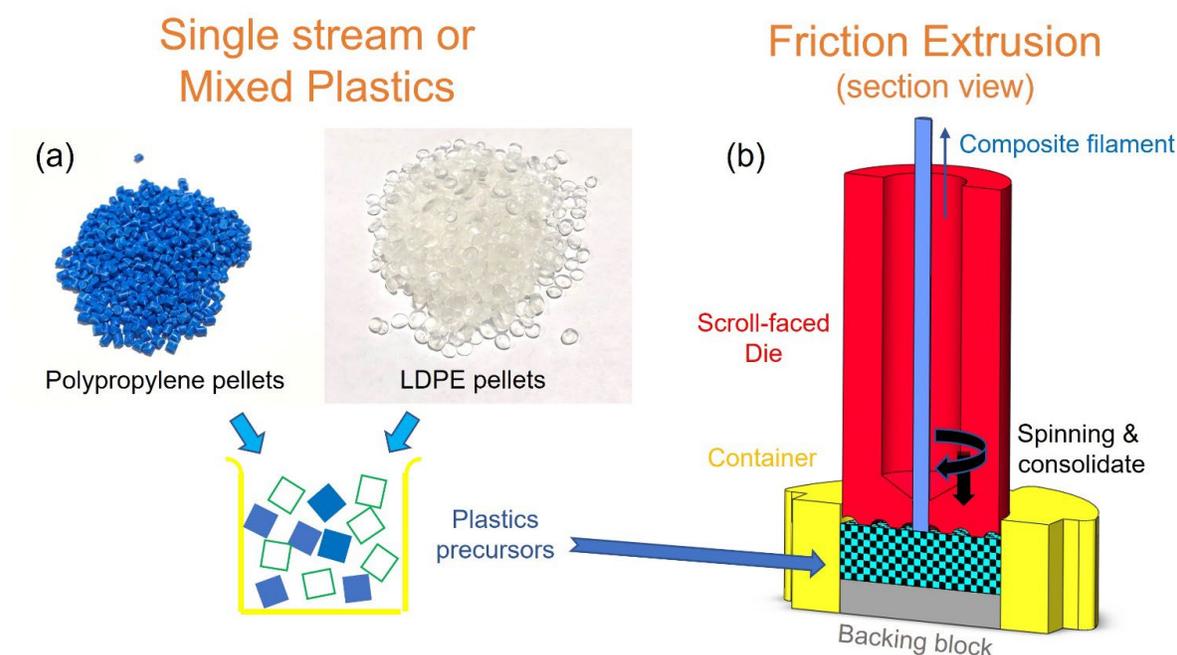


Figure 2. (a) Thermoplastic polymer precursors: polypropylene (PP) pellets and low-density polyethylene (LDPE) pellets, (b) Experimental setup of friction extrusion.

The setup for friction extrusion is illustrated in Figure. 2(b). It consists of the following tools: a scroll-faced extrusion die with a 100:1 extrusion ratio, a ring-shape precursor container with 25.4mm inner diameter, and a backing block to support the precursor in the container. Both container and die were made by H13 tool steel. A K-type thermocouple was inserted into the die face to measure the processing temperature during experiments. The Shear Assisted Processing and Extrusion (ShAPE™) machine was employed to perform the extrusion horizontally with controlled die rotational speed and die feed rate. Three types of precursors were processed in different batches: single stream PP pellets, single stream LDPE pellets, and the mixture of PP and LDPE pellets with 1:1 volume ratio. Before processing, the precursor pellets were loaded into the container to full capacity. Then the container was installed on the ram side of the ShAPE machine. Unlike conventional plastic extrusion, preheat was not required

for FE process. The die was installed on the spindle side and aligned with the center of the container as well as the central axis of the spindle. During processing, the rotating scroll-faced extrusion die was driven by the spindle and pressed against the precursor material in the container. The precursor was compressed, deformed, and heated under the friction at the die-polymer interface. Under the combination of high shear deformation and extrusion pressure, the polymer pellets were strained, mixed and then consolidated in the deformation zone at the die tip. A continuous filament was extruded through the orifice of the die. The die rotational speed and feed rate were controlled and recorded by the ShAPE machine. In this study, 400 rpm die rotational speed and 12 mm/min die feed rate were used to initiate the consolidation and heat generation at the start of the process, 80 rpm and 12 mm/min die feed rate was used at the steady-state extrusion stage. The instantaneous power input was recorded by machine and used to calculate the energy consumption. Due to equipment limitation, post-extrusion cooling system was not equipped on the machine in this early-stage study. The machine power data of single-stream LDPE extrusion process was used to determine the range of energy consumption of FE of thermoplastics in later section.

Melt behaviors of raw pellets and extruded filaments were measured on a DSC (TA Instruments Q2000) pre-calibrated with sapphire and Indium standards. A flat piece of specimen was cut from pellets or filaments along the cross-section, weighed, and sealed in a Tzero pan. Specimens were subjected to a cyclic heating-cooling-reheating protocol at 10 °C/min between 0°C and 200°C, under 50 mL/min N₂ flow. Microscope Miscibility of the PP and LDPE phases of extruded composite filaments was examined on an FTIR imaging microscope (Bruker LUMOS II) equipped with a TE-MCT detector and ZnSe optics. A flat surface was cut while a 1-inch section of filament was clamped on a sample holder (ST Japan MicroVice STJ-0116-SL30). After taking an optical microscopic image, 5~10 locations were chosen from the field of view for FTIR measurement. The aperture for each location was 100 μm to obtain sufficient FTIR signal-to-noise ratio. The FTIR data were taken for all chosen locations with attenuated total reflectance (ATR) from 4000 to 600 cm⁻¹ wavenumber as an average of 64 scans. FTIR spectra were compared with polymer library entries in Bruker OPUS software.

3.0 Results and discussion

3.1 Friction extrusion filaments

Friction extrusion thermoplastic filaments made from single stream PP pellets, single stream LDPE pellets, and mixture of PP and LDPE pellets are presented in Figure 3. Continuous filaments of 2.5 mm diameter and over one meter in length have been successfully extruded. As shown in Figure 3 (a)(c)(d)(e), all filaments have relatively smooth surfaces and are generally straight overall but with some twist. The twist happened when the filament rotated with the die after it exited the die throat. To avoid any twist, post-extrusion cooling system will be equipped in the future so that extruded filaments can be quenched and maintain a rigid shape. Another strategy to address the twisting issue is to use a different setup that the billet rotates but the die does not. From Figure 3(a) and (e), the overall color of the PP+LDPE filament is blue (from PP) with some localized white spots, implies a sound mixing of blue PP and transparent/white LDPE. The measured die face temperature at the steady-state of FE ranges from 130-140°C.

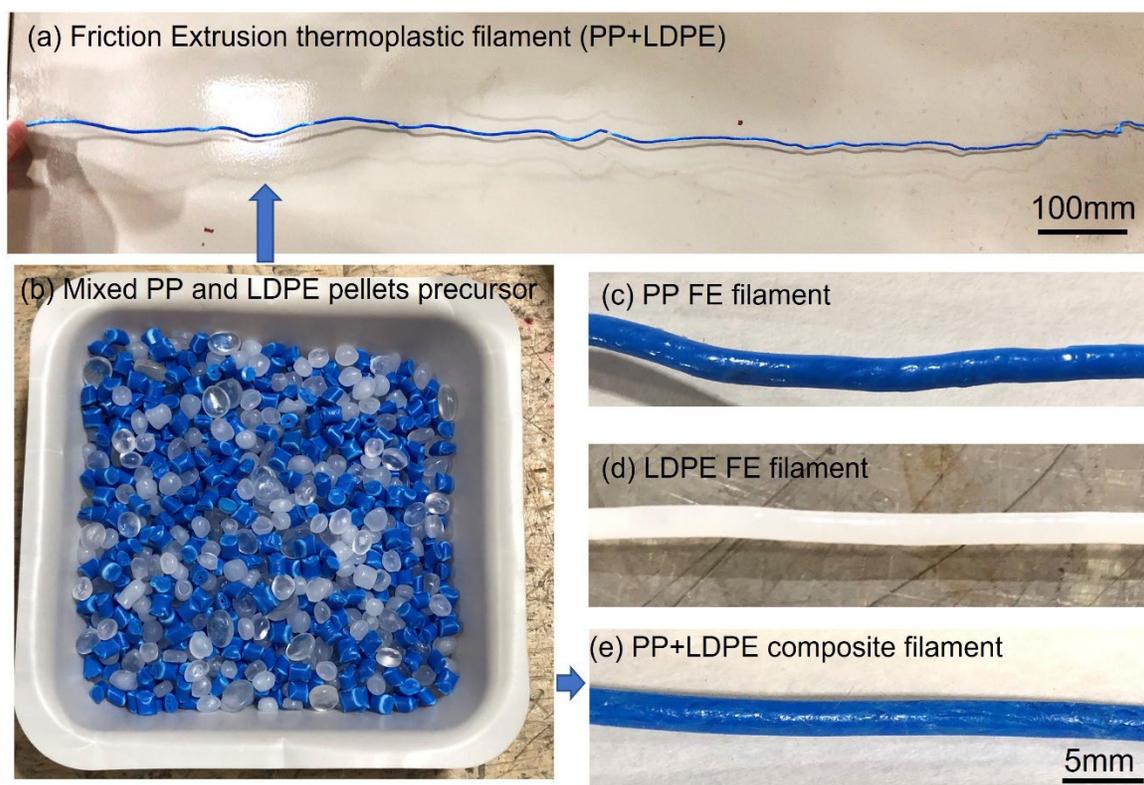


Figure 3. (a) Friction extrusion thermoplastic PP+LDPE filaments; (b) mixed PP and LDPE pellets precursor; Zoom-in pictures: (c) PP FE filament, (d) LDPE FE filament, (e) PP+LDPE composite filament.

3.2 Effects of FE on thermal properties of polymers

The heat capacity signals during reheating and cooling steps are displayed in Figure 4 and Figure 5 for precursor pellet of LDPE and PP, and extruded wire of LDPE and PP respectively. Melting temperature (T_m) and crystallization temperature (T_c) are obtained as the peak

temperatures during reheating and cooling steps. The heat of fusion (ΔH_m) and heat of crystallization (ΔH_c) are the peak areas where the baselines are extrapolated from high-temperature portions of heat capacity curves. Degree of crystallinity is calculated as $\chi = \Delta H_m / \Delta H_m^0$ where ΔH_m^0 is the heat of fusion for 100% crystallized LDPE (293.57 J/g [28]) or PP (146.44 J/g [29]).

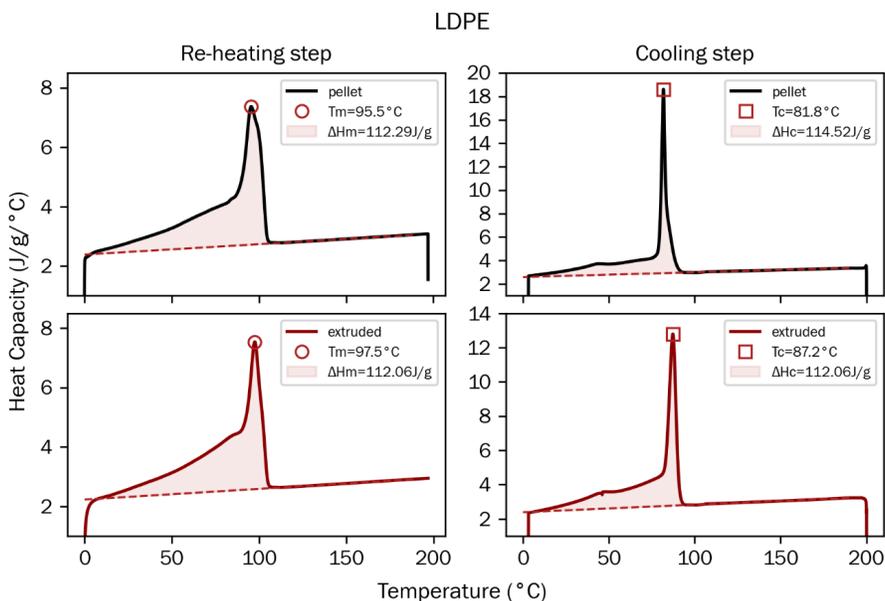


Figure 4. Heat capacity of LDPE during reheating and cooling steps for raw pellets and single-stream friction extruded samples.

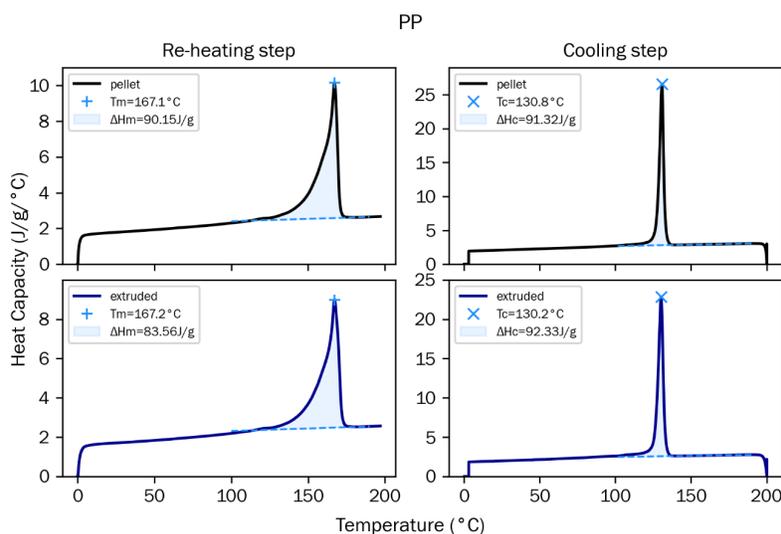


Figure 5. Heat capacity of PP during reheating and cooling steps for raw pellets and single-stream friction extruded samples.

Effects of friction extrusion process on thermal characteristics of LDPE and PP are different. T_m and T_c of single-stream friction extruded samples of LDPE increased by 2°C and 6°C respectively as shown in Figure 4 and in Table 1, while the T_m and T_c of PP did not change after friction extrusion (Figure 5 and Table 1). The increase in T_m and T_c of LDPE is consistent with “shear modification” effects where the LDPE with shearing histories exhibited lower storage modulus [30] and lower steady-state viscosity [31], i.e., promoted flowing. It was also pointed out in literature that the shear modification effects were reversible after long-time heat treatment, ruling out the concern of polymer degradation [31, 32]. Several theoretical explanations have been proposed for shear modification effects, presumably alignment of side chains [31] and disentanglement [32] during the shear history. As described in the experimental setup (section 2.2), the polymer interfacing with extrusion die was subjected to a large shear deformation as the die rotates. The proposed effects imposed by shearing, i.e., disentanglement or side chain alignment, could also cause crystalline perfection corresponding to an increase in T_m and T_c as observed in Figure 4 and Table 1. On the other hand, the T_m and T_c of PP remain the same after friction extrusion. Considering that the shear modification effects were reported for long-chain branched PP [33], it is very likely that the PP used in this study did not have much branching or entanglement and therefore no such effects were observed.

Another possible reason for the increase in T_m and T_c of LDPE after FE could be removal of plasticizers in the LDPE pellets during FE. When LDPE pellets were heated above its T_m in a vacuum oven, many bubbles were observed, which could come from volatile species acting like plasticizers in LDPE pellets. There was no such bubble formed in melted PP. Presence of plasticizer often reduces T_m [34]. The FE process reached 130 °C which is above the T_m of LDPE. It is possible that the volatile plasticizers were removed during FE and the T_m of extruded LDPE increased.

Table 1. Thermal characteristics of LDPE and PP before and after friction extrusion measured on DSC. Numbers in parenthesis are standard deviations.

	T_m (°C)	T_c (°C)	ΔH_m (J/g)	ΔH_c (J/g)	χ (%)
LDPE pellet	95.47	81.77	112.29	114.52	38.25
LDPE extruded	97.93 (0.44)	88.05 (0.99)	112.11 (0.9)	112.39 (1.97)	38.19 (0.31)
PP pellet	167.15	130.77	90.15	91.32	61.56
PP extruded	167.25 (0.2)	130.78 (0.53)	86.15 (2.42)	90.74 (1.49)	58.83 (1.65)

As for the peak areas, no significant change was observed for the heat of fusion (ΔH_m), degree of crystallinity (χ) or heat of crystallization (ΔH_c), for both LDPE and PP before and after FE, as shown in Table 1.

The individual phases were also detected on DSC where two melting peaks (and two crystallization peaks) corresponding to T_m (and T_c) of LDPE and PP phases were well separated as represented in Figure 6. The T_m and T_c peak temperatures of LDPE and PP phases in the composite filaments are very close to those of individual single-stream extrudates, with the differences being smaller than 1.5°C (see Table 2). The peak area (ΔH_m and ΔH_c) of LDPE+PP composite filament was significantly reduced. For LDPE, the melting and crystallization peaks only manifest as a tiny peak as shown in Figure 6 with an average ΔH_m of

19.35 J/g, much smaller than ΔH_m of LDPE pellet (112.29 J/g) or single-stream FE LDPE (112.11 J/g). There is a large deviation in ΔH_m of LDPE phase in the composite filament, being 11.28 J/g (Table 2). Given that DSC samples were cut as cross-sectional pieces from random locations along the filament, a large deviation indicates non-uniform composition along the longitudinal direction, which might be a result of gradual increase in processing temperature and partial melting of LDPE as the friction extrusion proceeds.

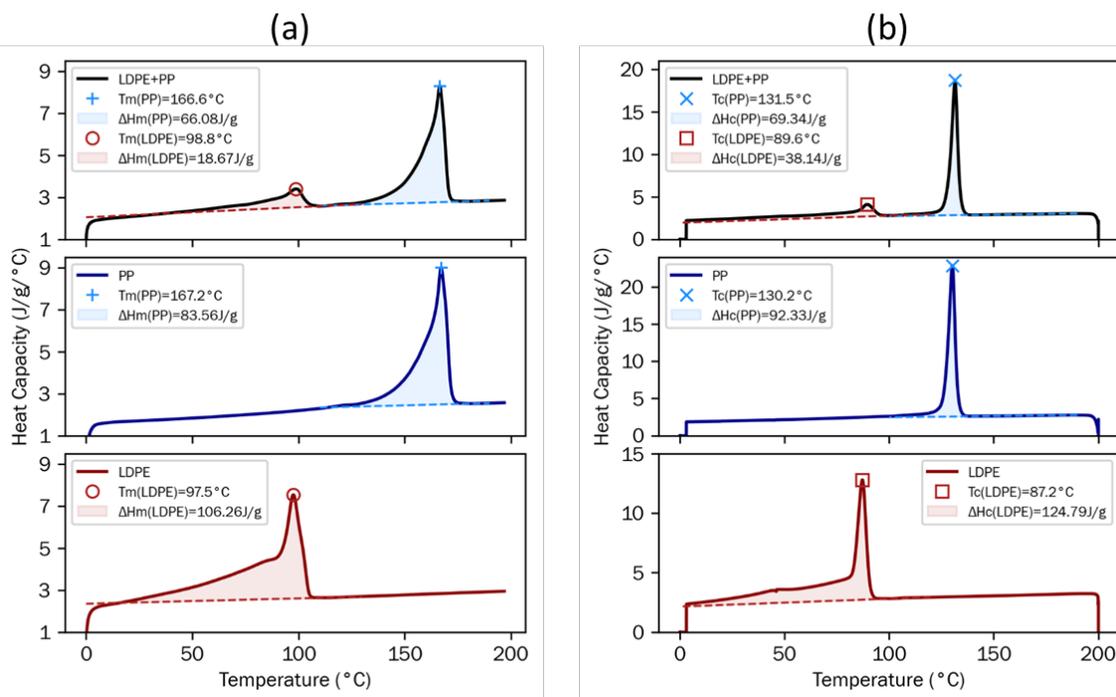


Figure 6. (a) Melting behaviors, (b) Crystallization behaviors of friction extruded LDPE+PP composite filament and single-stream PP and LDPE filaments measured on DSC during the reheating step.

Table 2. Thermal characteristics of LDPE and PP phases in the LDPE+PP composite filament compared to single-stream extrudates measured on DSC. Numbers in parenthesis are standard deviations.

	T _m (°C)	T _c (°C)	ΔH_m (J/g)	ΔH_c (J/g)
Single-stream LDPE	97.93 (0.44)	88.05 (0.99)	112.11 (0.9)	112.39 (1.97)
LDPE phase in the composite filament	98.88 (0.21)	89.55 (0.28)	19.35 (11.28)	43.2 (15.14)
Single-stream PP	167.25 (0.2)	130.78 (0.53)	86.15 (2.42)	90.74 (1.49)
PP phase in the composite filament	166.65 (0.19)	131.74 (0.29)	67.37 (9.41)	70.22 (9.24)

3.3 Energy analysis

Energy is a significant operating cost and primary greenhouse gas emission driver of conventional plastic extrusion sites. At a typical profile plastic extrusion site, approximately over 90% of total energy is consumed by the operation line (extruder motor, chiller, air compressor,

and water pump) while less than 10% of the total energy consumption supports administrative and other activities including offices, heating and lighting [35]. Therefore, energy analysis is key in research and development of plastic extrusion process toward improving both production cost and environmental impact. Energy consumption of plastic extruders is strongly depended on plastic materials at small scales and on production scales at large industrial scales. Specific energy consumption (SEC, energy per product mass) of industrial scale plastic extruders are summarized in Table 3 and Table 4. At commercial scales, the machine SEC is not heavily dependent on the type of plastics as shown in Table 4.

Table 3. Specific energy consumption (SEC) of conventional plastic extrusion machine or extrusion site

	Machine SEC (kWh/kg)		Site SEC (kWh/kg)	
Average	0.53		1.506	1.316
Source of data	Tangram*		EURecipe†	Tangram*

* Collected by Tangram Technology Ltd. on 91 extruders from 48 extrusion sites throughout the world [35]
† EURecipe report from a survey of European plastics processors in 2005 [35]

Table 4. Machine SEC estimated for extruding different plastics at commercial scales

Type of plastics	Machine SEC (kWh/kg)	Source of data
General	0.4-0.7	Estimated from PCL in [35]
Semi-crystalline (e.g., PE, PP)	0.5-1.0	Estimated based on material properties and extrusion line [36]
Amorphous (e.g., PS, PVC)	0.4-0.8	

The average machine SEC listed in Table 3 were collected by Tangram Technology from 48 extrusion sites [35]. Based on the Tangram dataset, a performance characteristic line (PCL) was developed to mathematically correlate SEC and process load. The PCL from 48 extrusion sites' data is $Site SEC = 4.33 \times (Production Rate)^{-1} + 0.49$ [35] and is plotted in Figure 7 as the solid blue line. In addition, the minimum and maximum machine SEC values in Figure 7 were estimated based on a simple assumption that site SEC is approximately 2-3 times of machine SEC [35].

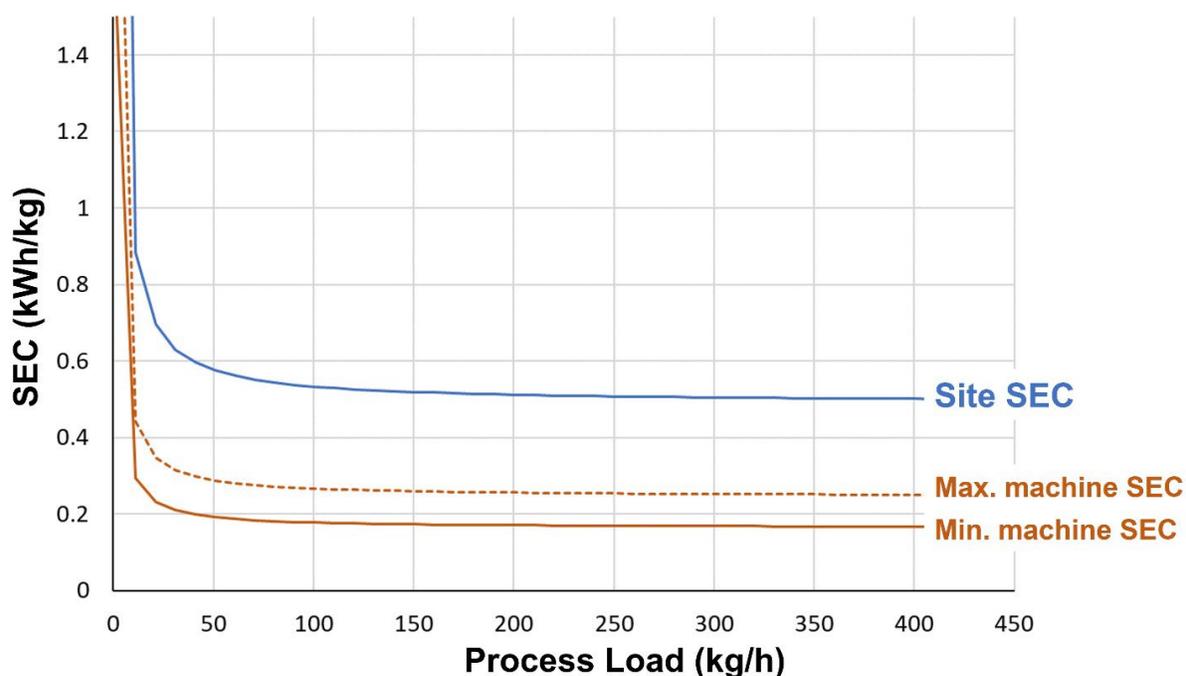


Figure 7. Performance characteristic line (PCL) model of conventional extrusion.

The PCL model in Figure 7 is used as a benchmark of conventional plastic extrusion process. Table 5 presents SECs from ShAPE machine and comparison to SEC calculated from the PCL model. ShAPE machine was operated at a maximum 0.04 kg/hr for the single-stream LDPE extrusion batch. Using the process load value, projected SEC of conventional extruder machine is approximately 33-50 kWh/hr. Note that the machine SEC of conventional extruder can decrease at higher process loads, based on the trend of PCLs in Figure 7. On the other hand, based on the energy data from experiments, energy consumption of ShAPE process for LDPE pellet is comparable at 7.1 kWh/kg of machine SEC. At 0.04 kg/hr load, the FE on ShAPE lowered machine SEC by 80% compared to the conventional plastic extruder. Although the results are only limited at low extrusion load, this energy analysis preliminarily suggests improvement of energy efficiency with FE compared to conventional melt-phase extrusion.

Table 5. SEC of ShAPE process (experimental data) and SEC of conventional plastic extruders (estimated from the PCL model).

Material	Process load (kg/hr)	Machine SEC (kWh/kg)	
		Friction extrusion, from ShAPE machine	Conventional extrusion, estimated from the PCL in Figure 7
LDPE (pellet)	0.04	7.1	33-50

4.0 Summary and conclusions

Processibility of polyolefin pellets by solid-state friction extrusion was investigated for LDPE, PP, and their blends. Filaments of 2.5 mm in diameter were successfully extruded. A slight increase in melting temperature (by 2°C) and crystallization temperature (by 5°C) of LDPE was found by comparing samples cut from the single-stream extruded LDPE filament to the raw pellets through temperature scans on DSC. This is consistent with the “shearing modification” effects reported for LDPE and long-chain branched PP and were attributed to reversible molecular changes such as disentanglement or side chain alignment. No other thermal characteristics were found significantly affected by friction extrusion process.

Consolidated filaments were also obtained from the blend of LDPE and PP pellets even though they are thermodynamically immiscible. Without using any compatibilization strategy, separated phases of LDPE and PP were observed under FTIR microscope. The size of each constituent is approximately at 0.1~10 mm scale. A lower feed rate or higher die rotational speed is required to further enhance mixing. Temperature scans on DSC of the LDPE+PP composite filament showed separate melting and crystallization peaks corresponding to LDPE and PP.

Based on the measured heat of fusion, 75.2 wt.% of PP was incorporated into the filament which is higher than expected (50.4%) presumably due to lack of mixing. Based on performance characteristic line model and process load data from extrusion process data, it is estimated that up to 80% improvement in energy efficiency can be accomplished using solid phase FE compared to conventional melt-phase processing. Improving energy efficiency of plastic extrusion/recycling process by solid phase processing could also be a cost-effective option in the near term to reduce the overall process greenhouse gas emission.

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