The Effect of Recycling Frequency on the Melt Flow Rate of Polypropylene Materials

Neilinda Novita Aisa[1,a)], Rohmat Setiawan[1,b)], Danny Wicaksono[1,c)], Eduardus Dimas[2,d)]

Author Affiliations
*[1]*Department of Machinery and Industry, Astra Polytechnic, Cikarang, Indonesia*
*[2]*Department of Certification and Competence, Astra Polytechnic, Cikarang, Indonesia*

Author Emails
a)neilinda.novita@polman.astra.ac.id
b)rohmat.setiawan@polman.astra.ac.id
c)danny.wicaksono@polman.astra.ac.id
d)eduardus.dimas@polman.astra.ac.id

Abstract. Widely used of plastic material regularly produce ecological and environmental issues due to increased number of waste. Recycle process is an alternative to overcome the plastic waste issues. Effect of using recycled plastic material is molecular structure changes that will affect to its physical and mechanical properties. The melt flow rate's (MFR) materials properties are closely related to the production of plastic products, especially to determine the type of forming process and temperature setting of each material. This study aims to determine the effect of the frequency of recycling on the flow rate properties of Trilene HI 10HO Polypropylene (PP) plastic materials using experimental methods. Based on analysis, the MFR value of recycled polypropylene material has increased as the frequency of recycling is increased. The MFR value did not change much until the 2nd recycling process, but it's changed drastically at the 3rd and 4th recycling process and the MFR value increased 4 times compared to the virgin material. And on the 6th recycling process, the MFR value increased up to 7 times. In addition to changes in the MFR value, recycled materials also have a darker color shift.

Keywords: Polypropylene, Recycled Material, Melt Flow Rate

I. INTRODUCTION

Polymers are one kind of materials that are commonly used in wide range of purpose such as food and beverages packaging, medicine, automotive and others [1] due to its ability to be easily manufactured especially in large quantities. The enormous amount of polymer has come into ecological and environmental issue as a result of its inability to decompose naturally [2]. The recycling process is the best way to handle waste polymer products in comparison to the traditional techniques (combustion of waste polymers or burial underground), which have detrimental impacts on the environment owing to the development of dust, fumes, and harmful gases [3]. Polypropylene is a linear – semi crystalline hydrocarbon polymer belonging to the thermoplastic polymer family with a high degree of crystallinity because it has a 95–98% isotactic phase and a 2–5% atactic [4].

MFR is an indirect measure of molecular weight and can be a simple indicator of how the addition of recycled materials affects the final process ability of the original resin [5]. In the plastics industry, one of the most useful data in determining the type and process conditions related to temperature regulation is the value of the melt flow rate (MFR) and melt volume rate (MVR) of the material [6] to control the volume and flow rate of the material to ensure product quality [7]. A high melt index indicates a low molecular weight [8]. Melt flow rate (MFR) or Melt Flow Index (MFI) is a convenient measure of the thermoplastic material passing through the die at a predetermined temperature and load. The melt flow rate of polypropylene was measured at 230 °C at 2.16 kg [9].

Almost all thermoplastics can be injection molded. These settings should then be adjusted for each specific mold to achieve the minimum molding cycle for optimum part performance and quality. The grades of resin for injection molding and extrusion are usually quite different. Whereas in extrusion a resin with high melt strength is desired so that the part will retain its shape while cooling, the opposite is true with injection molding. Resins with low melt viscosity are desired so that the flow through the runner system and gate and into the mold is done easily with minimum injection pressure. Injection molding grades would generally have low molecular weights and narrow molecular weight distributions [8].

TABLE 1. Properties of polypropylene trilene HI 10HO

Product Properties	Test Method*	Unit	Value
Melt Flow Rate (230^0C / 2.16 kg)	ASTM D 1238	g/10 min	10
Density	ASTM D 792	g/cm^3	0.9
Tensile Yield Strength (@50 mm/min)**	ASTM D 638	MPa	35
Tensile Yield Elongation **	ASTM D 638	%	13
Flexural Modulus (@1.3 mm/min)**	ASTM D 790A	MPa	1.500
Notched Izod Impact Strength (@23^0C)	ASTM D 256	J/m	30
Hardness, Rockwell	ASTM D 785	R-scale	100
Deflection Temperature (@0.455 MPa)	ASTM D 648	^{0}C	104
Vicat Softening Temperature	ASTM D 1525B	^{0}C	152
Melting Temperature	ASTM D3418	^{0}C	163

TABLE 2. Guidelines for Experimental Parameters

MFR (g/10 min)	Mass of test sample in cylinder (g)	Extrude cut-off-time Interval (s)
≥ 0.1 to ≤ 0.5	3 – 5	240
> 0.5 to ≤ 1	4 – 6	120
>1 to ≤ 3.5	4 – 6	60
>3.5 to ≤ 10	4 - 8	30
>10	4 - 8	5 to 15

Figure 1. As received granular polypropylene

II. MATERIALS AND METHOD

Materials

Polypropylene (PP) is known as a linier hydrocarbon polymer [4] which commonly used in injection molding, extrusion, blow molding, and thermoforming. In behalf of the production process versatility, PP is one of the highly recommended materials to be used in various applications, such as plastic containers, machinery and equipment parts, and fibers and textiles products. The melting point of polypropylene occurs in a range depending on the atactic material and its crystallinity [10]. Polypropylene (PP) Trilene HI 10HO in granular form was used in this research, the properties of the following materials can be seen in Table 1. Granular polypropylene can be seen in Fig 1.

Melt Flow Rate

The test standards for measuring the MFR value of plastic material are ASTM D1238 and ISO 1133. MFR can also be used to determine the reduction in plastic quality due to the molding process [11]. MFR has a measurement result of g/10 min. The schematic of MFR test is shown in Fig 2.

Equations (1) are guidelines for experimental parameters based on the material flow range according to ISO 1133 for the recommendation for sample weight in the cylinder and time interval. Value of MFR can be calculated using equation (1) [12]:

$$\mathbf{MFR}(\boldsymbol{\emptyset}, \boldsymbol{mnom}) \rightarrow \boldsymbol{tref\ x\ \frac{m/n}{t}} \qquad (1)$$

Where Ø is the temperature test in degree Celsius (^{o}C), *mnom* is mass of the piston in (kg), *m/n* is the fraction mass of sample material in grams (g) and the quantity of total sample, *t* is the cut of time in ten seconds. MFR testing was conducted according to the ISO 1133 standard. The experimental parameters are shown in Table 3.

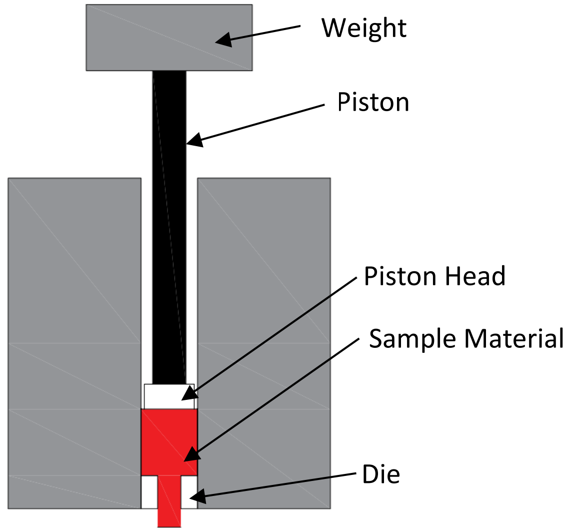

FIGURE 2. Basic Construction of Melt Flow Tester.

TABLE 3. Melt Flow Parameter of PP

Parameter	**Nominal**
Temperature	230^0C
Standard load	2.16 kg
Extrude Cutting-off time interval 1st recyle	30 second
Extrude Cutting-off time interval 2nd – 6th recycle	10 second
Total Sample/test	3 times

TABLE 4. Result of Melt Flow Tester

	Recycle 1		**Recycle 2**		**Recycle 3**	
	Sample Weight (gr)	**MFR (gr/10 min)**	**Sample Weight (gr)**	**MFR (gr/10 min)**	**Sample Weight (gr)**	**MFR (gr/10 min)**
Sample 1	1.50	10.00	0.74	14.80	2.07	41.40
Sample 2	1.95	13.00	0.65	13.00	2.10	42.00
Sample 3	1.80	12.00	0.70	14.00	2.04	40.80
Average	1.75	**11.67**	0.70	**13.93**	2.07	**41.40**
	Recycle 4		**Recycle 5**		**Recycle 6**	
	Sample Weight (gr)	**MFR (gr/10 min)**	**Sample Weight (gr)**	**MFR (gr/10 min)**	**Sample Weight (gr)**	**MFR (gr/10 min)**
Sample 1	2.19	43.80	3.39	67.80	3.95	79.00
Sample 2	1.95	39.00	3.30	66.00	3.81	76.20
Sample 3	1.89	37.80	3.09	61.80	3.55	71.00
Average	2.01	**40.20**	3.26	**65.20**	3.77	**75.40**

The parameter value as listed at Table 3 are obtained based on British Standard ISO 1133 [12]. After the recycling process was carried out once, it was found that the MFR value of the material changed greater than 10 g/10 minute. It is advised that the extrude cutting-off time interval for thermoplastic materials with an MFR value of >10 gr/10 min, be 5–15 seconds as shown in Table 2. In this experiment, the extrude cutting-off time interval of 10 seconds is used for the second recycling and subsequent cycles.

III. RESULT and ANALYSIS

Result

Table 4 shown the result of MFR Polypropylene material from the 1st recycling to the 6th recycling. Each type of recycling sample was tested three times. In the first recycled material, 1 sample was taken 3 times at 30 second extrude cutting-off time intervals. After it was known that the MFR value of the first recycled material was >10 gr/10 minute, then for the next test parameter setting, the extrude cutting-off time value was changed to 10 seconds, based on table 2.

The diagram at Fig 3 showed an increase value of MFR along with the increasing frequency of recycle. It's known that the properties of polymer materials will change with changes in temperature due to the movement of molecules [13]. As a result, along with the increasing frequency of material recycling, the color of the material also becomes darker, as shown in the Fig 5. Changes of color in the specimen occurred due to degradation of the material that subjected to high temperature over long period of time [8].

Analysis

Graph in Fig 3 depicts the change in MFR value in recycled materials. Materials that are 1st and 2nd recycled experience a change in the MFR value that is not so large compared to the MFR value of the pure material. The increase in the MFR value continues to occur along with the increasing frequency of recycling. In the 3rd and 4th recycling, the MFR value produced is much higher than before, or 4 times that of the pure material. In the 5th and 6th recycling, the MFR value increased again by almost 7 times compared to the pure material. From the data, it can be seen that polypropylene material maintains its MFR value to remain in the range or value that is almost the same after only one recycling. The higher the MFR value, the lower the viscosity of the material, or the material becomes very runny. The main factor that causes the decrease in material viscosity after recycling is temperature. Materials that are reheated lose specific gravity due to vaporized density [14] and specific gravity is one of the important factors that affect a material's flow properties.

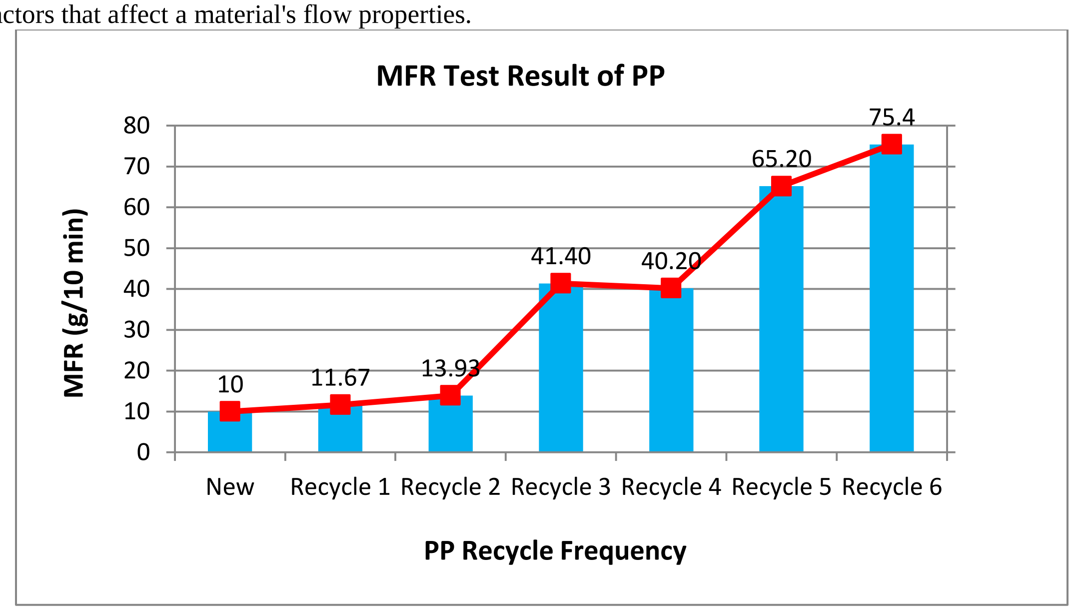

FIGURE 3. Result of melt flow rate at as received state and recycled state.

FIGURE 4. Melt flow rate result of as received specimen

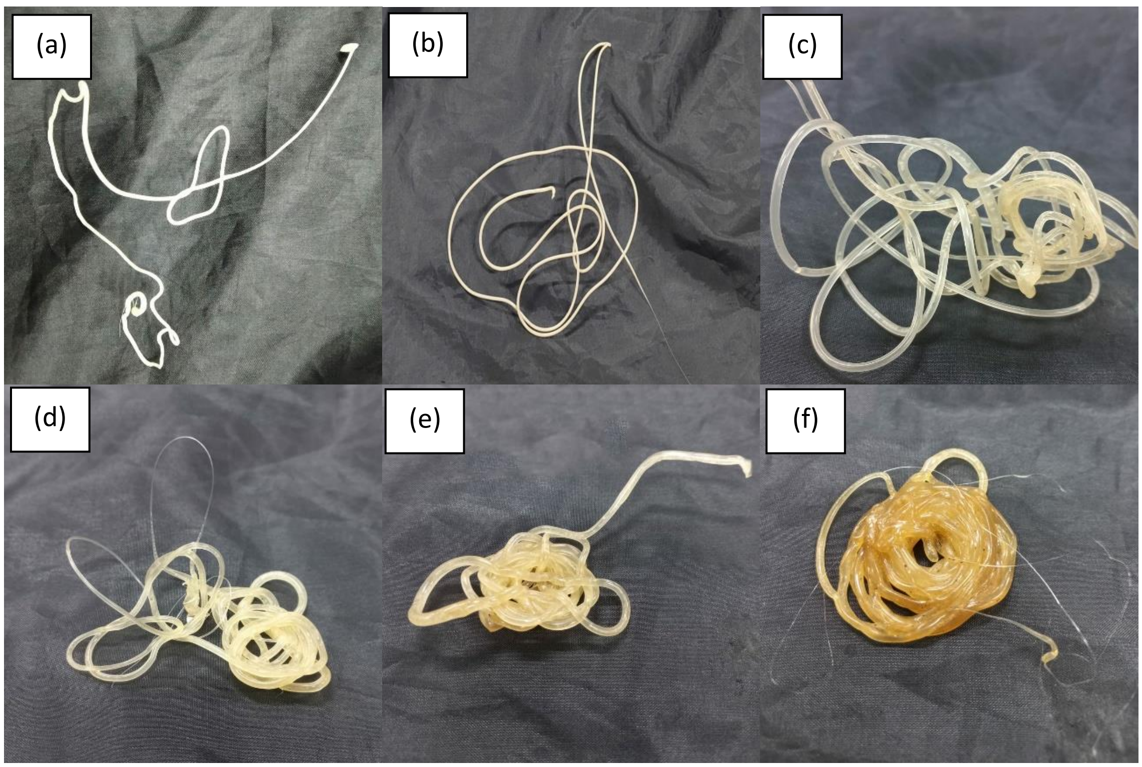

FIGURE 5. Melt flow rate result at (a) first recycle, (b) second recycle, (c) third recycle, (d) fourth recycle, (e) fifth recycle, (f) sixth recycle.

In addition to seeing changes in the MFR, the recycled material also undergoes visual changes, with the color of the material becoming darker, as indicated in Figure 5. This is probably due to a change in the composition of the plastic polymer during processing and application usage because of oxidation degradation. The oxidation degradation process will increase along with the presence of functional groups from the polymer. This is what causes irregular changes in mechanical properties and changes in color [15]. It is also known that compounds containing carbon, when burned in limited air, will produce CO and or CO_2 [16].

IV. CONCLUSION

Based on the results of research on the effect of the frequency of recycling on the flow properties of polypropylene material, it is known that the recycled polypropylene material has an increase in the MFR value, which causes a decrease in its viscosity as the frequency of recycling is increased. The MFR value did not change much until the 2nd recycling, but changed drastically at the 3rd and 4th recycling, which increased 4 times compared to the pure material. And on the 6th, recycling increased up to 7 times. In addition to changes in the MFR value, recycled materials have a darker color shift.

V. FUTHER RESEARCH

Therefore, in the application of recycled plastic materials, it is necessary to add addictive substances. One of the additives that can increase the MFR value of the material is stabilizer. The combination of specific stabilizers and co-stabilizers will make the process stable and maintain the life of the recycled material as long as it is used in an application (product). A good stabilizer for PP is a combination of sterically hindered phenols and phosphite or phosphonate, its combination is able to slow down the increase in the MFR value of PP material until the fifth recycle [15]. Further research will be conducted to identify the best stabilizer composition for retaining the ability of PP material until the sixth recycling.

VI. REFERENCES

[1] L. Sangroniz *et al.*, "Effect of shear rate and pressure on the crystallization of PP nanocomposites and PP / PET polymer blend nanocomposites," *Polymer (Guildf).*, vol. 186, no. October 2019, p. 121950, 2020, doi: 10.1016/j.polymer.2019.121950.

[2] S. Saikrishnan, D. Jubinville, C. Tzoganakis, and T. H. Mekonnen, "Thermo-mechanical degradation of polypropylene (PP) and low-density polyethylene (LDPE) blends exposed to simulated recycling," *Polym. Degrad. Stab.*, vol. 182, p. 109390, 2020, doi: 10.1016/j.polymdegradstab.2020.109390.

[3] R. T. Polymers, "Methods of Recycling , Properties and Applications of Recycled Thermoplastic Polymers," pp. 1–11, 2017, doi: 10.3390/recycling2040024.

[4] Z. M. Ariff, A. Ariffin, S. S. Jikan, N. Azura, and A. Rahim, "Rheological Behaviour of Polypropylene Through Extrusion and Capillary Rheometry."

[5] S. I. Darojat, "Analisis pengaruh waktu pemanasan awal dan massa sampel terhadap hasil uji indeks alir lelehan polipropilena," pp. 41–60, 2008.

[6] H. S. S. Asrof, M.F., dan Torno, "Pengaruh suhu proses dan tekanan injection moulding terhadap kekuatan benturan dan kekerasan pada material High density polyethylene..pdf." Jurnal Prosiding symposium nasional polimer IV Sentra Teknologi Polimer (STP)-BPOLYPROPYLENET, 188-192., 2003.

[7] J. Weszka, M. M. Szindler, M. Szindler, and C. Materials, "Determining the melt flow index of polypropylene : Vistalon 404," pp. 20–21.

[8] A. B. Strong, *Plastics Materials and Processing*, Third Edit. Upper Saddle River, New` Jersey, Columbus, Ohio.: Pearson, Prentice Hall, 2006.

[9] ASTM, "D1238-13 Test Method for Melt Flow Rates of Thermoplastics by Extrusion Plastometer," *Norma*, vol. 08, pp. 1–16, 2010, doi: 10.1520/D1238-13.

[10] H. A. Maddah, "Polypropylene as a Promising Plastic : A Review," vol. 6, no. 1, pp. 1–11, 2016, doi: 10.5923/j.ajps.20160601.01.

[11] I. P. S. A. Vinay Kumar , Rupinder Singh, "Melt Flow Index Melt Flow Index Smart Civil Structures Use of Thermosetting Polymers for," 2020.

[12] B. Standards, "Plastics — Determination of the melt mass-flow rate (MFR) and the melt volume-flow rate (MVR) of thermoplastics," *Int. Stand.*, vol. Fourth Edi, pp. 2; 12, 2005.

[13] Arifianto, "Analisis karakteristik termal pada kabel berisolasi dan berselubung PVC tegangan pengenal 300 / 500 volt = Analysis of thermal characteristic on PVC insulated cable with rating voltage 300 / 500V," *https://lib.ui.ac.id/bo/uibo/detail.jsp?id=126745&lokasi=lokal*, p. 8223.

[14] N. Juniarti, "Penetapan Bobot Jenis dan Rapat Jenis," *J. Univ. Hasanuddin*, pp. 2–6, 2009.

[15] L. Wisojodharmo *et al.*, "PENINGKATAN MUTU MATERIAL DAUR ULANG PLASTIK DENGAN PENAMBAHAN ADITIF," pp. 223–228.

[16] T. H. Pedersen, "Technical and Economic Assessment of Methanol Production from Biogas," no. May, 2012.