**PERFORMANCE OF A MODIFIED EXTRUDER FOR POLYESTER FIBER PRODUCTION USING RECYCLED PET****DESEMPEÑO DE UN EXTRUSOR MODIFICADO PARA LA PRODUCCIÓN DE FIBRA POLIESTER HECHAS DE PET RECICLADO**

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Abstract

In this study, we report the design and results of the operation of a modified extrusion equipment with spinning fiber devices. The performance of the equipment has been evaluated by producing polyester fibers from different recycled poly(ethylene terephthalate) (PET) of pharma- and bottle-grade. Flow performance along the extruder length was modeled and the pressure was calculated at standard conditions and considering a flow change of +10% in these standard conditions. The variation of drop pressure inside the extruder was modeled as a function of the Inherent Viscosity (*IV*) and humidity content (*X*). In addition, losses of *IV* were calculated in the temperature range from 290 to 300 °C for different initial PET Viscosity index (*IV*₀). We report the variation of mass flow as a function of pressure applied to the extruder at different temperature for PET recycled of pharma- and bottle-grade. The proposed model for analyzing the mass flow showed a good agreement with the experimental data showing a mean error < 3 %.

Keywords: recycled PET, extrusion process, spinning process, polyester fiber.

Resumen

En este estudio se reporta el diseño y resultados de la operación de un equipo de extrusión modificado con un sistema de hilatura de fibra. Fue evaluado el desempeño del equipo mediante la producción de fibras poliéster hechas de poli(tereftalato de etileno)[PET] (grado pharma y grado botella). Se modeló el comportamiento del flujo a través de la longitud del extrusor y la presión fue calculada a condiciones estándar y considerando un cambio de flujo de +10% en las condiciones estandar. Fue modelada la caída de presión en el extrusor en función de la viscosidad inherente (*IV*) y el contenido de humedad (*X*), así como también, la pérdida de *IV* en el rango de temperaturas de extrusión de 290 a 300 °C para tres diferentes viscosidad inherente iniciales (*IV*₀). Se presenta la variación del flujo másico del PET reciclado grado pharma y grado botella, en función de la presión aplicada al extrusor para diferentes temperaturas. Finalmente el análisis matemático del flujo másico muestra una buena correlación con los datos experimentales a nivel laboratorio, mostrando diferencias menores al 3 %.

Palabras clave: PET reciclado, proceso de extrusion, proceso de spinning, fibra.

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1 Introduction

Economically, the recycling process is the best way to reduce wastes of poly(ethylene terephthalate) (PET), therefore, many technologies have been developed for this purpose. The first effort in the world to recycle PET bottles was performed in 1977 (La Mantia *et al.*, 1994; Miller, 2007); and in the following decade, several studies were performed to analyze the properties of PET wastes recycled using extrusion processes. Several methods have been reported to obtain recycled recipients and bottles from PET. In particular, early studies focused on the use of waste PET for energy recovery (e.g., Daw-Ming *et al.*, 1996; Abu-Isa *et al.*, 1996; Saha and Ghoshal, 2005). Torres *et al.* (2000) performed a comparative study of the thermal and mechanical properties of bottles obtained from waste and virgin PET. They obtained elongation values of 200% for virgin PET and smaller values (around 10 % less) for recycled PET. These authors concluded that the cause of these results was associated to differences in the crystallinity. Oromiehie and Mamizadeh (2008) used three different methods for recycling PET bottles where processed virgin PET, recycled PET and mixtures of both types of PET, with and without the modifier polypropylene functionalized with maleic anhydride [PP-*graft*-MA]), were studied. This study reported that the intrinsic viscosity (IV) and the molecular weight (M_w) decreased with an increment in the content of recycled PET in the mixtures. This result was attributed to thermal effects and mechanical degradation of recycled PET. Besides, the properties of the functionalized blends were improved due to chemical and physicochemical interactions between components in the blend. Several studies have reported that, raw material flakes produced by the size reduction of PET waste should satisfy certain minimum requirements in order to achieve satisfactorily PET recycling (Seo and Cloyd, 1991; Cata *et al.*, 2007; Franceschini *et al.*, 2007). This is because the loss of molecular weight is presented in PET extrusion process due to hydrolytic (Franceschini *et al.*, 2007; Pirezadeh *et al.*, 2007) and thermal-mechanical degradation during melting process (Saeid Hosseini *et al.*, 2007). Degradation may also cause non-uniformity in the flow of the fused material causing negative effects in the subsequent processes and affecting the properties of final product (Abu-Isa *et al.*, 1996).

Another possibility for PET recycling is its application in the production of fibers, which can be

considered as a secondary recycling process in order to use them in other industries such as automotive industry (Gurudatt *et al.*, 2005). The melt spinning is the usual way for the production of these recycled fibers (Elarmi *et al.*, 2007; Litchfield, 2008). Note that melt-electrospinning can be used as alternative process (Lyons *et al.*, 2004; Rajabinejad *et al.*, 2009). For example, Gurudatt *et al.* (2003) obtained chips from PET waste and these chips were used for getting fiber filaments. These authors used virgin PET and waste PET from bottles in mixtures with different compositions. The analysis of the extrusion and stretching stages of the spinning process, these authors showed that the variations of molecular orientation is very important for the properties, efficiency and production of the final fiber. Abbasi *et al.* (2007) have reported that the values of crystallinity in samples of recycled materials were higher than those obtained for the virgin material. Consequently, the tenacity of these samples was high and the elongation was small. Martin and Rojas (2005) developed a patent for a new extrusion process of filament production from recycled PET using a simple screw. This equipment consisted of two parts: the first one is used to recycle PET with a stirrer and a condenser and the second one is related to the extrusion process. This development was proposed to obtain a constant quality in the extruded product, which is the main problem of the traditional PET extrusion recycling.

On the other hand, the main disadvantage of traditional designs of commercial extruders relies on it may take several minutes to access the screw. In addition, these extruders are manufactured by foreign companies other than Mexico. Therefore, it is necessary to have Mexican design extruders with better operability characteristics. A previous study focused on the design and operation of a modified extrusion equipment with spinning fiber devices (García, 2008). This extruder is based on a normal design but it has an adjustable support for the motor that allows the screw to be removed in just one minute.

In the present study, an analysis of the critical variables that impact the flow uniformity of the melt polymer and the degradation of recycled PET using this modified extruder has been performed. The content of this manuscript comprises: a) A brief description of the design of this modified PET extrusion and spinning process for textile applications, b) The mathematical analysis of the operation curves obtained in the extrusion process, c) Experimental results and the material characterization in the

extrusion and spinning stages and d) The validation of the extruder performance using a comparison between the experimental data and theoretical operation curves.

2 Methodology

2.1 Process design

The process design was performed considering the following specifications of the process and the final product for a textile fiber: a) operation temperature: 260 - 300 °C, b) drying temperature: 70 - 140 °C, c) maximum PET humidity: 0.02%, d) product denier: 1.5 to 3, e) fiber tenacity: 2 g_f/d, f) elongation: 20%, and g) residual retraction: < 5%. For the screw design, the characteristics of the raw material and the design parameters such as the compression ratio, residence time, angles of the helix and the relationship of length to diameter of the extruder were defined according to results reported in the literature (Martin and Rojas, 2005; Dale, 2000).

2.2 Mathematical analysis

To perform the analysis of the case of study, mathematical model reported by Dale (2000), which is based on PET degradation during reprocessing, was adapted for representing the extruder and raw materials used in the present communication.

The equation for the calculation of the total flow (Q) inside the extruder was obtained by solving the moment, heat and mass balances and considering the problem as a fluid movement between two surfaces, where one surface (i.e., screw) is movable. Rectangular coordinates in z direction (channel of the screw) were used and this model is given by

$$Q = \frac{\pi^2 D^2 N H \sin \theta \cos \theta}{2} - \frac{\pi D H^3 \sin^2 \theta \Delta P}{12 \eta} \frac{1}{L} \quad (1)$$

where N is the screw rotation speed, ΔP is the pressure drop, η is the melt viscosity of the material, D , H , L and θ are the diameter, depth, length and angle of the screw, respectively. Note that the melt viscosity η is a function of the inherent viscosity (IV) and temperature (T)

$$\eta = 0.098 e^{\left(\frac{6800}{T}\right)} (IV)^{5.1} \quad (2)$$

During the process, the degradation of the PET material may occur by chain scission reducing the average molecular weight, which can be measured via

the inherent viscosity at any residence time in the melt (IV_t). This viscosity is given by the following equation

$$IV_t = \left[\left[\left(\frac{1}{IV_0} \right)^{1.47} + \left[e^{\left[\frac{26.9 \cdot 17080}{T} \right]} \right] t \right]^{1.47} \right]^{-1} \quad (3)$$

where T is the absolute temperature, IV_0 is the initial inherent viscosity and t is the residence time in the melt, respectively.

On the other hand, if the PET polymer contains some water, the hydrolytic degradation can also occur during the process and it can be calculated using

$$IV_H = \frac{IV_0}{\left[1 + 21.9 (x) (IV_0)^{1.47} \right]^{0.68}} \quad (4)$$

where IV_H is the inherent viscosity after the reaction with water, IV_0 is the initial inherent viscosity of supplied PET and x is the weight of water (%) in supplied PET, respectively.

The analysis was focused on the degradation and the material uniformity in the process and this analysis was based on the strategy and method reported by Franceschini and Macchietto (2007). Therefore, a complete factorial experimental design and mathematical calculations of the operation curves were performed. The process analysis considers the variation levels for PET recycled in the following critical ranges: mass flow variation of $\pm 10\%$ (Dale, 2000; Incarnato, 2000; Torres and Robin, 2000; Martín and Rojas, 2005), pressure drop of $\pm 10\%$, intrinsic viscosity from 0.8 to 0.5, and raw material humidity less than 0.02% (Seo and Cloyd, 1999; Incarnato *et al.*, 2000; Abbasi *et al.*, 2007).

3 Experimental fiber extrusion

The extruder was maintained at constant temperature during 10 minutes before the initial feeding. The raw material was fed using a batch charge of 100 g, the operation conditions (temperature, pressure and mass flow) were continuously monitored and were kept constant during a time corresponding to three times the residence time of the material inside the extruder for to reach steady state before the evaluation of the test. The pressure was fixed for every test using different screw speed. The mass flow was determined at different conditions of pressure (from 2.68 to 4.14 Kg/cm²) and temperature (260, 280, 290 and 300 °C) for two different raw materials: bottle grade ($IV = 0.65$) and pharma grade recycled PET ($IV = 0.71$). The

results obtained in the mathematical modeling were compared with the experimental results.

4 Results and discussion

4.1 Extruder and spinning design

Figures 1 and 2 show schematic diagrams of the equipment used for obtaining recycled PET fibers including the extrusion and spinning stage. The main components of this equipment are: a) For the extrusion stage: a blending system (1 and 2), feeding hopper (3), barrel and screw of the extruder (4), gear box (5), direct current motor (6), adjustable motor support (7), extruder support structure (8), breaking plate (9), filtration system (10), gear pump (11), spin pack (12), cylindrical cooler (13), air distribution pipe (14), take up roll (15) and control panel (24); b) For the spinning stage: two groups of 5 rollers (16, 18), finishing container (17), drying and stretching system (19, 20), second stretching system (21), collecting filament reels (22) and package rotating changer (23).

The extruder is based on a normal design but it has an adjustable support for the motor that allows the screw to be removed in just one minute in order to analyze how the material changes inside the barrel at different process conditions (Rajabinejad, 2009). To remove the screw, the gear box has two disassembling gears that permit to separate them with a single bolt. Later, the motor is moved on the adjustable support for taking out the screw in an opposite direction to material flow. In the spinning stage, a slow first and second stretching are applied to the polyester fiber by using three groups of five rollers. This operation improves the molecular orientation and the crystallinity in order to increase the mechanical properties of the polyester fiber (Samperi, 2004; Abbasi, 2007).

Table 1. Operation conditions for processing virgin and recycled PET

Condition	Value
Residence time	10 minutes
Extrusion temperature	280 °C
First stretching	1.5 times
Second stretching	1.5 times
Drying temperature	120 °C
Spinning Speed	50 m/minute

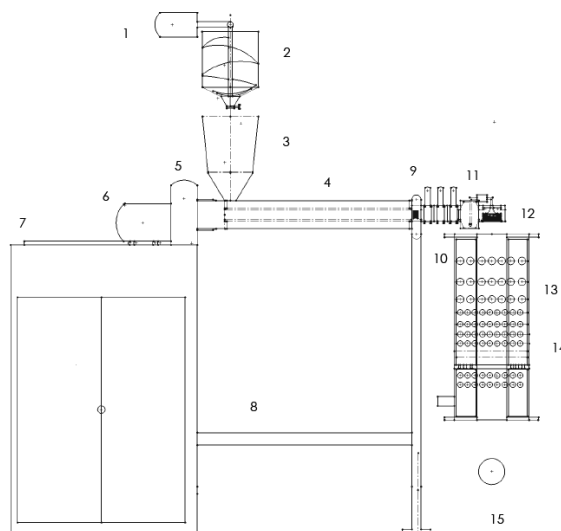


Fig. 1. Lateral view of the extrusion stage used for the production of recycled PET fibers.

Two drying systems were defined to improve crystallinity, dimensional uniformity and to reduce the fiber porosity (Abbasi, 2007), using heat rolls at temperatures between 10 and 100 °C over the glass transition temperature of PET. Using the same conditions in the stages of extrusion and spinning, virgin and recycled PET were processed to the conditions reported in Table 1.

4.2 Mathematical analysis of the process

Calculation of the flow rate Q was performed using Equation 1 with the geometry and the extruder operation conditions given in Table 2. For these calculations, we have considered a standard PET extrusion process with initial intrinsic viscosity of 0.6, a feed mass flow of 99 g/min and L/D extruder ratio of 30. The extruder diameter was 33 mm. Figure 3 shows the behavior of the flow rate along the extruder length. The changes of density or flow rate were produced by the temperature and the geometry of the extruder. These variations were more significant in the first two sections of the extruder, by the geometry and temperature differences between these sections. Pressure along the screw is affected mainly by its geometry in the last extrusion zone. For an extrusion process of recycled PET, this increasing pressure was mainly obtained by the total fusion of the material and the geometry in the last 34 cm of the extruder. If an increment of 10% for the mass flow variation in the feed is considered (Figure 4), the

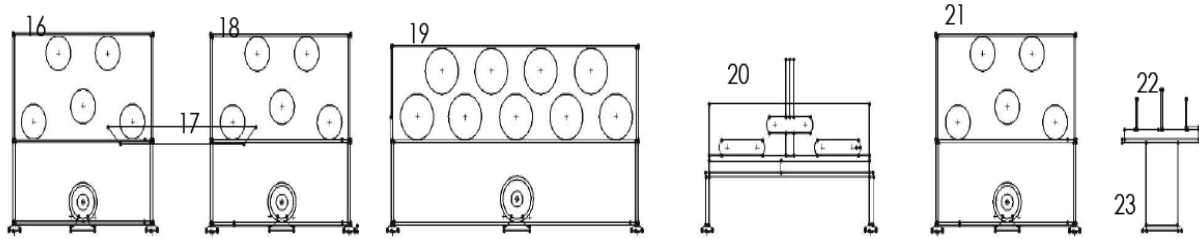


Figure 2. Lateral view of the spinning stage used for the production of recycled PET fibers.

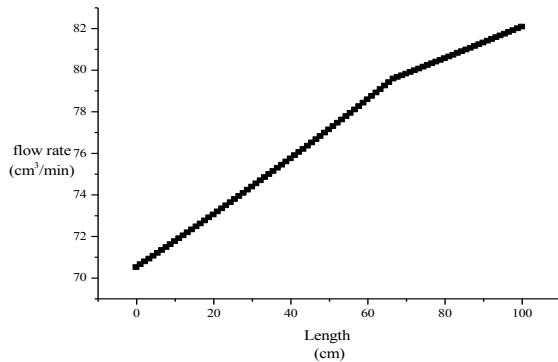


Fig. 3. Flow behavior inside the extruder obtained using Equation (1).

Table 2. Geometry and operation conditions of the extruder

Extruder zone	Feed	Compression	Dosing
Diameter (cm)	3.33	3.33	3.33
Length (cm)	33	33	34
Depth (cm)	0.6	0.38	0.16
	(average)		
Angle (°)	17.2	17.7	15
Operation temperature (°C)	280	285	290

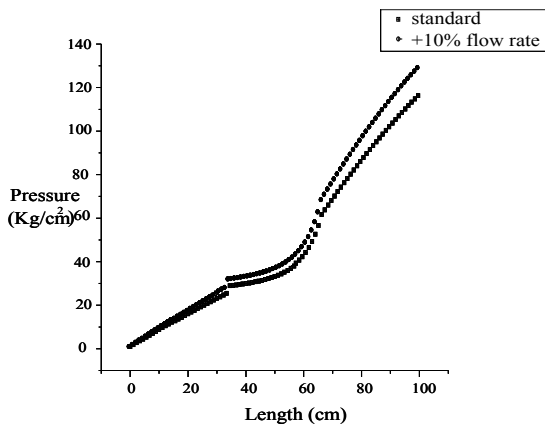


Fig. 4. Pressure inside the extruder of a standard PET extrusion process and curve with a flow rate variation of +10%.

pressure does not present considerable changes inside the extruder for the first two sections but, for the last zone (compression), the pressure presents a significant variation. This result is an important issue to be considered in the system control because this level of mass variation is a normal value in the recycling

process and it will impact the final diameter of the filaments produced using this type of extruders. The normal variations of the filament diameter are +/-10% in a commercial fiber production process. Note that this is the reason for minimizing this mass variation using the pressure control of the screw.

Mathematical analysis of the operation curves indicates that the variability of IV_0 , in the range of 0.5 at 0.8, must be reduced by choosing the appropriate raw materials to produce a quality product. On the other hand, it is necessary to handle the following variables at these ranges for to minimize the variability: screw speed of 30 to 45 rpm, temperature of 270 to 300 °C and humidity of 0.01 to 0.02 % by weight.

Figure 5 shows the variation in the pressure drop (dP) as a function of the humidity content (X , weight %) and inherent viscosity (IV). Figure 6 shows the degradation behavior of the material, which has been analyzed using the calculation of % of IV loss, using Equation 4, for three initial inherent viscosity ($IV_0 = 0.8, 0.6$ and 0.5). It is clear that IV variation (i.e., degradation) is more significant when polymer with high average molecular weight (high IV_0) is extruded.

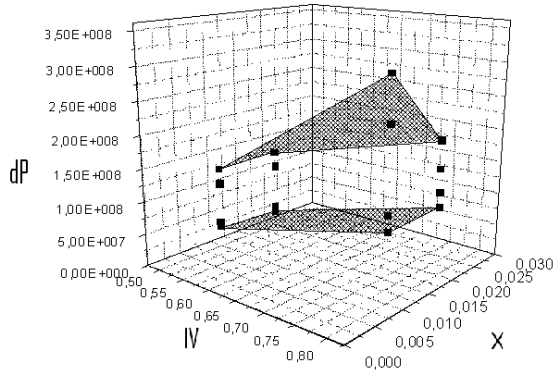


Fig. 5. Drop pressure as a function of inherent viscosity (IV) and humidity (X).

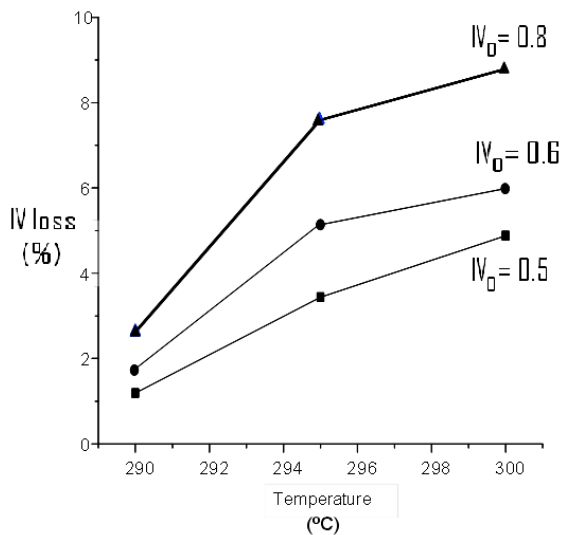


Fig. 6. IV loss (%) as a function of temperature for three initial inherent viscosities (IV_0).

4.3 Experimental results in the spinning stage

Figure 7 shows the relationship of the mass flow of pharma-grade recycled PET with respect to the pressure applied to the extruder for four different temperatures. Results indicated that, the variation of the mass flow is not so sensitive to the variations in the extruder pressure at 290 °C. On the other hand, Figure 8 shows the same relationship for the bottle-grade recycled PET for different operating temperatures. At 280 °C, the mass flow presented a minimum variation among the three studied temperatures regarding the analyzed range of pressures.

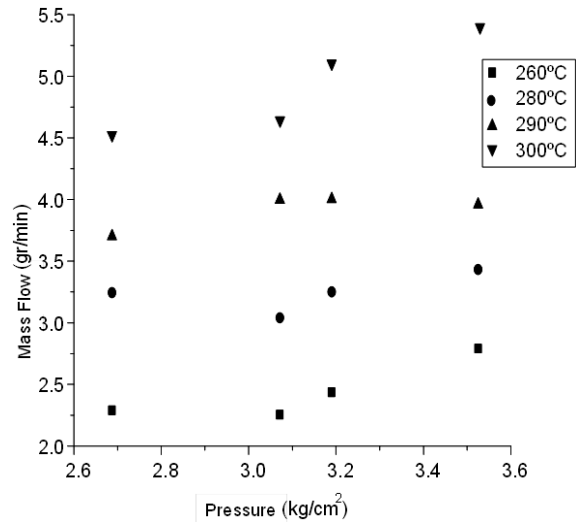


Fig. 7. Mass flow as a function of pressure at four different temperatures for pharma-grade recycled PET.

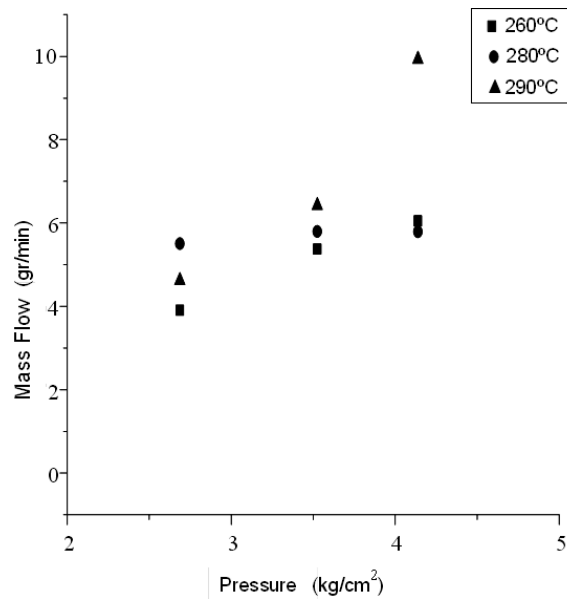


Fig. 8. Mass flow as a function of pressure at three different temperatures for bottle-grade recycled PET.

Finally, Fig. 9 shows the experimental mass flow of the pharma-grade recycled PET and the corresponding theoretical mass flows. The predicted results are very close to experimental data with a maximum error of 3%. In fact, very similar results are obtained for bottle-grade recycled PET. This result indicates that the model used to describe the performance of the experimental set up considering rectangular coordinates, is suitable for the modified

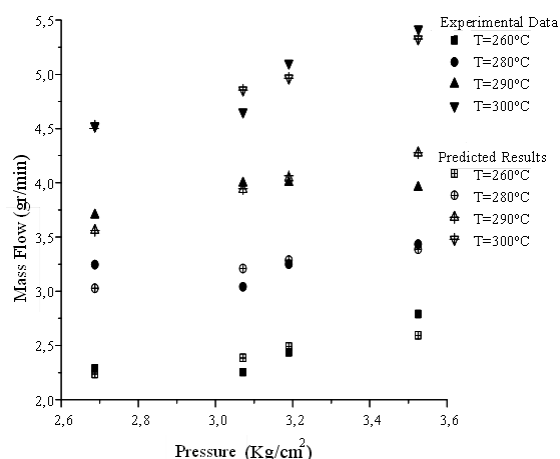


Fig. 9. Comparison of the experimental and calculated values of mass flow in the extruder.

extruder-spinning process developed in the present communication because the material inside the extruder flows similar to a laminar displacement. It is worth to mention that the quality and properties of the recycled PET fibers obtained using this modified spinning process are reported elsewhere (Tapia-Picazo et al., 2013).

Conclusions

The screw design should contemplate an appropriate handling of flows and pressure drop inside the extruder using adequate raw materials and adequate range of operation conditions in the critical variables and process parameters for recycling PET. Results of the mathematical analysis were used to analyze the impact of the characteristics of the raw material, pressure, temperature, feeding flow, extrusion speed and extruder design on the extrusion process of recycled PET. Process conditions have been determined in the extrusion system, which agree with the results obtained in the mathematical analysis of the operation curves and those reported in the literature. The mathematical prediction of the extruded mass flow agrees with the experimental data with a maximum error of 3%. Finally, the recommended conditions and the principal characteristics of raw recycled material for textile applications are: IV_0 in the range of 0.5 at 0.8, screw speed of 30 to 45 rpm, temperature of 270 to 300°C and humidity of 0.01 to 0.02% by weight.

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