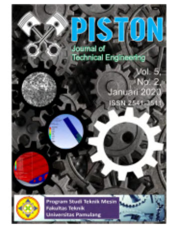
Journal of Technical Engineering: Piston Vol. 5, No. 2, (2022) 63-71

Journal of Technical Engineering:
PISTON



Utilization of Recycled PP-Talc Waste into Composite Products with the Hot Melt Mixing Method

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Received : 5 Septermber 2021 *Revision : 20 September 2021* *Accepted: 5 October 2021*

Abstract: Polypropylene (PP) is one type of plastic material often used. As the materials' specifications get higher, PP is combined with other materials to achieve the desired characteristics. One of them is Talc. Behind its significant growth, there is one main problem, namely the mismanagement of plastic waste that causes environmental pollution. Therefore, one method that can overcome this problem is the recycling method. This research was conducted to investigate the characterization between pure and recycled materials and the optimum composition obtained to get finished goods that were close to materials using pure materials. This research was conducted using PP-Talc scrap with 20% and 30% talc content, then combined with dry and hot melt mixing methods. The alloy material was then tested using a Scanning Electronic Microscope (SEM), Universal Testing Machine (UTM), and Simultaneous Thermal Analyzer (STA). The results showed that the recycled material's morphological changes, mechanical properties, and thermal properties almost resembled the pure material. Specific ratios of 20% and 30% recycled PP-Talc gave rise to new morphological changes and mechanical properties.

Keywords: Polypropylene-talc, recycling, thermal properties, mechanical properties, morphology

***Abstrak*:** *Polipropilena (PP) merupakan salah satu jenis material plastik yang sering digunakan. Seiring dengan spesifikasi dari bahan yang dibutuhkan semakin tinggi, maka PP dipadukan dengan material lain untuk mencapai suatu karakteristik yang diinginkan. Salah satunya adalah Talc. Di balik pertumbuhannya yang signifikan, terdapat satu masalah utama yakni salah kelola limbah dari plastik yang menyebabkan terjadinya pencemaran lingkungan. Oleh karena itu, salah satu metode yang dapat digunakan untuk menanggulangi masalah ini adalah dengan metode daur ulang. Penelitian ini dilakukan untuk mempelajari karakterisasi antara material Murni dan daur ulang serta komposisi optimum yang diperoleh untuk mendapatkan barang jadi yang mendekati bahan yang menggunakan material Murni. Penelitian ini dilakukan dengan menggunakan scrap PP-Talc dengan konten talc sebesar 20% dan 30%, kemudian dipadukan dengan menggunakan metode dry mixing dan hot melt mixing. Material paduan kemudian diuji dengan menggunakan Scanning Electronic Microscope (SEM), Universal Testing Machine (UTM), dan Simultaneous Thermal Analyzer (STA). Hasil penelitian menujukkan bahwa perubahan morfologi, sifat mekanik, dan sifat termal dari material daur ulang hampir menyerupai material murninya. Perbandingan tertentu dari PP-Talc daur ulang 20% dan 30% menimbulkan perubahan morfologi dan sifat mekanik yang baru.*

Kata kunci: *Polipropilena-talcum, daur ulang, sifat termal, sifat mekanik, morfologi*

INTRODUCTION

Polypropylene (PP) is one type of plastic material often used. Its use is very dominant because PP has good characteristics, such as low density, good heat and humidity resistance, and stable dimensional resistance [1,2,3]. As the materials' specifications get higher, PP is combined with other materials to achieve the desired characteristics. One of them is Talc ($Mg_3Si_4O_{10}(OH)_2$). Talc is a metamorphic mineral produced from the

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metamorphosis of magnesian minerals such as serpentine, pyroxene, amphibole, and olivine with the help of carbon dioxide and water [4]. The addition of talc to PP causes the resulting product to become more rigid so that the strength of the material can be increased without having to increase the thickness of the product dimensions. In addition, the addition of talc also increases thermal stability increases melting point, crystallization point, and crystallinity content [5,6,7]. It makes PP with a mixture of talc become one of the materials widely used in injection molding applications, especially for automotive.

Behind its significant growth, there is one main problem, namely the mismanagement of plastic waste, which causes environmental pollution [8,9,10]. Automobile waste comes from the runner, flash, and no good product (NG) during the injection molding. One of the methods is the recycling method to reduce this waste. Two methods can be used, namely primary recycling and secondary recycling [11,12]. Primary recycling refers to the reuse of a product in its original structure. Research related to primary recycling was carried out by [13] examining the effects of talc content and recycling on the properties of PP talc. This research found that the chemical structure and thermal behavior were not significantly affected by recycling, but the mechanical properties, especially break strain, decreased. Yield stress for PP/talc 80/20 was slightly lower than PP/talc 90/10 because the damage mechanism caused by matrix/filler debonding on PP/talc 80/20 was more severe than for PP/talc 90/10 during dynamic testing [14]. Moreover, it was confirmed by research by [15], which investigated the effects of recycling on PP's rheological and thermomechanical properties with a mixture of 12% Talc and 20% EPDM (Ethylene Propylene Diene Monomer) particles in various passes. This research found that the flow curve was more stable than virgin PP. However, the mechanical properties such as failure strain and impact energy at 20^{o}C experienced a significant decrease. This decrease in mechanical properties was caused by chain cutting (driven by chain winding). The chains with the highest molecular weights were usually broken during degradation and were located close to the macromolecular center [16,17].

Some of the research above showed that the recycling carried out on this PP talc material caused a significant change in mechanical properties [18]. Therefore, this research focused more on using recycled materials with secondary recycling methods. Secondary recycling used heat to melt and process the material into finished goods. This process did not involve any polymer changes during the process. This recycling involved the composition of recycled materials so that it was expected that the properties of the finished goods produced were close to materials using virgin materials. This research was expected to be a solution for handling waste in the automotive sector while reducing raw material costs to increase profits.

METHODS

Material

This research used the remaining products that used PP-talc as raw material. The remaining product was obtained from the radiator net support product and the left drawer cover. Support radiator net was a product that used PP as raw material with a mixture of talc as much as 20% (PT-20) of the material mass. In comparison, the left drawer cover used PP raw material with a mixture of talc as much as 30% (PT-30) of the material mass.Talc content can be seen on technical data sheet of both material. Both materials were recycled by the regrinding process and mixed with specific compositions.

Product Preparation

Both materials were recycled by the regrinding process and mixed with specific compositions. This composition was formed by adjusting the percentage by weight of the obtained material, namely 20% recycled PP-talc material (PTR-20) and 30% recycled PP talc material (PTR-30). The composition of the weight of the two materials can be seen in table 1 as follows:

Table 1. Composition of product preparation

Code	PTR-20 (%)	PTR-30 (%)	PTR-20 (g)	PTR-30 (g)
A	10%	90%	30	270
B	25%	75%	75	225
C	50%	50%	150	150
D	75%	25%	225	75
E	90%	10%	270	30

Product preparation was carried out in two stages: dry and hot melt mixing. The first stage was done by the dry mixing method. The dry mixing method was carried out using a henschel mixer with a capacity of 500 grams with a stirring speed of 1500 rpm. A dry mixer was carried out for three minutes for each sample. It was done to ensure that the two ingredients were well mixed and remove moisture that occurred during the storage of the ingredients. The next mixing stage was carried out by the hot mixing method. This method was carried out using a Thermo-Haake Twin Screw Extruder with a chamber capacity of 38 grams. The hot melt mixing process was carried out with a screw speed of 200 rpm for 4 minutes at a temperature of 225°C. The hot melt mixing method was carried out to produce a mixed product that was evenly distributed and dispersed.

Characterization

Universal Testing Machine (UTM). The PP-talc sample that has been treated with hot melt mixing was then prepared to be a test sample in the form of a micro tensile test sample with dimensions of ± 1mm thickness, ± 15.5 mm gauge length, and ± 5.5 mm width. The micro tensile test sample preparation was made by hot pressing the PP-talc sample mixed with the hot melt mixing method into the micro tensile test sample mold. The micro tensile samples were printed using a hot pressing tool by adjusting the operating temperature of the top plate and the bottom plate of hot pressing at a temperature of 200˚C for the bottom plate and 200˚C for the top plate, where the pressure on the hot-pressing hydraulic tool was set at 40 tons for two minutes. After that, the hot pressing was immediately cooled using water for two minutes. The micro-tensile test was carried out using the Chatillon LF Plus Universal Tensile Machine LLOYD Instruments, Ltd., with a loading capacity of up to 1 kN. The test referred to the ASTM D1708-06 standard, with a 10 mm/min pulling speed.

Scanning Electron Microscope (SEM). This test prepared PP-talc samples by freezing with nitrogen then breaking. Morphological changes due to the recycling process and the mixing process were seen through SEM. SEM testing was carried out using the FE-SEM machine Inspect F50 type, focusing on the morphology formed on the faults in each variable. The results of this test were morphological changes that occurred in the material due to the recycling process.

Simultaneous Thermal Analysis (STA). This test was conducted to determine the thermal behavior of the recycled material. It was caused by the possibility of material degradation of PP-talc and the possibility of changes in the material's melting point. The STA test was carried out using the Perkin Elmer Simultaneous Thermal Analysis 6000 machine. In this test, the sample was heated at 50 – 300^{o}C with a heating rate of 10^{o}C/min. The test used nitrogen gas with a 20 ml/min flow rate.

RESULT AND DISCUSSION

Product Characteristics

The characterization of raw materials was carried out using UTM and SEM. The results of testing the modulus of elasticity and tensile strength of PP Talc can be seen in Table 2.

Table 2. Mechanical properties of PT-20 and PT-30

No.	Material Type	Testing Method	Modulus of Elasticity (MPa)	Tensile Strength (MPa)
1	PP Talcum 20% (PT-20)	ASTM D-1708 - 06	111.7	24.6
2	PP Talcum 30% (PT-30)	ASTM D-1708 - 06	179.2	28.7

The value of the modulus of elasticity for PT-30 was more significant than that for PT-20. It indicated that the addition of talc could reduce the elasticity of PP. In addition, the tensile strength value of PT-30 was higher than the tensile strength value of PT-20. It proved that the talc content in PP affected the value of its tensile strength. The more talc content mixed, the higher the tensile strength of the material [7]. In addition, SEM testing was carried out to see the fracture morphology of the raw material in Figure 1 as follows:

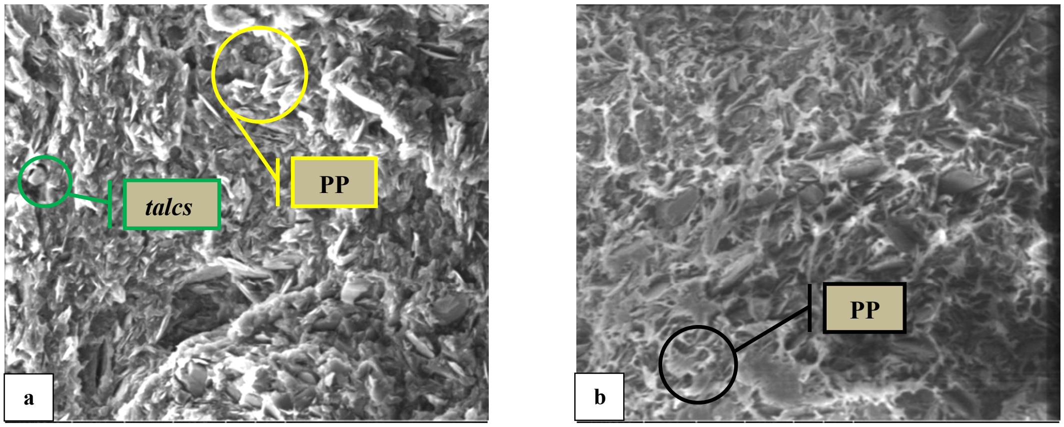

Figure 1. Fracture Morphology Comparison Between (a) PT-20 dan (b) PT-30 with 1000 x Magnification

PT-20 had a fractured shape that PP dominated. The presence of talc particles was complicated to find because the talc particles might be so small that morphological observations made it impossible to estimate the dispersion and distribution of talc. In addition, the fracture surface could be seen with dark or light-colored surfaces. This difference in surface color could occur due to differences in surface depth due to fractures carried out on the test object. Dark-colored surfaces had a deeper surface than light-colored surfaces [19]. At PT-20, the fracture surface still had a slightly dark color. PT-30 in Figure 1b had a fracture surface that resembled PT-20 in Figure 1a. The fracture surface was still dominated by PP and had dark and light-colored surfaces. However, PT-30 had a more even dark surface dispersion than PT-20. This uniform dark surface dispersion could be assumed that PT-30 became more brittle than PT-20 [19]. Furthermore, the morphology of the scrap material can be seen in Figure 2 as follows:

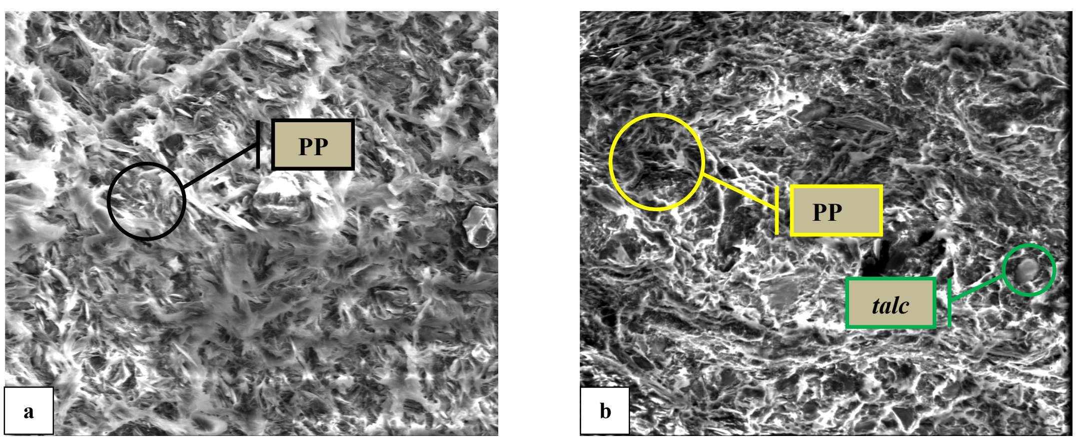

Figure 2. Fracture Morphology Comparison Between (a) PT-20 dan (b) PT-30 with 1000 x Magnification

The recycled material was shown in Figure 2a for PTR-20 and Figure 2b for PTR-30. The fracture surface shown did not significantly differ from the fresh material. The recycled material still had a fracture surface which PP dominated, and there were dark and light surfaces on various surfaces.

The Effects of Waste Composition on Composite Morphology

The comparison between the alloy materials of PTR-20 and PTR-30 can be seen in Figure 3. The morphology of these alloys differed according to the composition of the alloy. PTR-20 (10%)/PTR-30 (90%) had a morphology similar to the dominant material, PTR-30. In contrast, PTR-20 (90%)/PTR-30 (10%) materials morphology was similar to PTR-20. Therefore, it can be assumed that the PTR-20 (10%)/PTR-30 (90%) and PTR-20 (90%)/PTR-30 (10%) materials most likely had properties similar to the dominant material. PTR-20 (50%)/PTR-30 (50%) has a different morphology when compared to PTR-20 (10%)/PTR-30 (90%) or PTR-20 (90%)/PTR-30 (10%). As seen in Figure 3b, the fracture surface formed had dark and light colors spread over almost the entire surface. However, the surface was not dominated by any colors in this distribution. Therefore, it could be assumed that PTR-20 (50%)/PTR-30 (50%) had properties of PTR-20 and PTR-30.

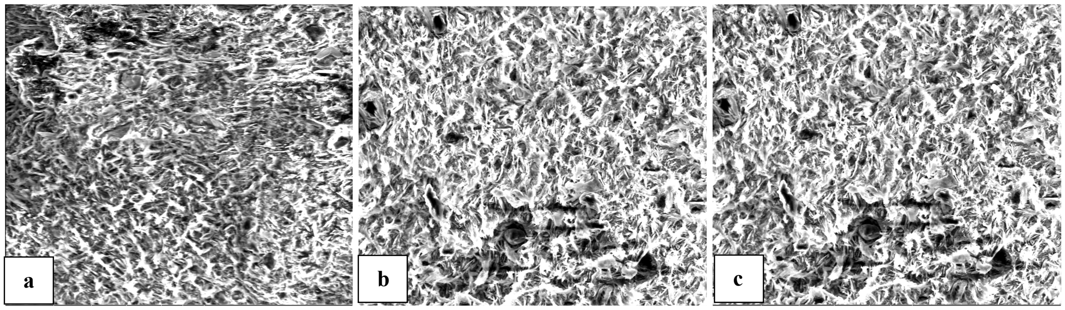

Figure 3. Fracture Morphology Comparison (a) PTR-20% (10%)/PTR-30% (90%) (b) PTR-20% (50%)/PTR-30% (50%) (C) PTR-20% (90%)/PTR-30% (10%) with 1000 x Magnification.

The Effects of Waste Composition on Mechanical Properties of Composites

The mechanical behavior reviewed in this research was the elasticity comparison between virgin material and recycled material. The comparison of the modulus of elasticity can be seen in Table 3 as follows:

Table 3. Mechanical properties of virgin material and recycled material

No	Material Type	Testing Method	Modulus of Elasticity (MPa)
1	PP Talcum 20% (PT-20)	ASTM D-1708 - 06	111.7
2	PP Talcum Recycled 20% (PTR-20)	ASTM D-1708 - 06	108.0
3	PP Talcum 30% (PT-30)	ASTM D-1708 - 06	179.2
4	PP Talcum Recycled 30% (PTR-30)	ASTM D-1708 - 06	144.8

The stiffness decreased in PP-talc recycled material, both 20%, and 30% content. The production process (injection molding) carried out on PP-talc material would cause a decrease in the modulus of elasticity [13]. The heating that occurred during the production process caused the PP-talc to melt. Only PP melted; talc did not melt because the melting point reached 1400^{o}C [21]. The talc granules agglomerated with other talc granules due to the injection process during production by not melting talc during the production process. The agglomerated talc caused the space in the PP-talc to get bigger, causing the material to be more elastic [13]. Particle size of talc was 0.15 µm, so it tends to agglomerate [22]. Therefore, the comparison of the modulus of elasticity and tensile strength of each formed alloy can be seen in Figure 4 and 5 as follows:

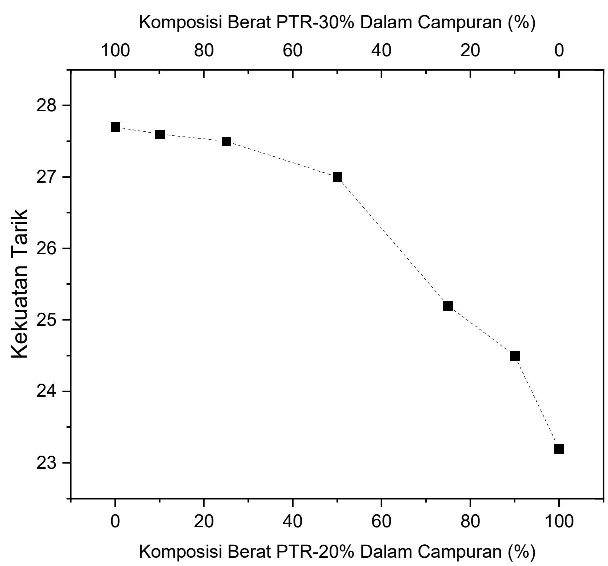

Figure 4. Comparison Graph of the Modulus of Elasticity Value between Test Samples

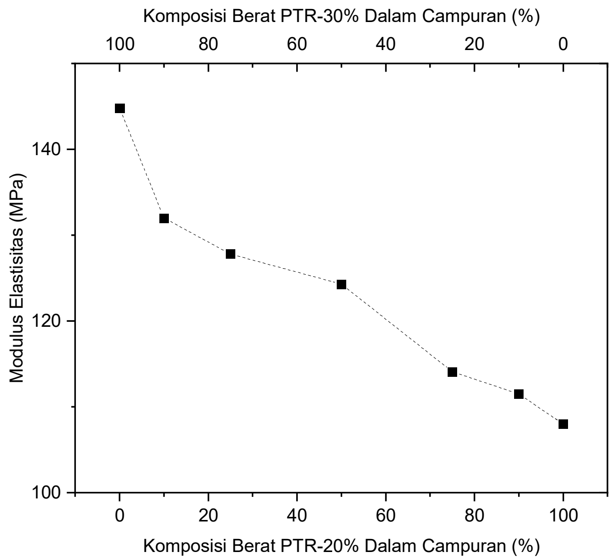

Figure 5. Comparison Graph of Tensile Strength Values between Test Samples

The mechanical behavior of the PTR-20 and PTR-30 alloys was also observed. The value of the modulus of elasticity and tensile strength possessed by the alloy material decreased along with the increase in the composition of PTR-20 in the alloy, as shown in Figures 4 and 5. The value of the modulus of elasticity obtained was close to the dominant material of the alloy. It was also experienced in the value of the material's tensile strength. The value of the material's tensile strength would decrease along with the increase in the composition of PTR-20 in the alloy. The value of the tensile strength approached the dominant material. From these two observations, it can be assumed that the alloy material had a mechanical behavior that resembled the dominant material.

Thermal Properties of Composites

Thermal Properties were studied in this research using the Differential Scanning Calorimetry (DSC) and Thermo Gravimetric Analysis (TGA) testing techniques which were tested simultaneously using a Simultaneous Thermal Analyzer (STA) which can be seen in Figure 6, 7, and 8:

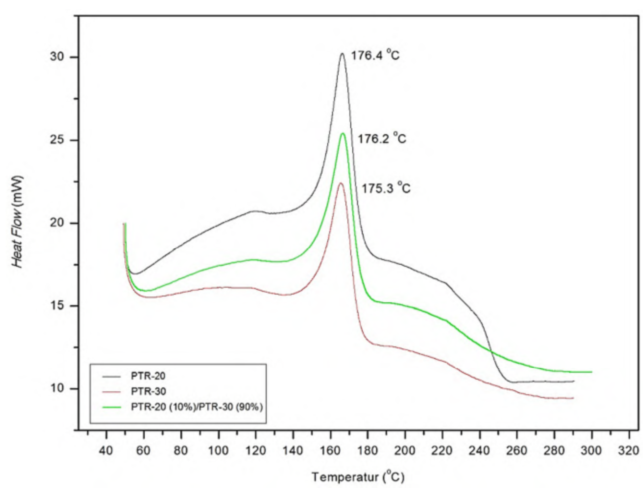

Figure 6. Melting Point Comparison of Test Samples on the Curve Using Differential Scanning Calorimeter (DSC) Technique

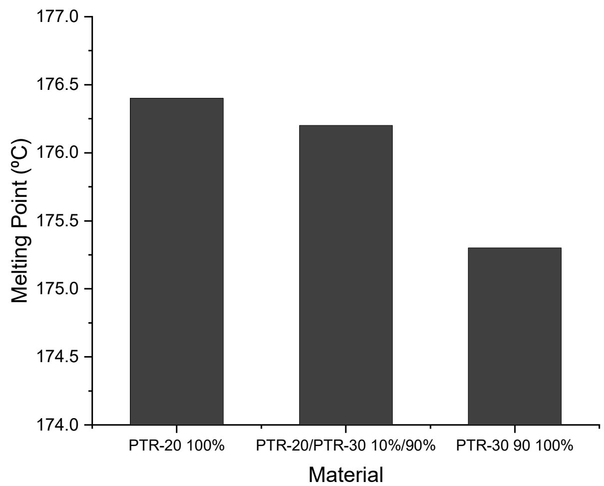

Figure 7. Melting Point Comparison of Test Samples on the Graph

From Figures 6 and 7, the more talc content in the composite, the lower the melting point of PP-Talc. However, the melting point of each sample was not too much different, which is only about 1^{o}C. When compared with the melting point of virgin PP, which was around 176^{o}C [5], the melting point value of the three samples was still within the realm of the melting point of PP. It indicated that the addition of talc in PP would not affect the melting point value of PP. In addition, other factors, such as the use of colors, namely carbon black and titanium dioxide, were neglected due to their use in the composites.

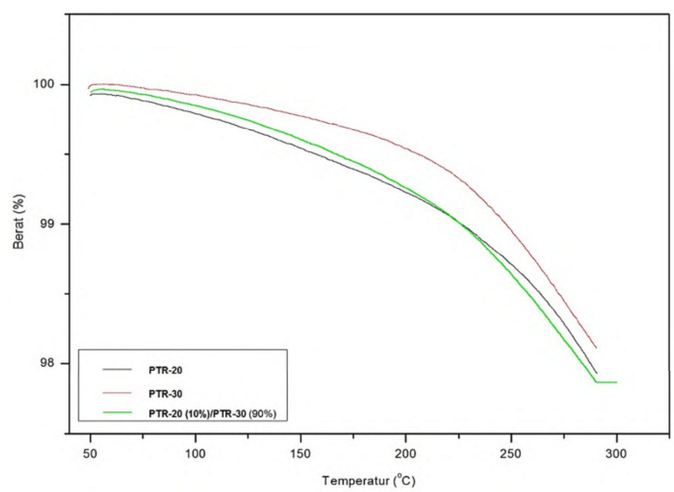

Figure 8. Comparison of Sample Measurement Result Using Thermgravimetric Analysis (TGA) Technique

Based on Figure 8, it can be seen that the thermal stability of each sample was known. The measurement results obtained did not experience significant differences in the three samples. Up to a temperature of 300^{o}C, all three experienced a decrease in mass that was not too large. The magnitude of the decrease in mass in the three samples was also not much different. It was because PP-talc has not experienced material degradation at temperatures below 359^{o}C, and only the release of trapped water vapor occurred during the heating process. When passing through the melting point, the material would change into a liquid state and facilitate the release of water vapor, thus causing a more significant decrease in mass than before passing through the melting point. Therefore, when heated to 300^{o}C, PP-talc recycled material only released water vapor trapped during production and did not experience material degradation [20]. Using temperatures below 300^{o}C, PP-talc recycled material was still feasible to produce based on these measurements.

CONCLUSIONS

Based on the data from the tests and analyzes that have been carried out, several conclusions can be drawn, including:

1. PT-30 had a higher modulus of elasticity and tensile strength than PT-20. The more talc content would increase the stiffness of the composite.
2. The morphology of the recycled PP-talc material did not change much compared to the virgin material. The PTR-20/PTR-30 composite had a morphology similar to the dominant material.
3. There was a decrease in the value of the elastic modulus of the recycled material compared to the virgin material. It was due to the injection molding process causing a decrease in the material's stiffness due to talc agglomeration. The mechanical properties of the PTR-20/PTR-30 alloy tended to resemble the dominant material.
4. The melting point of the recycled PP-talc material and its alloys were around the melting point of virgin PP (± 176^{o} C). PP-talc recycled material and its alloys could still be produced in the production process temperature (injection molding).

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