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Obtaining an Empirical Equation for Correcting the Melt Flow Index of Virgin and Recycled Polypropylene Mixtures and Analysis of Mechanical Properties of the Blends

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Abstract

Polypropylene is one of the most used materials in the world because of its easy processability and good mechanical properties under different applications. As a result, a lot of polypropylene waste is being generated and recycling is important.The problem with recycling polypropylene is the changing in the properties when compared to virgin polypropylene. Processability parameters and mechanical properties of traction and impact can be changed after recycling this material, affecting companies in the polymer transformation sector. Thus, this work aims to obtain an empirical equation for the correction of the melt flow index of virgin and recycled polypropylene mixtures and also conducts a study on the mechanical properties of the mixtures comparing them with those of virgin polypropylene. The empirical equation found shows a linear relationship between the melt flow index of the mixture and the percentage amount of virgin polypropylene. From the mechanical properties, it was observed that the stiffness of the mixtures is similar to that of virgin polypropylene, however the energy absorbed under impact is considerably lower, compromising the use of recycled polypropylene for such application.

Index terms— recycled polypropylene, melt flow index, mechanical properties of polymers.

1 Introduction

olymeric materials are an important group of the materials engineering because their easy production and wide range of applications. This group includes materials such as plastics and rubbers ranging from low density liquids to rigid solids [1].

The growth in the consumption of plastics in the world is evident. In 1964 around 15 megatons (Mt) were generated, in 2014 this production reached 300 Mt. This production is expected to double again within 20 years, and by the year 2050 it has practically quadruple [2].

Is estimated that if plastics were replaced by other packaging materials, there would be an increase of approximately 60% in the volume of waste produced, 57% electricity consumption over the entire product life cycle and between 78 Mt to 170 Mt of complementary greenhouse gas emissions [3].

Despite the use of plastics in general, more specifically the polymers, to exhibit a decrease in the generation of waste, their discard, as well of other consumable materials, also contributes to the production of urban waste and for the waste in the oceans with the aggravation of not being biodegradable, soon this residue remains for long periods in the nature [4].

One way to mitigate such problem is the reuse of polymeric materials, by secondary or mechanical recycling which according to the Union of the Plastic Material Industry of the State of São Paulo (Sindiplast) and the Environmental Company of the State of São Paulo (CETESB), " **??**?]is the converting of plastic waste into granules that can be reused by the productive sector to make other products [?]" [5].

One of the most used plastics in the world is the polypropylene (PP) wich, due to the great increase in its productive capacity caused its cost to decrease, so it may be used in new much applications. PP has characteristics that allow it to be subjected to many processing techniques such as Injection, Extrusion, Thermoforming, Rotational Molding and Blowing [6]. The main application of PP is in the food sector, representing 32% in products such as: packaging, lids, tupperwares, jars, bottles and gallons. In the consumer goods sector PP represents 17% of the market and is applied in packaging and boxes. In civil construction represents 3% of the market and, among others, it replaces asbestos in fibre cement tiles and water tanks, and it serves as a partial substitute of concrete in slabs with the use of BubbleDeck [7]. Also, it is a raw material for the Pack Less, a plastic pallet. In the automotive sector it represents 9% of the market, being used in dashboards, door panels, bumpers, grilles, for example. [8]. Therefore, all this material when discarded can be recycled and originate new products. In Brazil, about 8.2% of the post-consumer polypropylene is recycled, which is processed into pellets through mechanical recycling and subsequently produces packaging, Year 2020 Abstract-Polypropylene is one of the most used materials in the world because of its easy processability and good mechanical properties under different applications. As a result, a lot of polypropylene waste is being generated and recycling is important.The problem with recycling polypropylene is the changing in the properties when compared to virgin polypropylene. Processability parameters and mechanical properties of traction and impact can be changed after recycling this material, affecting companies in the polymer transformation sector. Thus, this work aims to obtain an empirical equation for the correction of the melt flow index of virgin and recycled polypropylene mixtures and also conducts a study on the mechanical properties of the mixtures comparing them with those of virgin polypropylene. The empirical equation found shows a linear relationship between the melt flow index of the mixture and the percentage amount of virgin polypropylene. From the mechanical properties, it was observed that the stiffness of the mixtures is similar to that of virgin polypropylene, however the energy absorbed under impact is considerably lower, compromising the use of recycled polypropylene for such application. automobile components and other recycled products [9].

However, in its use, recycled PP has different properties in relation to virgin. One of these properties is the Melt Flow Index (MFI), defined by many authors as the material's processability index. The value of MFI is important for defining the polypropylene transformation process and also in the processability and quality of the parts produced with this material, being that PP's that have a higher MFI are more suitable for injection processes [10].

In practice, when a polypropylene is recycled presents a higher melt flow index than when virgin. The recycling causes changes in chemical structure, in the melt viscosity, crystallization behaviour, and tensile and fracture properties. The main effect of recycling is the lowering of the melt viscosity, which is attributed to molecular weight decrease and consequently increases the melt flow index [11]. From experience, it is known that changing the MIF can be responsible for problems during the plastics process transforming such as bubbles, fillers, poor filling of the mold, in injected parts in which this material is used.

In this context, and considering the PP's used for making parts in a plastics processing company in the state of Rio Grande do Sul, Brazil, which uses both recycled and virgin PP's, the purpose of this work is to obtain an equation to correct the melt flow index of virgin and recycled polypropylene mixtures in order to keep it within the processability values required by the company and standardize the material used. To verify if the parts injected with the mixture between virgin and recycled PP's attend the technical specifications, the mechanical properties of traction and impact were tested.

Its important to note that recycled polypropylene, in addition to having an interesting ecological bias, is also more economically viable. Therefore, virgin and recycled PP mixtures can attend projects requirements and economic and environmental demands.

2 II.

3 Materials and Methods

4 a) Materials

Neat polypropylene (PPv), commercial grade PP CP442XP, was provided by Braskem S.A. (Brazil) with a melt flow index of 26g/10 min (230ºC / 2.16 kg), with yield stress of 32 Mpa.

Polypropylene from secondary recycling (PPr) with a many different melt flow index: 5.91, 5.93, 7.70, 8.23, 8.36 and 12.29 g / 10 min (230ºC / 2.16 kg) was provided by Coplast -LTDA from Manaus city (Amazonas, Brazil).

5 b) Methods

6 i. Mixture preparation

The materials used for the measure of the melt flow index were blended in the proportions shown in Table 1. The specimens for the tensile and impact tests were injected in the proporcional for 30% of virgin PP and 70% of recycled PP, following the dimensional requirements of ASTM D638-02a and ASTM D256-02, respectively. Table 2 shows the parameters used during their injection. ii. Obtaining an equation to melt flow index of virgin and recycled polypropylene mixture The obtaining of an equation relating the melt flow index with the different proportions of PPv and PPr, was carried out empirically, carrying out tests with the different mixtures. After

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the rheological tests were performed, MFI x %PPV graphics were generated, and so the linear equations and the general equation was obtained of the melt flow index as a function to percent of virgin polypropylene in the mixtures.

7 c) Characterization

8 i. Melt Flow Index -MFI

The experiment was accomplished in IFRS -Campus Farroupilha, in Farroupilha city (Rio Grande do Sul, Brazil), using an INSTRON CEAST equipment, type 7023, at a temperature of 230°C , 2.16 kg and 420 seconds to stabilization in according to ASTM D1238.

9 ii. Differential Scanning Calorimetry (DSC)

The crystallization (Tc) melting (Tm) temperatures, and percentual of Polypropylene and polyethylene were measured by differential scanning calorimetry on a DSC 6000 PerkinElmer. Samples of 8 mg were analyzed under nitrogen atmosphere under a flow rate of 20 mL/min. In order to erase their thermal history, samples were initially heated at 30°C to 200°C with rates of 50°C/min and kept at 200ºC for 2 min. Then, they were cooled down to 30°C with rates of 20°C/min to determine the crystallization temperature. A second heating to 200ºC with rates of 20°C/min was conducted to evaluate the melting temperature of the composite.

The percentage of polyethylene (PE) and polypropylene (PP) in the composite was calculated using the melt enthalpy reference value obtained from the curves of the samples analyzed. To obtain the results, the method developed by Zamin [12] was used, in which, by the ratio Y = (Î?"HPE / Î?"HPP), is possible to determine PE percentage in the material, by the equation 1:$Y = 0.001X^2 -0.0029X + 0.0804$**(1)**

Where:Y = (Î?"HPE / Î?"HPP) ratio X1 (+) = Percertage of PE in te material X2 (-) = Disregard

iii. Fourier Transform Infrared Spectroscopy -FTIR Fourier transform infrared (FTIR) spectroscopy analysis was performed using a PerkinElmer Frontier spectrophotometer with an attenuated total reflectance (ATR) accessory. Each spectrum was obtained using five scans ranging from 4000 to 600 cm -1 at a resolution of 4 cm -1 .

10 iv. Mechanical properties

The tensile test were performed according to ASTM D638-02a, operating at a speed of 5 mm/min and at room temperature. Five specimens of each mixture were tested and then the mean values of elastic modulus, yield strength and yield strain were obtained. To perform the tensile tests a universal testing machine model WDWV100E, made by Time Group was used, located in the test laboratory of the IFRS -Campus Erechim, controlled by WinWDW software, and has a 100kN load cell and a strain gauge.

Impact tests were performed according to ASTM D256-02 in a controlled environment, with a room temperature of 23 ± 2°C and a relative humidity of 50 ± 10%. The impact strength value was obtained by the ratio of the average energy absorbed by 8 specimens tested according to the dimensions specified in the standard. The tests were performed with a Zwick Izod impact testing machine, located at the IFRS -Farroupilha Campus. The hammer used for the test provides 4.0 J of energy and was released from a height of 610 mm in relation to base.

11 III.

12 Results and Discussion

13 a) Differential Scanning Calorimetry

The Table 3 show the results of melting enthalpy (Î?"H), melting temperature (Tm), crystallization temperature and percentage of polyethylene in the material. The table shows that the melting temperatures of the materials have not changed since it is a blend, both were close to those found in the literature, HDPE 125 °C to 135 °C and PP 160 °C to 170 °C [12,13]. The values shown in Table 3, indicate that the material supplied by the Coplast company is not only composed of polypropylene, but has of 20% polyethylene. Therefore, the MFI variation does not depend on the percentage of polyethylene present in the composition of the material.

14 b) Fourier Transform Infrared Spectroscopy -FTIR

The FTIR-ATR spectra of the recycled polypropylenes (PPr) are presented in Figure 1. The FTIR spectra PPr matrices show characteristic bands of PP [14], with stretching of CH and CH 2 bonds between 2965 and 2820 cm -1 , and asymmetric and symmetrical deformations of CH and CH 3 at 1447 and 1378 cm -1 , respectively. Still in Figure 1, is possible to observe that in PPr4 there are changes in the range from 3600 to 3000 cm-1, related to the O-H groups, showing a peak that may be an indication of a lot of humidity in this sample. Its difficult to distinguish between PP and PE just by the FTIR test, as both have similar bands; which proves the presence of both materials is the DSC test.

In the graph of Figure 1 it is also possible to observe the presence of calcium carbonate in the material, through the absorption bands at 873 and 719 cm -1 , which are characteristic of CaCo 3 [16]. The PP's bands are in accordance with the ones found in the literature and can be seen in Table 4. As the MFI considered ideal is between 9 and 13 (g/10 min) for injection processes in the company requesting the research, to obtain the equation and analyze the mechanical properties, recycled polymers with MFI above 9 (g/10 min) were not used in mixtures with PPv100. The Table 5 shows the measured values of melt flow index and the nomenclature used for each raw material studied. Year 2020

15 c) Melt Flow Index

To evaluate the melt flow index of the raw material with which the company works, rheology tests were carried out on them, the first tests being carried out using 100% of the recycled material (PPr100) and 100% of the virgin material (PPv100). The PPv100 presented a MFI of 26 (g/10 min) according to the value provided by Braskem. The Table 6 shows the results of melt flow index of the recycled polymer samples with different percentages of virgin. Where:

MFIe: Melt Flow Index of the mixture (g/10min)

x: Percentage of PPv in the mixture (%) R: Correlation coefficient of the line (1)

Through the results, the general equation obtained to increase the Melt Flow Index between PPv and PPr mixtures, which is show below:MFIm = MFIr + 0.120712x R=0.988468 (1.6)

Where:

MFIm: Melt Flow Index of the mixture (g/10min) MFIr: Melt Flow Index of the recycled polypropylene (g/10min)

x: Percentage of PPv in the mixture (%) R: Correlation coefficient of the line (1)

Where the MFIr is the linear coefficient of the equation, the value of 0.120712 which is multiplied by the percentage of PPv, it is an average of the slope coefficients of the lines found, and the R (correlation coefficient of the line) is an average of the R's of the all lines, which shows how close to an ideal equation it is. This general equation can be used for any MFIr value as long as mixture with PPv CP202XP.

16 e) Mechanical Properties

After the study of the mixture of virgin and recycled material to correct the MFI value of some recycled PP and obtained the general equation which will solve the company's problem, tensile and impact tests were carried out with the aim of analyzing the mechanical properties of the different mixtures and comparing them with properties of virgin raw material used.

The data obtained by the tensile test are shown in Table 7, where PP M1 has an MFI of 9.50 g/10 min, and PP M2 has an MFI of 10.68 g/10 min. Table 7 shows that with the increase in the melt flow index of polypropylene, the yield strenght of the polypropylene also rises. In a comparison between the two mixtures made PP M1 and PP M2 it is possible to visualize an increase of 4.35% in the yield strength.The virgin material has a value of about 50% above that presented by the mixture between PP V and PP R of IF = 10.691 (g /10 min).For the chosen mixtures the values of IV.

17 Conclusions

In the present work, the mixtures of virgin and recycled polypropylene were analyzed in order to obtain an equation to basically correct the melt flow index required by the company to correct the injection process. The results showed that there is a proportional relationship between the melt flow index and the amount of virgin polymer in the mixture. Therefore, for correction and elevation of the melt flow index of recycled polymer, a certain amount of virgin polymer must be insert to reach the desired index for processing. The evaluation of the mechanical properties showed that the mixtures of virgin and recycled polypropylene maintains rigidity similar that samples with 100% virgin polypropylene, but under impact the mixtures presents a drastic drop in the absorbed energy. Therefore, for applications that require impact, recycled polymers would have their use compromised. Year 2020 elastic modulus (E) are very close, so both samples have the same stiffness. It is also observed that both mixtures have lower E values than virgin material and therefore are less rigid.

As for the yield strain, was observed in table 7 that the values found for both PP M1 and PP M2 are equal. As with the elastic modulus, the highest deformation value is in the virgin material, about 23.5% in relation to PP M2 .

When observed the results of the energy absorbed in the impact, also in the Table 7, is possible to verify that both mixtures of virgin and recycled polymer absorb little energy under impact when compared to 100% virgin material.

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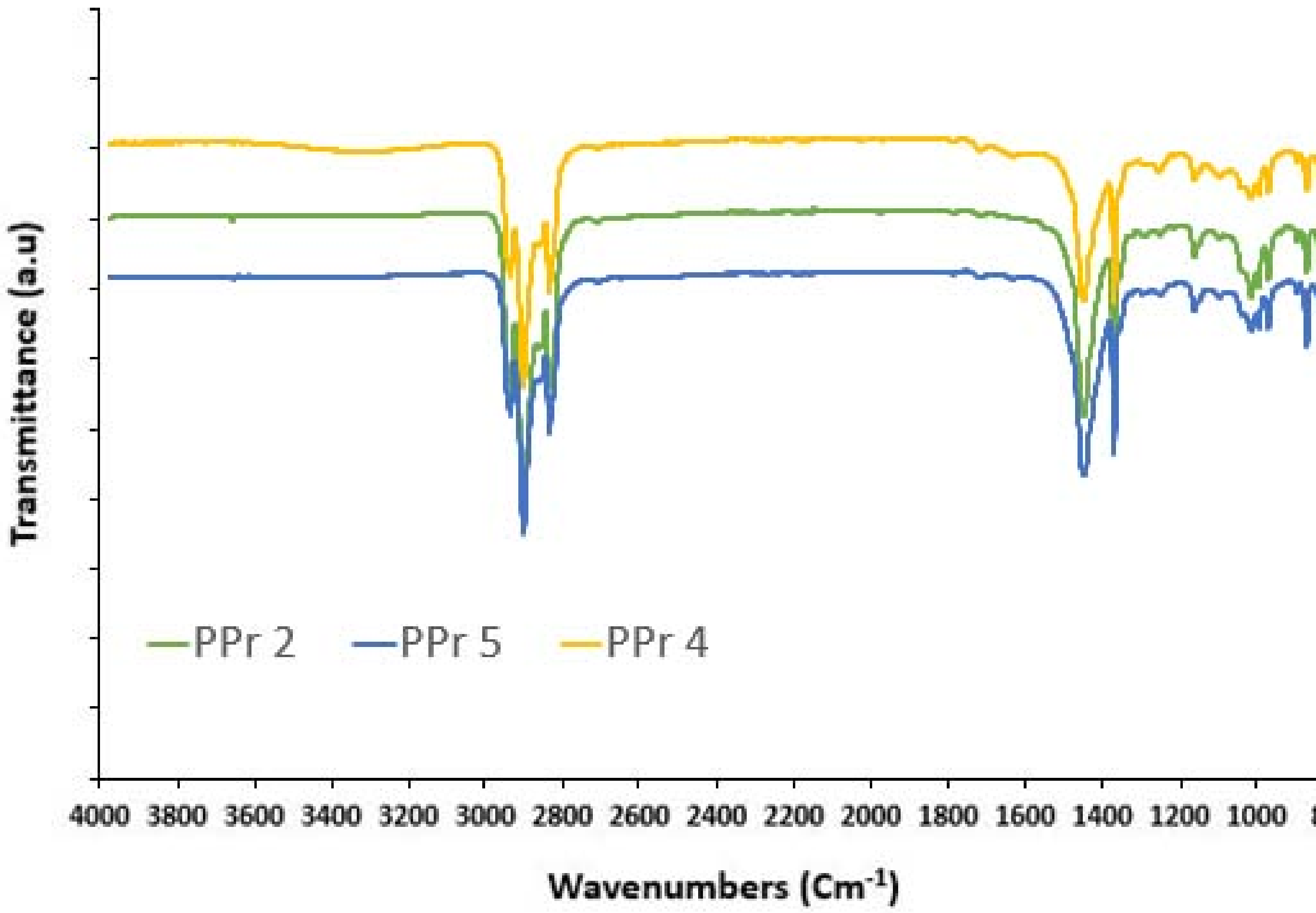

1

Figure 1: Figure 1 :

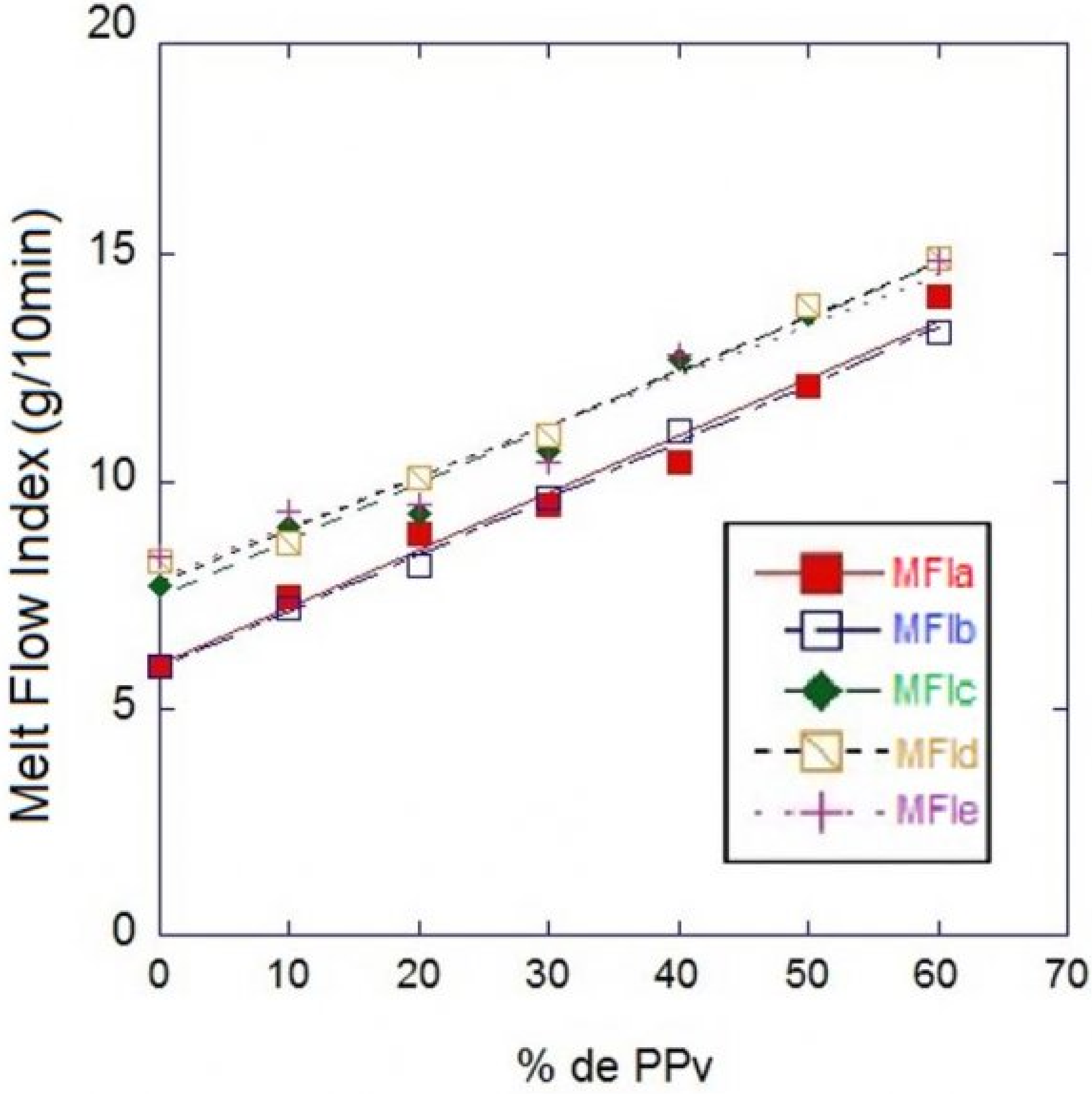

Figure 2:

1

Sample	Virgin (%)	Composition Recycled (%)
PPv100	100	-
PPr10	90	10
PPr20	80	20
PPr30	70	30
PPr40	60	40
PPr100	-	100

Figure 3: Table 1 :

2

Injection Parameter	Value
Maximum holding pressure (MPa)	35
Maximum injection pressure (MPa)	50
Maximum back pressure (MPa)	8
Decompression pressure (MPa)	55
Injection time (s)	5
Temperature of zones 1 to 4 (°C)	200
Temperature of zone 5 (injection) (°C)	195

Figure 4: Table 2 :

3

Sample	MFI (g/10min)	Tm PP (ºC)	Tm PE (ºC)	Î?"H PP (J/G)	Î?"H PE (J/G)	Tc (ºC)	Y = Î?"HPP Î?"HPE/	% PE
PPr2	5.912	162.83	128.88	42.08	22.09	118.09	0.525	22.19
PPr4	8.36	162.34	128.03	46.72	20.35	118.16	0.436	19.51
PPr5	7.70	163.12	128.86	43.38	19.48	118.14	0.449	20.19

Figure 5: Table 3 :

4

Wavenumber (cm -1) PP Pe	Peak Assignment	Reference
2952, 2918 and 2838	C-H stretching	[14]
2720	CH bending and CH3 stretching	[15]
1456	CH3 asymmetric deformation	[15]
1376	CH3 symmetric deformation	[15]
1165	Bending vibration of tertiary carbon	[15]
974, 841 and 808	C-H deformation out-of-plane	[15]
873	C-O, calcite, CaCO3	[17]
719	CO3 , stretching	[18]

Figure 6: Table 4 :

5

MFI measured (g/10 min)	MFI PP
26.00	MFI PPv100
12.286	MFI PPr1
5.912	MFI PPr2
5.93	MFI PPr3
8.36	MFI PPr4
7.70	MFI PPr5
8.23	MFI PPr6

Figure 7: Table 5 :

6

Percentage of virgin PP (% PPv)	MFI PPr2 (g/10 min)	MFI PPr3 (g/10 min)	MFI PPr4 (g/10 min)	MFI PPr5 (g/10 min)	MFI PPr6 (g/10 min)
0	5.912	5.93	8.36	7.70	8.23
10	7.45	7.2	9.33	8.64	9
20	8.83	8.15	9.47	10.06	9.28
30	9.5	9.64	10.4	11	10.68
40	10.4	11.13	12.81	12.45	12.71
50	12.1	13.3	14.446	13.9	13.67
60	14.1	13.3	14.88	14.92	14.91

Figure 8: Table 6 :

7

Year 2020 32 ( ) Volume Xx X Issue V Version I J lobal Journal of Researches in Engineering Gl	Composition PPM1 30% PPv 70% PPr2 PPM2 30% PPv 70% PPr5	MFI (g/10min) 9.50 10.691	? e (Mpa) 20.23 ± 0.49 21.11 ± 0.13	? l 5.67 ± 0.18 5.67 ±0.18	E (Mpa) 1726.40 ± 355.65 1684 ± 176.38	Impact Resistance (J) 0.1225 ±0.0361 0.135 ± 0.0358
	100% PPv Cp 202Xp © 2020 Global Journals	26	32	7	2023.42	3.6195

Figure 9: Table 7 :

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