Several properties of filament fibers made from recycled bottles of mineral water using melt spinning method

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Abstract. Waste mineral water bottles made of PET called post-consumer POSTC-PET packaging with recycling code no.1 can be made into another material other than the bottle by using a mechanical recycling process. In this experiment carried waste recycling process bottled mineral water bottles of PET into filament fibres with the aid of a melt spinning. From the resulting experimental filament fibres diameter of 14-15 microns, obtained the draw ratio is 1/46, 573,5 – 699,8 MPa tensile strength, modulus of elasticity of 2,01 – 2,45GPa, moisture regain of 2,84. Keywords. PET; Bottle; Fiber; Melt; Spinning; Drawing.

1. Introduction

The use of Polyethylene terephthalate also known as PET or PETE (a plastic resin materials used for making packaging materials such as bottles and food containers) is increasingly becoming paramount among manufacturers, as they used these PET bottles to package their products because it (PET) is an excellent barrier material with high strength, thermo stability and transparency [1]. Consumer also prefers or chooses PET because it is inexpensive, lightweight, resealable, shatter resistant and recyclable [2]. More than 75% of plastic water bottles are never recycled; they’re simply thrown away and generate more than 121 million tons of waste each year [3]. Unfortunately, the postconsumer drink bottles made of polyethylene terephthalate are approaching 5 million tons/year worldwide and cannot be reused. This is banned not only by hygienic requirements, regulated by consumer protecting acts, but also because of substantial degradation in molecular of PET, which is followed by a decrease in melt viscosity and mechanical strength [4,5].

Two major processes have been applied in order to recycle post consumer PET (POSTC-PET) flakes. These processes are chemical recycling and mechanical recycling [6]. Chemical recycling (chemolysis) of POSTC-PET is achieved by total depolymerisation into monomers or partial depolymerisation into oligomers. The chemicals used for the depolymerisation of PET include water (hydrolysis), methanol (methanolysis) and EG (glycolysis) [7,8,11]. The main disadvantage of PET chemolysis is its high cost [9]. The mechanical recycling of POSTC-PET normally consist of contamination removal by sorting and washing, drying and melt processing [6].

The removal contaminants from POSTC-PET is a vital step in the mechanical recycling process od PET. Contaminant removal consists of several processes in which POSTC-PET bottles are sorted,
ground and washed. The sorting process is basically separating PET bottles from PVC, polyethylene and other plastic containers. The sorting of PET bottles is an important and critical step. High level of contamination by other materials causes great deterioration of POSTC-PET during processing. PVC and other plastic container can be manually removed from PET bottles. The fact that manual separation PVC and other plastic container are becoming more expensive indicates that micronyl process would be more feasible to use in the industry. After sorting, PET is ground into flakes in order to be easily reprocessed. PET flakes are washed following grinding. Hot wash using NaOH and detergent at 80 °C followed by a cold wash with water only. Drying regarded as an essential step in POSTC-PET recycling. Minimizing the moisture content of POSTC-PET reduces the hydrolytic degradation effect and leads to higher recycled PET melt strength. Most manufacturers use drying condition ranging from 140 – 170 °C; 3 – 7 h [10]. In typical operating conditions, no more than 50 ppm water is allowed to be present in the PET flakes and this is normally achieved by using desiccated dryers operating before feeding to the extruder [9]. Melt processing POSTC-PET can be processed in a normal extrusion system into useful granules. However, due to the contaminants mentioned above, extrusion of PET at 280 °C with the presence of contaminants reduces in molecular weight (MW) due to degradation reactions [9].

The main advantage of mechanical recycling of POSTC-PET is the fact that the process is relatively simple, environmentally friendly and requires low investment. The main disadvantage of mechanical recycling is the reduction of MW during processing, otherwise known as the reduction of viscosity. A great sense of awareness of this problem by researchers and manufacturers was generated in the past two decades to find ways to maintain viscosity during POSTC-PET processing [6]. Crystalinity of recycled materials (PET) was higher than those obtained for virgin materials [14]. Consequently, the tenacity of samples from used material was higher and the elongation was smaller. Herein, it is convenient to highlight that there are other scientific and industrial reports about the production of fibers from recycled PET [15-18]. Unfortunately, the technical details are not described because of the industrial secrecy [19].

For commercial PET flakes, DSC shows that Tg inflexion is at 83,9 °C and there is a weak endotherm due to a phase transition at 176 °C and Tm at 246 °C. After it has been quenched from melt, note that there is a lower Tg transition at 78,2 °C, a strong crystallization exothermal peaking at 167 °C and Tm at 245,8 °C. Note that the enthalpy of melting has fallen from 40,57 J/g to 36,34 J/g as a result of the lower order of crystalinity [20]. Usually, melt-polymerized PET has an IV of about 0,67. It is quite possible by using Solid State Polymerization process to raise IV to 0,80 – 1,10. High IV polymer is needed for high tenacity PET fibers for sling, tyre cords, and very much so for PET bottle resin. PET bottle grade resin polymer has IV 0,92. Recycled PET bottles are ground up, dried and remelt-spun into polyester yarn for fiber fill of for carpet yarns. One manufacturer [21] says that the so-called ‘food grade’ PET, made from recycled bottles, gives a more hard-wearing polyester carpet yarn. The higher yarn MW, which increases fiber toughness and work-to-break parameter, is the reason [20].

For commercial non-olefin polymers, including PET, PBT, Polycarbonates, Nylon, there are additives which giving hydrolytic stability in applications such as food contact packaging and medical devices. Its afforded by Doverphos S9228, which is claimed to offer better colour and stability than 2,4-di-t-butylphenyl phosphite, and also to function as an effective secondary anti-oxidant in combination with hindered phenols. Based on its pentaerythritol diphosphite technology, it can be used as stabilizer [22].

Typically PET textile fiber properties have diameter above 12 micron, with moisture regain value at 20 °C and rH 65 % is about 0,4 %, specific gravity (SG) is about 1,34, modulus of elasticity (E) is about 1000 g/tex or equal to 13 GPa, tensile strength is about 25 – 54 g/tex or equal to 325 – 656 MPa, elongation at break (EL) is about 12 – 55 % and melting point (Tm) is about 250 °C [23,24]. High tenacity PET fiber tensile strength reached 1,139 GPa at elongation at break about 7 %. As
hydrophobic material, PET has a moisture regain of 0.4 % at rH 60 %. Therefore, PET fiber was difficult to wet and rapidly build up static electrical charges by friction because as water effectively leaks away, voltage is produced [26]. The Fibers and Filaments magazine reports the commercial use of recycled PET in textile products such as fiber fillings, non wovens, apparel, carpet, home textiles and Bulk Continuous Filaments (BCF) [27].

The purpose of the present study was to mechanically recycling waste POSTC-PET bottle to become PET filament using melt spin apparatus and investigate the draw ratio based on diameter change characterized by optical microscopy to tensile and moisture properties of the fibre.

2. Melt spin apparatus
Melt spinning is the simplest method of fibre manufacture, mainly because it does not involve problems associated with the solvents. It is therefore the preferred method, provided polymer gives stable melt. A major development era is in 1970, which was the transition from conventional spinning 1000 m/min to high speed spinning 3000 m/min and higher. In the direct spinning process, the homogenous and spinable melt produced by polymerization may be directly passed to the spinning machine at the gear pump stage. When the winding speeds are high, the yarn may be directly dropped to the wind-up device with no godets being used. The later version of melt spin machine allows the yarn tension on take-up to be controlled using S-shaped wraps around cold godets.

Melt spin apparatus is a kind of melt spin filament maker which the polymer melts and compressed by the dead weight cylinder made from steel. The basic principle of this melt spin apparatus is that the weight of the stick creates compression to the viscous liquid of the polymer. The compression strength allowed the polymer to flew down passed the spinneret that have single hole and small diameter. This type of apparatus is built at Laboratorties Logam in Fakultas Teknik Mesin & Dirgantara ITB Bandung. Design of the apparatus is shown in figure 1. The apparatus contains 5 main parts, temperature control unit; heating unit; tube; stick; drawing unit. The total dimension is 2 m height and 50 cm wide. Stick diameter is 15 mm and have 1 kg weight, spinneret diameter is 0.5 mm. Ring type heater used to heat the cylinder for polymer melts. The drawing unit has 5 differential speeds to create different draw ratio.

Figure 1. Design of melt spinning apparatus
The distance between spinneret and drawing unit is about 0.8 – 1.2 m long. The heating control unit controlled the rise of temperature about 6.75 °C/min and getting lower 10 °C below its set. There is also an asbestos belt surrounding the cylinder to reduce heat loss.

3. Experimental
600 ml Waste PET bottle with trade mark AQUA Danone as POSTC-PET raw material were prepared and separated from the brand label and the cap. Mechanical recycling used to make PET bottle flakes. Washing with neutral soap in 60 °C 15 min, rinsing with warm and cold water and drying in 100 °C 60 min is done before cutting the bottle manually with scissor into 25 mm2 flakes. A check on the composition by FTIR and TGA gave about 83 % PET. Thermal properties of PET bottle flakes by DSC and TGA gave Tg 85 °C, Tm 320 °C and Tdeg 350 °C. Therefore the thermal processing is done near Tm.

Prior to melt spinning, the flakes were dried by heating in 100 °C for 60 min and stored in dry carrier contains silica dessicants. Flakes inserted manually after the temperature display on the apparatus show 75 °C. The temperature set to 300 °C to reached 285 - 290 °C. The stick inserted manually after the temperature reached 250 °C, and compressed manually after reached 285 °C. The flakes then flew down from the spinneret after compression.

The filament created by drawing and winding the melted flakes using first speed (lowest) of drawing unit. With the spinneret to drawing distance about 1 m, its also created solidification of the filament. The drawing and also winding are done for approximately 3 min to create filament diameter stability. Drawing and winding material is made from 3 mm thickness silicone tube. After winding is done, the filament is removed from the silicone tube to characterized.

The draw ratio (DR) were determined from the following equation :

$$DR = \frac{d_0}{d_1}$$

(1)

Where d0 is the diameter of filament created without drawing action, and d1 is the diameter of filament created with drawing at first speed in drawing unit.

4. Characterization
Optical characterization to determined the diameter of drawed and undrawed filament compared to human hair were done at Laboratorium Logam FTMD ITB. Mechanical characterization to determined strength, elastic modulus and elongation at break, and moisture properties to determined moisture regain were done at Laboratorium Evaluasi Kimia Tekstil STTT. Filaments birefringence was measured with Wild Heerbrugg stereo microscope and captured using dinocapture 2.0 software.

Filaments mechanical properties including tensile strength, elongation and modulus elasticity was measured with MesdanLab Strength tester in accordance with ASTM D 3822-01 using load cell 1000 N, polyurethane clamp, clamp speed 50 mm/min and clamp distance 25 mm. Tensile test environment condition is 22 °C; rH 65 %. Strength and modulus of elasticity calculated manually using microsoft excel.

Filaments moisture properties including moisture regain (MR) and moisture content (MC) prepared using SNI standard 8100:2015. The filaments and the glas stopper flask were dried using Memmert atmosafe universal oven. Both were weighed using Precisa XB220A analytical balance. The equation related between MR, MC and weight differences are shown below.

$$MR = \frac{(\text{wet weight} - \text{dry weight})}{\text{dry weight}}$$

(2)

$$MC = \frac{(\text{wet weight} - \text{dry weight})}{\text{wet weight}}$$

(3)
5. Results & Discussion

From the observation using stereo microscope and dinocapture software, from figure 2 it was observed that the first drawing diameter of filament is about 14.15 micron to 15.63 micron and compared with human hair (73 micron) is 4.8 times thinner. But if compared with diameter from typical PET fibers (12 micron), its a little thicker. It well understood because the melt spinning apparatus is using single drawing that merge two process (drawing and winding) in single unit, unlike industrial PET fiber which have several stages to draw the fiber into accepted diameter that can make the fiber become thinner and also using winder as winding unit separated from drawing unit.

Figure 3 results the other microscopy observation for non drawing melt spinning filament diameter is about 675.63 micron to 694.00 micron. From above data and equation, we can calculate the draw ratio is 1/46. This draw ratio is depended on differential speed between stick and drawing unit.

Figure 2. Stereo microscopy image of filament cross section with drawing at first speed (compared with human hair).

DL0 (72.96 micron) & DL3 (73.06 micron) are measurement of human hair, DL1 (14.15 micron) & DL2 (15.63 micron) are measurement of the drawed PET filament.
From Mesdanlab Tester, the average maximum load to break the filament is 11 cN. With average elongation at break is about 28.5%. Then after calculating (using diameter 14.15 – 15.63 micron), the tensile strength of filament is about 573.5 – 699.8 MPa, modulus of elasticity is 2.01 - 2.45 GPa. The result of fiber tensile strength is on top limit to typical PET fiber which has range 325 – 656 MPa. Then the fiber modulus of elasticity is lower than typical PET fiber due to the elongation at break about 28.5% if the assumption is fiber is fully elastic or no plastic deformation. The actual modulus of elasticity may go higher due to PET behaviour is thermoplastic.

The moisture regain calculated by the difference between wet weight and dry weight, divided by dry weight. The weight of flask is 143.5907 g, the weight of flask included PET fiber before drying is 143.9525 g, the weight of flask included PET fiber after drying is 143.9425 g. The moisture regain resulted is 2.84%, a significantly higher than typical PET fiber (0.4%). The higher results may depended on the lower contents of PET which only 83% as shown on TGA, the others content such as additives may have a hydrophilic nature that may increase the moisture regain.

Furthermore, the higher result of moisture regain will affects to less statical electrical charges by friction and also increase wet ability related to hydrophilicity, both are decreases voltage shocks.

6. Conclusion
Single filament fiber made from waste POSTC-PET bottle were succesfully prepared and characterized. Optical microscopy result that the diamater is a little bit thicker from typical PET fiber. The tensile strength is on top limit to typical PET fiber, but the modulus of elasticity is lower due to high elongation at break (with assuming that the fiber is fully elastic). The moisture regain is higher that typical PET fiber which has advantages to reduce poor properties related to hydrophobicity. Therefore, its suitable to be PET fiber related to textile use.
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