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PHYSICOMECHANICAL AND RHEOLOGICAL PROPERTIES OF COMPOSITE POLYMER GRANULES PREPARED FROM POLYPROPYLENE AND POLYETHYLENE WASTE REINFORCED WITH MINERAL FILLERS

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Abstract

This study prepared composite polymer granules based on polypropylene (PPJ160) and hig h-density polyethylene (HDPE 7000F) waste modified with bentonite and calcium carbonate ($CaCO_3$), and comprehensively investigated their physicomechanical and rheological properties. The compositions were compounded using a twin-screw extruder, and the melt flow index (MFI), tensile strength, elongation at break, flexural strength, elastic modulus, and impact toughness were determined using standard test methods. The results showed that increasing the HDPE content improved the flowability and deformability of the composites, while slightly reducing their tensile strength. Bentonite-modified composites demonstrated higher tensile strength and elongation, whereas the addition of $CaCO_3$ increased flexural strength and elastic modulus. The PHBC series exhibited superior structural properties. PHBC-1, containing 70% PPJ160, 10% HDPE, 5% MAPP, 10% bentonite, and 5% $CaCO_3$, was recommended as the optimal composition. The obtained results confirm the possibility of producing high-performance composite granules through the integrated recycling of polymer waste with local mineral fillers.

Keywords: *polypropylene, polyethylene, composite, polymer, granules, bentonite, calcium carbonate, MAPP, rheology, material*

Introduction

The recycling of polymer waste has become an important approach to mitigating environmental pollution and promoting the efficient utilization of secondary raw materials. In particular, the modification of polypropylene (PP) and high-density polyethylene (HDPE) waste with mineral fillers has attracted considerable attention due to its potential for producing composite materials with enhanced performance characteristics. Bentonite is widely used as a reinforcing filler because of its high specific surface area and excellent reinforcing capability, whereas calcium carbonate ($CaCO_3$) improves the stiffness and flexural strength of polymer composites. In addition, maleic anhydride-grafted polypropylene (MAPP) acts as an effective compatibilizer, enhancing the interfacial adhesion between polymer phases and improving the overall mechanical performance of the composites. The present study aims to prepare composite polymer granules based on polypropylene and polyethylene waste modified with bentonite and $CaCO_3$, to evaluate their rheological and physicomechanical properties, and to identify the optimum composition for potential applications in the production of construction materials.

Materials and Methods

Composite polymer granules prepared from polypropylene waste (PPJ160), high-density polyethylene (HDPE 7000F), maleic anhydride-grafted polypropylene (MAPP) as a compatibilizer, bentonite, and calcium carbonate ($CaCO_3$) were used as the materials for this study. To evaluate the reinforcing effect of bentonite and the stiffening effect of $CaCO_3$, two groups of composite formulations were prepared. The first group (PHB) contained 10 wt.% bentonite, whereas the second group (PHBC) consisted of 10 wt.% bentonite and 5 wt.% $CaCO_3$. The compositions of the prepared composite formulations are presented in (Table 1).

Table 1. *Composition of Composite Polymer Granules Prepared from Polypropylene and Polyethylene Waste with Mineral Fillers*

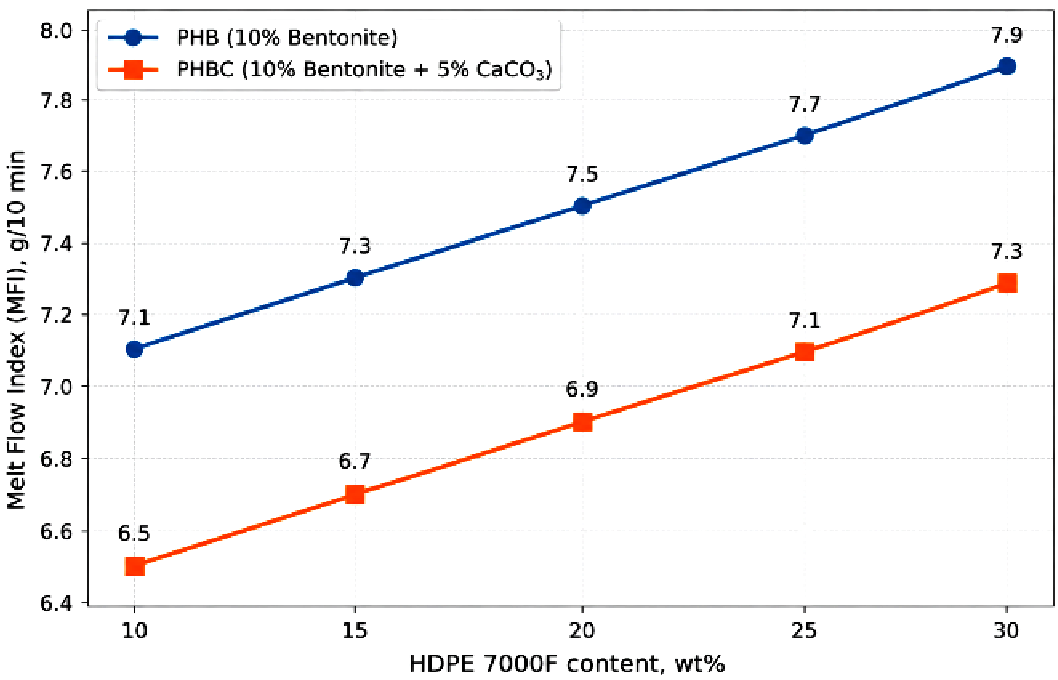
Sample Code	PPJ160 (wt.%)	HDPE 7000F (wt.%)	MAPP (%)	Bentonite (wt.%)	$CaCO_3$ (wt.%)
PHB-1	75	10	5	10	–
PHB-2	70	15	5	10	–
PHB-3	65	20	5	10	–
PHB-4	60	25	5	10	–
PHB-5	55	30	5	10	–
PHBC-1	70	10	5	10	5
PHBC-2	65	15	5	10	5
PHBC-3	60	20	5	10	5
PHBC-4	55	25	5	10	5
PHBC-5	50	30	5	10	5

Prior to the preparation of the composite granules, PPJ160 polypropylene waste and HDPE 7000F were shredded, while bentonite and calcium carbonate ($CaCO_3$) were dried at 105 °C to remove moisture. All components were weighed according to the specified formulations and dry-mixed in a laboratory mixer for 10–15 min. The prepared mixtures were compounded using a twin-screw extruder at processing temperatures ranging from 170 to 210 °C and a screw rotation speed of 60 ± 5 rpm. The extrudates were subsequently cooled in a water bath and pelletized into granules with a size of 3–5 mm. Before testing, all specimens were conditioned at 23 ± 2 °C and 50 ± 5% relative humidity for at least 24 h. The processing characteristics of the composite granules were evaluated by measuring the melt flow index (MFI) according to ASTM D1238 (ISO 1133) at 230 °C under a load of 2.16 kg. The MFI values were expressed in g/10 min. The tensile properties, including tensile strength and elongation at break, were determined using a universal testing machine in accordance with ASTM D638 (ISO 527–2). The maximum tensile stress (MPa) and elongation at break (%) were recorded during testing. The flexural strength and flexural modulus were measured using the three-point bending method following ASTM D790 (ISO 178). The maximum flexural stress (MPa) and flexural modulus (GPa) were calculated. The impact strength of the composite granules was evaluated using the Izod impact test in accordance with ASTM D256 (ISO 180). The impact resistance was expressed as absorbed impact energy in kJ/m^2. For each formulation, at least five specimens were tested. The experimental data were processed and reported as mean ± standard deviation (Mean ± SD). A comparative analysis was performed to evaluate the effects of HDPE 7000F, bentonite, and $CaCO_3$ contents on the rheological and physicomechanical properties of the composite materials.

Results and Discussion

The melt flow index (MFI) is one of the most important parameters for evaluating the processing characteristics of composite polymer granules. It characterizes the flow behavior of the polymer melt and provides an assessment of the material's suitability for processing techniques such as extrusion, injection molding, and other polymer-forming operations. Therefore, the MFI values of composite granules prepared from PPJ160, HDPE 7000F, MAPP, bentonite, and bentonite/$CaCO_3$ were investigated. The experimental results are presented in (Figure 1).

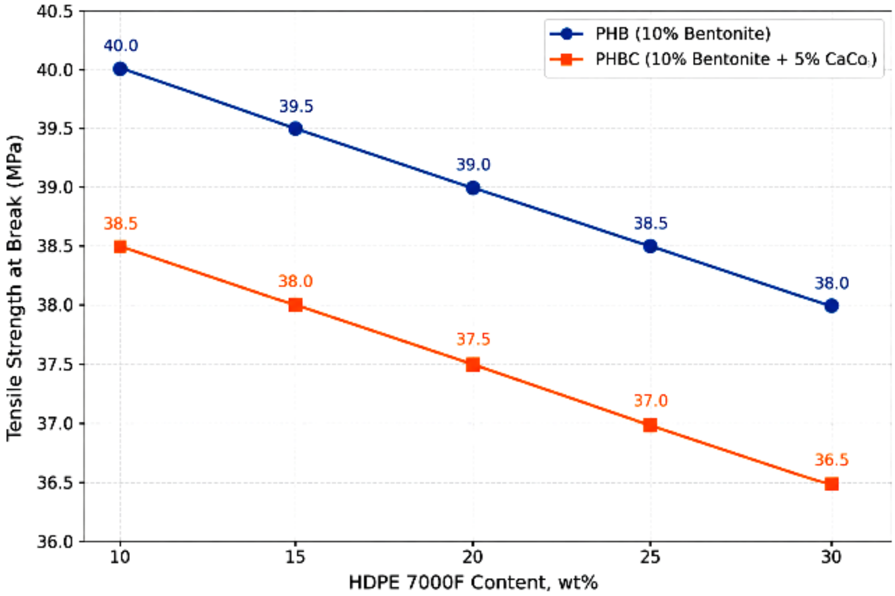
Figure 1. *Effect of HDPE 7000F Content on the Melt Flow Index (MFI) of PHB and PHBC Composite Granules*



A comparative analysis of the PHB and PHBC series (Figure 1) showed that the MFI values of the PHBC composites were consistently lower than those of the corresponding PHB formulations. For example, at an HDPE content of 20 wt.%, the MFI decreased from 7.5 g/10 min for PHB-3 to 6.9 g/10 min for PHBC-3, and a similar trend was observed for all formulations. This reduction is attributed to the addition of 5 wt.% $CaCO_3$, which, together with bentonite, forms a dispersed phase within the polymer matrix, restricts polymer chain mobility, and increases melt viscosity. Nevertheless, the reduction in MFI was moderate, indicating that $CaCO_3$ had only a limited effect on the processability of the composites. The highest MFI values were recorded for PHB-5 (7.9 g/10 min) and PHBC-5 (7.3 g/10 min). Overall, the MFI values ranged from 6.5 to 7.9 g/10 min, demonstrating that all formulations possessed sufficient flowability for extrusion and pelletizing processes.

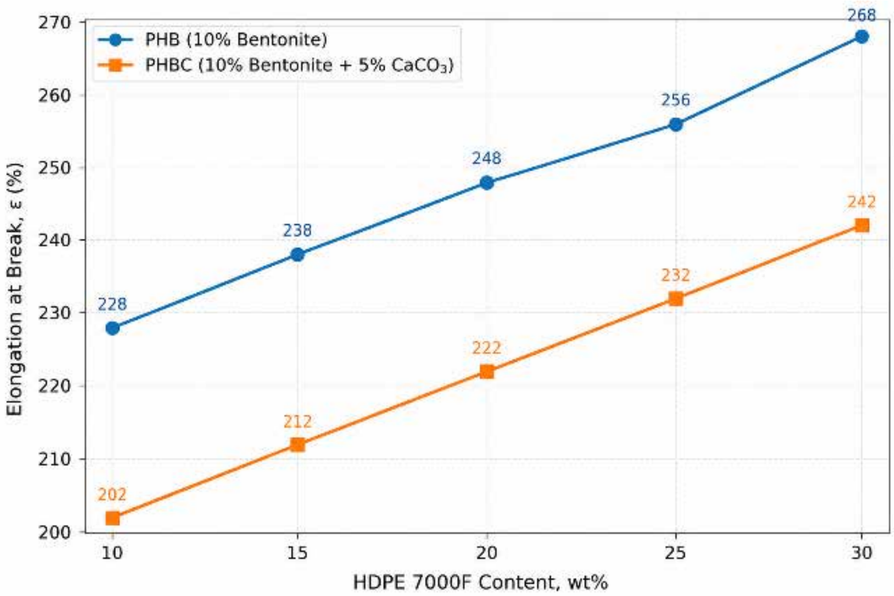
The tensile strength of the PHB and PHBC series of composite polymer granules was also investigated. The experimental results are presented in (Figure 2).

Figure 2. *Effect of HDPE 7000F Content on the Tensile Strength of PHB and PHBC Composite Granules*



As shown in Figure 2, the tensile strength decreased gradually with increasing HDPE 7000F content in both the PHB and PHBC series. In the PHB series, tensile strength (σ_t) decreased from 40.0 MPa (PHB-1) to 38.0 MPa (PHB-5), while in the PHBC series it declined from 38.5 MPa to 36.5 MPa. The highest tensile strength was obtained for PHB-1, owing to its higher PPJ160 content (75 wt.%). Increasing the HDPE content from 10 to 30 wt.% enhanced composite deformability but slightly reduced tensile strength because of the lower fraction of the reinforced crystalline polypropylene phase. The relatively small decrease (only 2 MPa in the PHB series) indicates that MAPP and bentonite effectively improved interfacial adhesion and reinforcement within the polymer matrix.

Figure 3. *Effect of HDPE 7000F Content on the Elongation at Break of PHB and PHBC Composite Granules*



The PHBC series consistently exhibited lower tensile strength than the corresponding PHB formulations. For instance, the tensile strength decreased from 39.0 MPa for PHB-3 to 37.5 MPa for PHBC-3. This reduction is attributed to the incorporation of

5 wt.% $CaCO_3$, which increases stiffness but also creates stress concentration sites during tensile loading, thereby limiting deformation and slightly reducing tensile strength. Overall, the results demonstrate that bentonite-modified composites provide more effective reinforcement than bentonite/$CaCO_3$-filled systems in the PPJ160/HDPE matrix.

In addition, the elongation at break of the PHB and PHBC series of composite polymer granules was investigated. The experimental results are presented in Figure 3.

As shown in Figure 3, the elongation at break increased steadily with increasing HDPE 7000F content in both the PHB and PHBC series. In the PHB series, elongation at break (ε) increased from 228% (PHB-1) to 268% (PHB-5), while in the PHBC series it increased from 202% to 242%. This improvement is attributed to the higher ductility of HDPE compared with polypropylene, which enhances composite flexibility and promotes greater polymer chain orientation during tensile deformation. The highest elongation at break was obtained for PHB-5 (268%), while MAPP further improved interfacial adhesion between the PPJ160 and HDPE phases, preserving composite integrity during deformation. Although the PHBC series exhibited the same increasing trend, its elongation at break remained consistently lower than that of the corresponding PHB formulations. For example, PHB-3 showed an elongation at break of 248%, compared with 222% for PHBC-3. On average, the PHBC composites exhibited 20–30% lower elongation than the PHB series. This reduction is attributed to the incorporation of 5 wt.% $CaCO_3$, whose particles restrict polymer chain mobility and reduce the plastic deformation capability of the matrix. These results indicate that bentonite-modified composites exhibit superior deformation properties while maintaining greater flexibility than bentonite/$CaCO_3$-filled systems. Overall, the highest impact strength among the investigated composites was observed for the PHB-5 formulation, reaching 39 kJ/m², whereas the lowest value was recorded for PHBC-1 (33 kJ/m²). The impact strength (a_n) of all composite formulations ranged from 33 to 39 kJ/m², indicating that the prepared materials possess sufficient resistance to dynamic loading and impact stresses. The physicomechanical and rheological properties of composite polymer granules prepared from PPJ160, HDPE 7000F, MAPP, bentonite, and $CaCO_3$ were comprehensively evaluated. The PHBC series exhibited superior physicomechanical performance compared with the PHB series. In particular, PHBC-1 demonstrated the highest flexural strength (64 MPa) and flexural modulus (2.29 GPa), indicating excellent stiffness and structural performance for construction applications. These findings are further supported by the physicomechanical and rheological data summarized in Table 2.

Table 2. *Physicomechanical and Rheological Properties of Composite Polymer Granules Modified with Inorganic Mineral Fillers*

Sample ID	(MFI) g/10 *min*	Tensile Strength σ_t, MPa	Elongation at Break ε, %	Flexural Strength σ_f, MPa	Flexural Modulus E_{exp}, GPa	Impact Strength a_n, kJ/m²
PHB-1	7,1	40,0	228	62	2,14	35
PHB-2	7,3	39,5	238	61	2,09	36
PHB-3	7,5	39,0	248	60	2,04	37
PHB-4	7,7	38,5	258	59	1,99	38
PHB-5	7,9	38,0	268	58	1,94	39
PHBC-1	6,5	38,5	202	64	2,29	33
PHBC-2	6,7	38,0	212	63	2,24	34
PHBC-3	6,9	37,5	222	62	2,19	35
PHBC-4	7,1	37,0	232	61	2,14	36
PHBC-5	7,3	36,5	242	60	2,09	37

Among the investigated formulations, PHBC-1 (70 wt.% PPJ160, 10 wt.% HDPE 7000F, 5 wt.% MAPP, 10 wt.% bentonite, and 5 wt.% $CaCO_3$) was identified as the optimum composition because of its balanced combination of stiffness, flexural strength, elastic modulus, and impact resistance. This formulation is considered a promising material for manufacturing construction profiles, façade panels, decorative cladding, cable ducts, and other structural products. The incorporation of 15 wt.% mineral fillers (10 wt.% bentonite and 5 wt.% $CaCO_3$) resulted in the best mechanical performance, demonstrating the effectiveness of locally available mineral fillers in reinforcing polymer waste-based composites. These findings confirm that the integrated recycling of polymer waste and mineral fillers is a feasible approach for producing high-performance composite granules for engineering and construction applications.

Conclusion

Composite polymer granules based on polypropylene (PPJ160) and high-density polyethylene (HDPE 7000F) waste, modified with bentonite and calcium carbonate ($CaCO_3$), were successfully prepared, and their rheological and physicomechanical properties were comprehensively evaluated. The results demonstrated that increasing the HDPE content enhanced the melt flow index and elongation at break, while causing a slight reduction in tensile strength. The bentonite-modified PHB series exhibited superior tensile strength and deformation properties, whereas the PHBC series containing $CaCO_3$ showed improved flexural strength and flexural modulus. Among the investigated formulations, PHBC-1 (70 wt.% PPJ160, 10 wt.% HDPE, 5 wt.% MAPP, 10 wt.% bentonite, and 5 wt.% $CaCO_3$) was identified as the optimum composition because it provided the best balance of mechanical and processing properties. These findings demonstrate that the integrated recycling of polymer waste with locally available mineral fillers offers an effective approach for producing high-performance composite polymer granules suitable for construction materials and other structural engineering applications.

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