The Interaction Effect of $CaCo_3$ Composition, Injection Temperature, and Injection Pressure on the Tensile Strength and Hardness of Recycled HDPE

Hendra Prasetyo, Yon Haryono, and The Jaya Suteja

University of Surabaya, Surabaya 60292, Indonesia
jayasuteja@staff.ubaya.ac.id

Abstract. The mechanical properties of recycled High-Density Polyethylene (rHDPE) are inferior compared to non-recycled HDPE. To overcome this problem, Calcium Carbonate ($CaCO_3$) is added to improve the material's mechanical properties. The temperature and injection pressure changes can affect the material's mechanical properties. This research uses a factorial design methodology with two replications to investigate the interaction effect of the CaCO3 composition, injection temperature, and injection pressure on recycled HDPE's tensile strength and hardness. The results showed that the interaction between the $CaCO_3$ composition and temperature, the $CaCO_3$ composition and pressure, and temperature and injection pressure increase the value of tensile strength. The interaction among the three independent variables at a high level has the most influence on tensile strength. The hardness value is influenced by the interaction between the $CaCO_3$ composition and temperature, the $CaCO_3$ composition and pressure, and temperature and injection pressure at all levels. The presence of $CaCO_3$ has the most significant effect on the specimen's hardness. However, the interaction with temperature and pressure reduces the effect of $CaCO_3$ in increasing the hardness value.

Keywords: Interaction Effect, $CaCo_3$ Composition, Injection Parameters, Mechanical Properties, Recycled HDPE.

1 Introduction

High-Density Polyethylene (HDPE) is a thermoplastic polymer made from petroleum. It is the largest constituent of plastic waste and is required to be recycled. The mechanical properties of recycled HDPE (rHDPE) are not the same as those of non-recycled HDPE. According to Tesfaw et al., the tensile strength of rHDPE is inferior to that of non-recycled HDPE [1]. The use of rHDPE as a mixture of non-recycled HDPE leads to an increase in manufacturing efficiency and reduces the extraction of non-recycled material. Manas et al. stated that the higher the percentage of rHDPE mixed with non-recycled HDPE, the lower the hardness value of the mixture [2]. To overcome the decrease in tensile stress of a mixture of non-recycled HDPE and rHDPE, calcium carbonate ($CaCO_3$) can be added to the mixture. $CaCO_3$ is one of the calcium salts that can

M. Hartono et al. (eds.), *Proceedings of the 4th International Conference on Informatics, Technology and Engineering 2023 (InCITE 2023)*, Atlantis Highlights in Engineering 21,
https://doi.org/10.2991/978-94-6463-288-0_15

be found in natural stone and limestone. The $CaCO_3$ has been added to fill thermoplastics for different applications in automotive, packaging, and construction [3, 4].
Previous research has been conducted to investigate the influence of $CaCO_3$ composition on HDPE. The tensile strength and ductility of the compound decrease as the percentage of $CaCO_3$ increases in the range of 5 to 20 percent [5]. And according to Kherici et al., the stiffness of the HDPE decreases with the addition of $CaCO_3$ from 10 to 35 percent [6]. In contrast, the research by Awan et al. showed that the addition of $CaCO_3$ in the range of 5 to 10 percent shows a significant improvement in mechanical results and results in an increase in tensile strength, toughness, impact strength, and hardness [7]. Alshammari et al. also reported that the tensile, flexural, and Izod impact strengths are improved as a function of $CaCO_3$ and fumed silica fillers loading in the HDPE matrix [8]. This research investigates the influence of $CaCO_3$ in the range of 5 to 25 percent In addition, the increase in the percentage of nanoscale $CaCO_3$ from 5 to 20 percent increases the impact strength but does not decrease the yield strength of HDPE [9].
Some researchers investigated the influence of the injection molding process parameters on the mechanical properties of the HDPE. Parameters of the injection molding process that can be varied are temperature, injection pressure, injection time, holding, and cooling time. The effect of the injection temperature and pressure on the mechanical properties of the HDPE is significant. Meanwhile, the injection time, the holding time, and the cooling time affect the production time. Zhu et al. stated that higher temperature and injection pressure increases tensile strength value [10]. Karagöz and Tuna found that a higher melting temperature decreases the strength of tensile and bending [11].
Other research has been conducted to investigate the influence of $CaCO_3$ on recycled HDPE. Atikler et al. found that the tensile strengths of $CaCO_3$-filled compounds show a decreasing trend as the filler loading increases in the range of 10 to 40 percent [12]. The research by Ngothai et al. shows that the tensile strength of the mixture of non-recycled HDPE and rHDPE is slightly affected by the particle size of $CaCO_3$, where the smaller the size, the greater the tensile strength [13]. In addition, the tensile strength of the composites tends to increase with the increase of CaCO3 in the range of 10 to 40 percent.
According to the literature, the mechanical properties of HDPE and rHDPE are influenced by the composition of $CaCO_3$ and various process parameters of injection molding. The results of previous studies indicate that there are different conclusions about the effect of $CaCO_3$ on the mechanical properties of HDPE and rHDPE. In addition, it is important to understand the influence of the $CaCO_3$ composition and injection molding process parameters interaction to improve the mechanical properties of the rHDPE, as the interaction among them may bring different effects compared to the influence of each of them. This research aims to investigate the interaction effect of CaCO3 composition, injection temperature, and injection pressure on the tensile strength and hardness of recycled HDPE.

2 Materials and Methods

The rHDPE used in this research is made of pure HDPE HD5218EA plastic pellets that had been processed by using an injection molding machine, as shown in Figure 1. The pure HDPE plastic pellet has a melt flow index of 18 g/10 min and a 950 kg/m^2 nominal density. The pure HDPE material is heated until it is softened, injected into a mold in the shape of the desired final object, and then cooled until it solidifies again. The injection temperature used to produce the rHDPE is 160 °C and the injection pressure is 150 bar. The final object of the injection molding process is a tensile test specimen according to ASTM D638, as shown in Figure 2 [14]. After that, the tensile test specimen made of HDPE is cut into small pieces by using a crusher machine to produce the recycled HDPE material. The crusher has a 7.5 mm mesh to ensure that the rHDPE size becomes smaller than 7.5 mm.

Fig. 1. Injection molding machine.

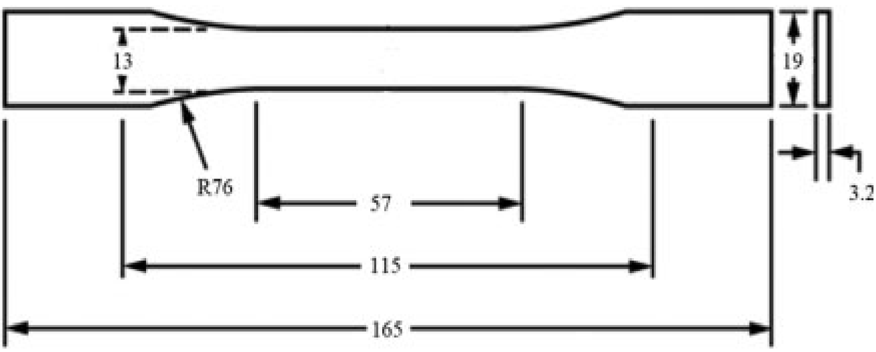

Fig. 2. Tensile test specimen.

The type of $CaCO_3$ used in this research is Precipitated Calcium Carbonate. Before the experiment began, $CaCO_3$ was crushed into powder form. The weight of $CaCO_3$ and rHDPE is measured by using a digital scale according to the investigated composition of $CaCO_3$ and rHDPE. Then, the $CaCO_3$ is mixed with the small piece of rHDPE using a mixer until they are mixed evenly. The investigated values of the $CaCO_3$ composition are 10%, 20%, and 30%. This value is determined based on the preliminary experiment result. The preliminary experiment showed that the composition of $CaCO_3$ up to 30 % increases the tensile strength and hardness of the rHDPE. However, the $CaCO_3$ composition above 30 % reduces the tensile strength and increases the hardness of the rHDPE.

The injection temperature values investigated in this research are 140 °C, 150 °C, and 160 °C. This lowest value is determined based on the melting temperature of HDPE, which is 130 °C. The highest temperature used is 160 °C to ensure that the injection molding process uses the lowest energy for heating. The injection pressure parameter values used in the research are 115 bar, 130 bar, and 145 bar. Based on the preliminary experiments, the use of a pressure of 100 bar resulted in poor specimens because the melted rHDPE had not fully filled the cavity and instead blocked the runner. The use of an injection pressure value of 160 bar for injection temperatures ranging from 150 °C to 170 °C produces flashing because the clamping force cannot hold the pressurized melted rHDPE inside the cavity.

This research uses factorial design methodology with two replications to find the effect of the interaction of $CaCO_3$ composition, injection temperature, and injection pressure on the tensile strength and hardness of rHDPE. The response variables of this research are tensile strength and hardness. The independent variables are $CaCO_3$ composition, injection temperature, and injection pressure. The values of each independent variable are divided into three levels. The values for each level are shown in Table 1. The research conducts 33 x 2 equal to 54 experiments to print 54 tensile test specimens according to ASTM D638, as shown in Figure 2.

Table 1. Value of each independent variable.

Parameters	Levels		
	Low	Middle	High
$CaCO_3$ percentage (%)	10	20	30
Injection temperature (°C)	140	150	160
Injection pressure (bar)	115	130	145

The tensile strength value of each specimen is measured using a Universal Testing Machine GT 7001-L30 universal testing machine manufactured by GOTECH Testing Machines Inc. The hardness value of each specimen is measured using the Shore D durometer according to the ASTM D2240 standard for the hardness test [14]. The durometer has a range between 0 and 100 HD with 0.5 HD resolution.

3 Results and Discussion

The average tensile strength value of the specimens that are injected using various values of $CaCO_3$ composition, injection temperature, and injection pressure are shown in Table 2. The data is analyzed using Analysis of Variance (ANOVA) to determine the effect at each independent variable value. Then, the result of the experiment is followed up by an experiment geometry analysis to determine the quantitative effect of the interaction of the parameters. The experiment geometry is carried out to calculate the magnitude of the effect on the response variable influenced by the value change of the interaction between or among the independent variables.

Table 2. Average tensile strength value.

Temperature	Pressure	10% $CaCO_3$	20% $CaCO_3$	30% $CaCO_3$
140 °C	115 Bar	11.44	11.02	11.23
140 °C	130 Bar	10.82	11.29	10.96
140 °C	145 Bar	10.90	11.21	10.79
150 °C	115 Bar	11.66	11.94	10.60
150 °C	130 Bar	11.65	11.26	12.49
150 °C	145 Bar	11.50	12.14	13.29
160 °C	115 Bar	11.77	11.07	11.81
160 °C	130 Bar	11.89	11.31	12.82
160 °C	145 Bar	10.95	11.83	14.47

Based on the results of the ANOVA analysis in terms of the tensile strength response, the interaction of the $CaCO_3$ composition - temperature, $CaCO_3$ composition - injection pressure, and the interaction of the $CaCO_3$ composition – temperature – injection pressure has a significant influence on the tensile strength of the specimen. However, the interaction of temperature with injection pressure has a less significant effect.

The results of the experiment geometry calculation for tensile strength are shown in Figure 3. The interaction value shows that the increase or the decrease in tensile strength value is caused by the interaction of two or three independent variables for each level. The figure indicates a result similar to that of the result of ANOVA analysis. The interaction between the $CaCO_3$ composition and temperature, the interaction between the $CaCO_3$ composition and pressure, and the interaction between temperature and injection pressure increase the tensile strength value. In each interaction, the $CaCO_3$ composition only increases the tensile strength value at medium to high levels. The highest increase in tensile strength is influenced by the interaction between the three independent variables at a high level. The interaction between the three independent variables at a high level increases the value of tensile strength by 1.55.

The increase in the percentage of $CaCO_3$ increases the value of tensile strength because the presence of CaCO3 increases the degree of crystallinity degree and the entropy of crystallization of rHDPE [16]. As a result, it increases the strength of the molecular bonds of the rHDPE. The presence of $CaCO_3$ also increases the density and increases the interaction between materials [17]. Consequently, the tensile strength increases. However, the interaction between $CaCO_3$ and other process parameters at a low level decreases the tensile strength value. At a low level of temperature, the crystalline ratio decreases and the molecular bonds of the rHDPE with $CaCO_3$ are weakened [11]. At a low injection pressure level, the molecular structure between $CaCO_3$ and rHDPE is less dense than that at high-level pressure [18].

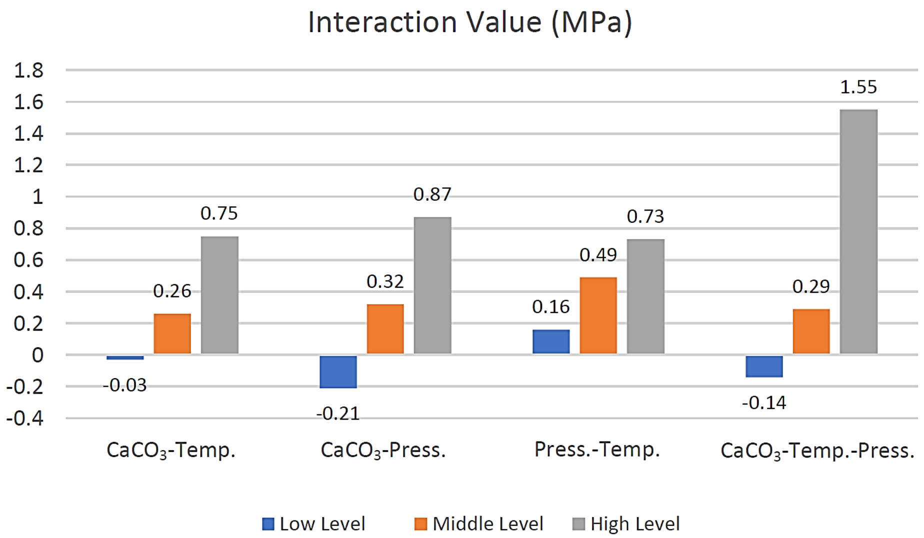

Fig. 3. Interaction value of the tensile strength.

Table 3 shows the average hardness value of the specimens that are injected using various values of the composition of $CaCO_3$, the injection temperature, and the injection pressure. The result of ANOVA shows that only the interaction of the $CaCO_3$ composition with temperature significantly influences the hardness of the specimen. The interaction of the $CaCO_3$ composition with temperature increases the hardness of the specimen by 2.16. Meanwhile, the other interactions have a less significant effect.

Table 3. Average hardness value.

Temperature	Pressure	10% $CaCO_3$	20% $CaCO_3$	30% $CaCO_3$
140 °C	115 Bar	45.75	49.75	50.92
140 °C	130 Bar	46.42	48.50	49.08
140 °C	145 Bar	46.50	49.42	50.33
150 °C	115 Bar	47.17	49.33	51.00
150 °C	130 Bar	47.25	49.83	50.97
150 °C	145 Bar	46.67	49.17	50.92
160 °C	115 Bar	46.33	48.23	48.92
160 °C	130 Bar	46.08	47.51	51.25

160 °C	145 Bar	46.92	49.83	52.50

Figure 4 shows the results of the experiment geometry calculation for the specimen's hardness. The interaction value indicates the influence of the interaction of two or three independent variables for each level on the hardness value. The figure shows that the interaction between the $CaCO_3$ composition and temperature, the interaction between the $CaCO_3$ composition and pressure, and the interaction between temperature and injection pressure at all levels increase the hardness value. The increase of the hardness is influenced by the interaction among the three independent variables only at medium and high levels.

The presence of $CaCO_3$ has the most significant effect on the specimen's hardness. $CaCO_3$ has a higher density compared to HDPE material. Therefore, $CaCO_3$ as a filler increases the density of the rHDPE specimen and the interaction between the materials. As a result, the increase of $CaCO_3$ percentage increases the hardness value. However, the interaction with temperature and pressure reduces the effect of $CaCO_3$, as the crystalline ratio, the molecular bonding between $CaCO_3$ and rHDPE, and the molecular structure of $CaCO_3$ and rHDPE mixture may significantly reduce the effect of $CaCO_3$ in increasing the hardness value. The interaction with temperature influences the hardness value, as the temperature influences the crystalline ratio and the molecular bonds of the rHDPE. In addition, the interaction with injection pressure influences the hardness value because the pressure affects the molecular structure of the $CaCO_3$ and rHDPE mixture.

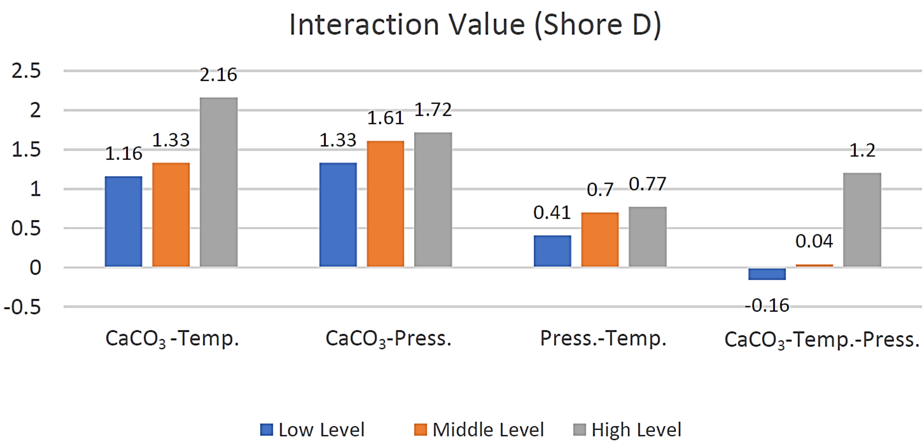

Fig. 4. Interaction value of the hardness.

4 Conclusions

This research investigates the interaction effect of the $CaCO_3$ composition, injection temperature, and injection pressure on recycled HDPE's tensile strength and hardness. The result shows that the interaction between the $CaCO_3$ composition and temperature,

$CaCO_3$ composition and pressure, and temperature and injection pressure increase the value of tensile strength. The percentage of $CaCO_3$ influences the value of tensile strength because the presence of $CaCO_3$ influences the degree of crystallinity degree, the entropy of crystallization of rHDPE, the density, and the interaction between materials. The increase in the percentage of $CaCO_3$ increases the value of tensile strength. The temperature affects the crystalline ratio and the molecular bonds of the rHDPE with $CaCO_3$. The increase in crystalline ratio and the molecular bonds increases the tensile strength. The injection pressure influences the molecular structure between $CaCO_3$ and rHDPE. The denser the molecular structure between $CaCO_3$ and rHDPE increases the tensile strength. Therefore, the interaction among the three independent variables at a high level has the greatest influence on tensile strength. The interaction between the three independent variables at a high level increases the value of tensile strength by 1.55. In addition, the hardness value is influenced by the interaction between the $CaCO_3$ composition and temperature, $CaCO_3$ composition and pressure, and temperature and injection pressure at all levels. The interaction of the $CaCO_3$ composition with temperature increases the hardness of the specimen by 2.16. The presence of $CaCO_3$ has the most significant effect on the specimen's hardness. $CaCO_3$ increases the density of the rHDPE specimen and the interaction between the materials. As a result, the presence of $CaCO_3$ increases the hardness of the specimen. The temperature affects the crystalline ratio of the rHDPE with $CaCO_3$. The decrease in crystalline ratio decreases the hardness. The injection pressure influences the molecular size of $CaCO_3$ and rHDPE. The coarser the molecule of the specimen, the less the hardness value of the specimen. Therefore, the interaction with temperature and pressure reduces the effect of $CaCO_3$, as the crystalline ratio and the molecular bonding between $CaCO_3$ and rHDPE reduce the effect of $CaCO_3$ in increasing the hardness value.

References

1. Tesfaw S, Bogale TM, Fatoba O.: Evaluation of tensile and flexural strength properties of virgin and recycled high-density polyethylene (HDPE) for pipe fitting application. Materials Today: Proceedings 1(62), 3103-13 (2022).
2. Manas D, Manas M, Mizera A, Stoklasek P, Navratil J, Sehnalek S, Drabek P.: The high density polyethylene composite with recycled radiation cross-linked filler of rHDPEx. Poymers 10(12), 1361 (2018).
3. Peng Y, Musah M, Via B, Wang X.: Calcium carbonate particles filled homopolymer polypropylene at different loading levels: mechanical properties characterization and materials failure analysis. Journal of Composites Science 5(11), 302 (2021).
4. Hesabi M, Brennan S, Boulard Q, Le Blanc F, Major I.: On the possibility of replacement of calcium carbonate by a high-performance, economically viable filler in polyethylene composites. Journal of Thermoplastic Composite Materials 14(08927057211031956), (2021).
5. Elleithy R, Ali I, Al-haj Ali M, Al-Zahrani SM.: Different factors affecting the mechanical and thermo-mechanical properties of HDPE reinforced with micro-CaCO3. Journal of reinforced plastics and composites 30(9), 769-80 (2011).
6. Kherici S, Benouali D, Nouredine C.: The Effects of Calcium Carbonate Filler on HDPE Pipe. Advances in Science and Technology. Research Journal 16(3), 213-8 (2022).

7. Awan MO, Shakoor A, Rehan MS, Gill YQ.: Development of HDPE composites with improved mechanical properties using calcium carbonate and NanoClay. Physica B: Condensed Matter 606(412568), (2021).
8. Alshammari BA, Alenad AM, Al-Mubaddel FS, Alharbi AG, Al-Shehri AS, Albalwi HA, Alsuabie FM, Fouad H, Mourad AH.: Impact of Hybrid Fillers on the Properties of High Density Polyethylene Based Composites. Polymers 14(16), 3427 (2022).
9. Yuan Q, Shah JS, Bertrand KJ, Misra RD.: On processing and impact deformation behavior of high density polyethylene (HDPE)–calcium carbonate Nanocomposites. Macromolecular Materials and Engineering 294(2), 141-51 (2009).
10. Zhu J, Chen JC, Kirby E.: Tensile strength and optimization of injection molding processing parameters using the Taguchi method. The International Journal of Modern Engineering 4 (2), (2004).
11. Karagöz İ, Tuna Ö.: Effect of melt temperature on product properties of injection-molded high-density polyethylene. Polymer Bulletin 78(10), 6073-91 (2021).
12. Atikler U, Basalp D, Tihminlioğlu F.: Mechanical and morphological properties of recycled high-density polyethylene, filled with calcium carbonate and fly ash. Journal of applied polymer science 102(5), 4460-7 (2006).
13. Ngothai Y, Putra H, Ozbakkaloglu T, Seracino R.: Effect of $CaCO_3$ size on the mechanical properties of recycled HDPE. The University of Adelaide, Australia, (2009).
14. Standard AS.: D638-14: Test method for tensile properties of plastics. West Conshohocken, USA: ASTM International (2014).
15. Standard AS.: D2240, Standard Test Method for Rubber Property-Durometer Hardness. ASTM International, West Conshohoken, PA, USA (2005).
16. Eiras D, Pessan LA. Influence of calcium carbonate nanoparticles on the crystallization of Polypropylene. Materials Research 12:523-7, (2009).
17. Li J, Tang C, Wang D, Pei X, Shi B. Effect of discrete fibre reinforcement on soil tensile strength. Journal of Rock Mechanics and Geotechnical Engineering 6(2), 133-7 (2014).
18. Maries GR, Chira D, Bungau C, Costea T, Moldovan L. Determining the influence of the processing temperature by injection and of the subsequent pressure on the surface's hardness and indentation modulus of the products made of HDPE, PMMA, PC+ ABS through nanoindentation-G-series basic hardness modulus at a depth method. Mater Plast 54(2), 214-20 (2017).