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Research Article

Investigation of the influence of mineral fillers on the structural and mechanical characteristics of polyethylene high-density (PEHD) composites reinforced with alumina and talc

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Abstract

This study explores the impact of incorporating a blend of alumina and talc, ranging from 1 to 7%, into high-density polyethylene (HDPE) to modify its structural, rheological, and mechanical properties. Comparative evaluations with HDPE/Alumina and HDPE/Talc composites were conducted. The dual-filler combination was achieved through dry grinding, followed by melt processing with a Brabender plastograph. FTIR spectroscopy analysis of HDPE/ (alumina + talc) composites revealed interactions between aluminum and oxygen, with a peak at 700 cm-1 indicating alumina integration into the HDPE matrix, influencing composite properties. Impact strength decreased with talc inclusion but significantly improved with alumina. Notably, a 5% blend of HDPE/(alumina-talc) fillers exhibited the highest impact resistance. Tensile stress showed peaks at 1% talc content and a 3% filler mixture, emphasizing synergistic effects and talc's superiority over alumina. Synergism in elongation at break was observed for HDPE/ (alumina + talc) composites. Additionally, HDPE/Alumina composites displayed the highest modulus of elasticity, while HDPE/(Alumina + Talc) composites had the highest melt flow index. Surface treatments promoted uniform filler dispersion within the HDPE matrix, enhancing mechanical properties. Overall, composites with binary fillers harnessed synergistic effects, combining advantages from both components.

1. Introduction

In modern engineering, the versatility of materials is paramount, with engineers adapting materials to suit the evolving needs of society and the demands of cutting-edge technologies. The advancement of macromolecular materials, ignited by the advent of synthetic polymers, has spurred the emergence of new materials, revolutionizing industries over recent decades [1, 2]. Polymers, now integral to multitude everyday objects, have progressively supplanted traditional materials such as mineral glasses, ceramics, and metals, owing to their superior attributes in terms of both lightweight construction and durability. In this dissertation, our focus centers on high-density polyethylene (HDPE).

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Polymer composites are complex materials comprised of two or more distinct phases, typically featuring a polymer matrix and reinforcing or filler components. This structural composition enhances properties beyond those of individual constituents. Thermoplastics are often favored over thermosets due to their recyclability and thermoformability. Conventional fillers, such as glass, aramid, and carbon fibers, along with additives like calcium carbonate, play a pivotal role in creating robust yet lightweight materials, thus driving advancements in composite technology [3]. The manufacturing process of composite materials involves several stages that integrate blending components possessing diverse physicochemical properties. These stages typically encompass intricate procedures such as material selection, mixing, polymerization, and molding. Each step contributes to the final composite's desired characteristics, ensuring optimal performance and functionality in various applications [4]. Fillers can incorporate particles, layered materials, fibers, or clusters that are enclosed within natural or synthetic polymers. These polymer composites exhibit significant promise for applications across various industries such as aeronautics, automotive manufacturing, cement production, electronics, medical equipment, consumer goods, and packaging. This is owing to their outstanding properties, which are enhanced by the presence of reinforced fillers [5].

The incorporation of fillers into polymers stands as a key technique for enhancing the properties of finished products and broadening the scope of plastic material applications. Moreover, it represents a cost-effective approach to crafting materials tailored to highly specific needs. Mineral fillers are renowned for their ability to enhance electrical conductivity, thermal resistance, and mechanical strength [6, 7]. Within the scope of this study, our attention is drawn to two mineral fillers: alumina, talc, and their amalgamation. Research conducted by F.Z. Benabid and colleagues [8] has revealed that incorporating alumina (Al_2O_3) into low-density polyethylene (LDPE) enhances its thermal stability and impact resistance. Conversely, O.K. Mallem et al. [9] found that the addition of talc improves the tensile strength of composites. Mechanical treatment of mineral fillers, typically employed, aims to enhance the quality of polymer/filler interfaces, ensuring effective dispersion. Moreover, the researchers [8, 9] demonstrated that severe grinding provides a more effective solution for increasing the specific surface area of the mineral.

According to K. Liu et al. [10], treating talc significantly enhances the Charpy impact strength and tensile strength of polypropylene/talc composites. Polyethylene (PE) finds extensive use across a spectrum of applications. Within the confines of this study, the combination of alumina and talc is introduced into HDPE to enhance specific properties, paving the way for potential applications.

The main objective of this research is to explore the impact of incorporating alumina, talc, and their combination (synergistic advantages) on the properties of HDPE, and to evaluate the efficacy of blending these two additives a distinctive aspect of this initiative. The method of co-mixing through grinding is a fast and economical approach utilized to reduce production costs, prevent filler agglomeration, and improve dispersion within the HDPE matrix.

2. Experiment

2.1. Materials

The polyethylene selected for this study is sourced as HDPE HYA 600 Blow Molding Resin, belonging to the ExxonMobil group- Chemical Company 22777 Springwoods Village Parkway Spring, TX 77389-1425. USA, and is provided in granule form with a density of 0.96 g/cm^3. Talc, specified as ETFINE 8CF, is provided by FOURNIER COMPOSITES SA (ZA DE RAGON 5 RUE DE COULOMB, 44119 TREILLIERES, FRANCE). with a particle size (d80) of approximately 38 μm and a whiteness of 85%.

The alumina GF68527187 used in this study is procured from Sigma-Aldrich Corporation PO Box 14508 Saint Louis, MO 63178 United States.

2.2. Preparation of Polymer/Filler Composites

In the process of creating polymer/filler composites, blends based on high-density polyethylene (HDPE) were meticulously crafted through melt processing. This was achieved using a Brabender plastograph operating at a temperature of 220°C and a rotational speed of 30 rpm for 10 minutes. The composites were systematically prepared, incorporating varying filler proportions (alumina, talc, alumina-talc mixture) set at 1, 3, 5, and 7%.

Following the production process, the resulting composites underwent grinding using a DREHER Brabender mechanical grinder, followed by compression molding to create thin films and test specimens. The preparation of these specimens for diverse mechanical tests involved the utilization of automatic compression press from the CARVER brand. The tests, including tensile and impact assessments, were conducted under specific working conditions: a plate temperature of 200°C, a preheating duration of 10 minutes, a compression time of 5 minutes, and subsequent air-cooling. Sample dimensions conform to ASTM standards.

2.3. Characterization

Infrared analyses (IR) are a crucial tool for discerning the intricacies of chemical bonds within materials. This method involves exciting molecular bonds in a sample using infrared radiation with wavelengths between 2.5 and 5 µm and frequencies from 4500 to 400 cm^{-1}. The specific absorptions within this frequency range offer valuable insights into the compound's structural composition. When the radiation frequency aligns with molecular vibrations, emitted energy is absorbed by the bonds, producing characteristic transmission bands representing elongation and angular deformation vibrations, such as rocking, scissoring, wagging, and twisting [11- 13]. The analyzes were carried out using a "Perkin Elmer 1000" type device with a resolution of 4 cm^{-1}. The different spectra present the transmittance (%) as a function of the wave number (cm^{-1}).

The experimentation involved the utilization of a Charpy testing apparatus, comprising a robust module with a hammer situated at its free end, a designated space for the test specimen, and an indicator dial to measure the absorbed energy during the impact, as outlined in reference [14-16]. This testing device is fundamental in assessing the impact resistance of materials, providing valuable data on their mechanical properties under specific conditions. The non-impact notched specimens were prepared with dimensions of 63x12.7x3mm^3. The Charpy test is conducted using a device of type CEAST 6546/000 with an energy of 7.5 joules.

The tensile test, a pivotal mechanical examination, serves to assess a material's deformation capacity when subjected to fluctuating stress levels. In this destructive procedure, a gradually increasing deformation is applied at a constant rate, and the corresponding force needed for this deformation is measured. This critical test is executed under ambient temperature conditions, as referenced in [17-20]. Through the tensile test, valuable insights into a material's mechanical behavior and its response to varying stresses can be garnered. The tensile specimens, manufactured with dimensions (115x13x3mm), are subjected to the tensile test controlled by a computer. After having fixed the initial length at a value of 115mm, the specimen is embedded between the two jaws, one of which is fixed and connected to a 2000N force sensor. The other jaw is mobile and connected to a drive system having a stretching speed of 5.$mm.min^{-1}$.

The Melt Flow Index (MFI) is a widely employed method in the plastics industry for characterizing thermoplastic materials. Using a melt flow meter, this technique measures the mass of molten polymer flowing under a calibrated load for a specific duration, providing a key indicator of viscosity and molecular weight. Known for its speed and comparability, MFI is crucial for the practical study of a polymer's molten state behavior [21]. The experiments utilized a "MELT-INDEXER" model 5 apparatus, featuring a vertical cylinder in an oven with a standard die. Tests were conducted under a 2.16 kg load at 200°C with Melt flow index (g/10 min)

3. Results and Discussion

3.1. Chemical Structure Analysis

3.1.1. Fourier Transform Infrared Spectroscopy (FTIR)

The Infrared (IR) analysis relies on exciting the molecular bonds of a sample using infrared radiation with frequencies ranging from 4500 to 400 cm^{-1}.

- HDPE/Filler Composites

Fig. 1, depicting the FTIR spectra of HDPE/Al_2O_3 composites incorporating fillers in the range of 1 to 7%, reveals noteworthy trends. The distinctive HDPE bands manifest as a prominent peak between 2800-3000 cm^{-1}, corresponding to the elongation vibration (C-H) of polyethylene, and a peak around 1480 cm^{-1}, indicative of the deformation vibration (C-H) of the polyethylene (CH_2) group. Absorptions within the range of 700-750 cm^{-1} signify the rotational vibration (CH_2) of polyethylene.

The distinctive HDPE bands previously elucidated persist in HDPE/Al_2O_3 composites spectra, albeit with a subtle decline in intensity correlating with an escalating filler content. Notably, the discernible alteration lies in the emergence of two distinct peaks indicative of alumina. Specifically, in the 500-400 cm^{-1} range, there is a manifestation of peaks attributed to Al-O bonds, signifying the interaction between aluminum and oxygen within the composite [22]. Additionally, a discernible peak at 700 cm^{-1} corresponds to the Al-OH bond [22], further elucidating the integration of alumina into the HDPE matrix. These observed shifts in the spectra signify the influence of alumina content on the composite.

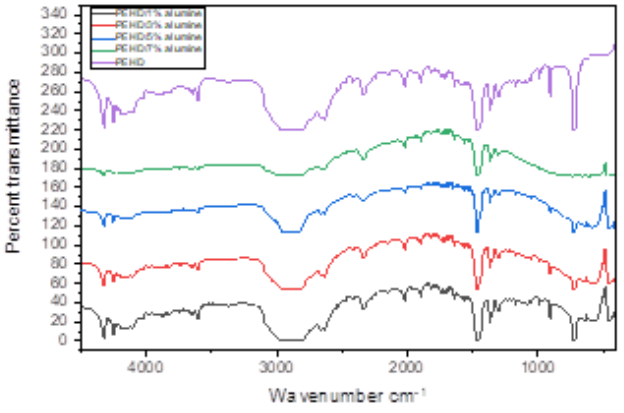

Fig. 1. FTIR spectra of HDPE/alumina composites

The findings from the FTIR spectroscopy analysis of HDPE/ (alumina+talc) composites are presented in Fig. 2. The spectral profile depicted mirrors the characteristic peaks of HDPE and alumina. Notably, the spectrum exhibits the concurrent presence of new absorbance

peaks attributed to talc, indicating their successful dispersion within the HDPE matrix. For talc, the peak at 3700 cm^{-1} is assigned to the surface hydroxyl groups as Si-OH and Mg-OH. The stretching band at 966 cm^{-1} is attributed to the Si-O tetrahedral layer and those at 3674, 664 and 549 cm^{-1} to the MgO/MgOH octahedral layer. The siloxane group (Si–O–Si and Mg–O–Si) stretching vibrational bands exist with intense peaks 460- 420 cm^{-1} [9].This amalgamation signifies the formation of the HDPE/(alumina-talc) composite, elucidating the compositional synergy of the individual components in the resulting material.

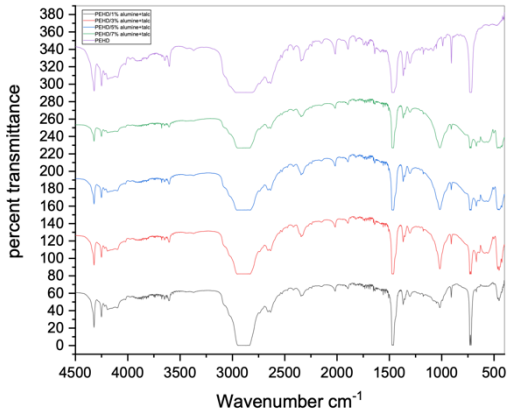

Fig. 2. FTIR spectra of HDPE/alumina+talc composites

3.2. Mechanical Properties

3.2.1. Impact Strength

The impact strength values ak (smooth specimens) of unloaded HDPE samples (ak=9.169 J/cm^2) and composites loaded with 1%, 3%, 5% and 7%, was investigated. Fig. 4 provides a comprehensive overview of the impact strength variation, specifically the Charpy impact strength, across different composites. The results unveil a distinct trend, wherein the introduction of talc is associated with a reduction in impact strength. Conversely, the inclusion of alumina demonstrates a noteworthy enhancement in the impact strength of the composites. This finding aligns with the observations made by Tuen et al [23], who reported superior flexural strength but diminished impact resistance in talc-based composites when compared to their $CaCO_3$-based counterparts.

Remarkably, a pivotal observation surfaces concerning the composite containing a 5% blend of the two HDPE/(alumina-talc) fillers, registering the highest impact resistance value (ak=13.15 J/cm^2). This improvement can be attributed to the finely granulated nature achieved through meticulous grinding during the blending process of the two fillers. This process results in a high specific surface area and an optimized interface [23, 24]. The absence of filler/matrix interaction favors particle/particle interaction, preventing an increase in the particle size of the fillers (brittle zones). This intricate combination mitigates brittleness in the material, underscoring the significance of the blending process in optimizing the composite's mechanical properties.

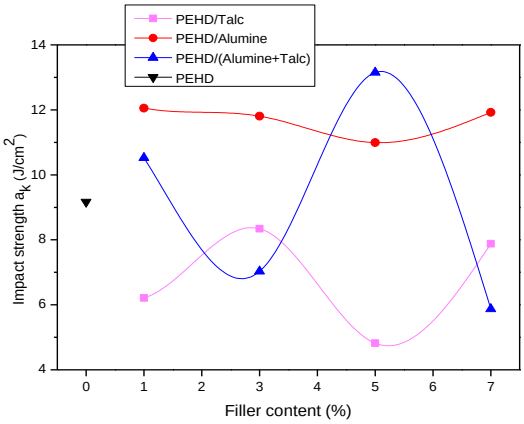

Fig. 3. Variation in Charpy ak impact strength (J/cm^2) of HDPE and its composites as a function of filler content

3.2.2. Tensile Test

Figs. 4-5 and 6 show the variations in tensile properties, i.e. stress at break, and strain at break and modulus of elasticity of the different compositions respectively. From the obtained results, it can be seen that the incorporation of fillers (alumina, talc and their mixtures) in the HDPE matrix has considerably affected its tensile properties.

- Stress at Break

The ultimate tensile stress values for HDPE/mineral filler composites were investigated σr (HDPE)=17.5 MPa Fig. 4 shows an increase in tensile strength, except for an optimum at 1% talc content and a second optimum at 3% mixture of the two fillers (synergism effect), with no improvement for the other composites. It should be noted that talc composition values are higher than those for alumina.

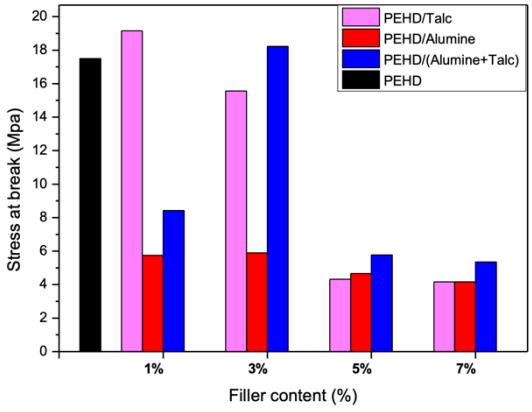

Fig. 4. Variation in Stress at break of HDPE and its composites

- Strain at Break

From Fig 5, we can deduce that there is no improvement in elongation at break by incorporating fillers compared with virgin HDPE.A decrease in this property is indeed

noticeable for composites with higher filler content, which have the lowest values of elongation at break. Synergism in elongation at break was observed for the HDPE/ (alumina+talc) composite with 3, 5 and 7% filler content, compared with the two HDPE/alumina and HDPE/talc composites. More than a synergetic effect, this improvement is due to the fine particle size obtained after milling, as larger particles provide more stress concentrations where a crack can be initiated more easily [22, 23], and in addition to this, the presence of aggregates also creates brittle zones. This means that composites containing a mixture of two fillers have the best dispersion of fine particles in the polymer matrix compared to composites with separate fillers.

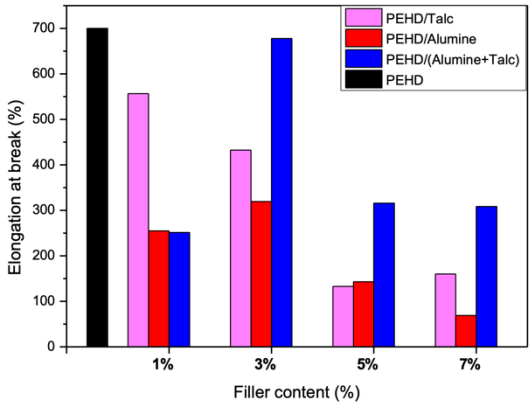

Fig. 5. Variation in elongation at break for HDPE and its composites

- Modulus of Elasticity

The best results in terms of modulus of elasticity were observed for composites containing 5% alumina, 1% talc and 7% mix of two fillers (Fig. 6). It was also found that the highest value of modulus of elasticity E (1200 MPa) was observed for the composite containing 5% alumina and consequently the increase in HDPE stiffness by the addition of fillers.

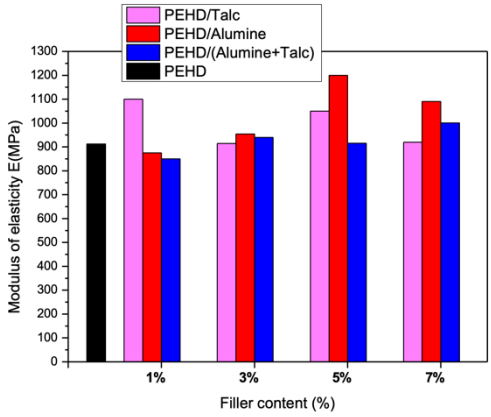

Fig. 6. Variation in modulus of elasticity of HDPE and its composites

The research shows that adding fillers like talc and alumina, or their mix, affects HDPE's tensile properties significantly, even in small amounts. Good dispersion and increased surface area of the fillers toughen the HDPE matrix. Just 1% of untreated talc can improve

HDPE's mechanical properties, likely due to its hydrophobic nature aiding deformation resistance.

Blending the fillers makes the material stiffer, stronger, and more stretchable before breaking, compared to using them separately, thanks to effective dispersion post-grinding. Chemically and mechanically treated talc, alumina, and their mix enhance certain properties of rigid HDPE, showing a synergistic effect when combined.

3.2.3. Rheological Analysis

- Melt Flow Index

The melt flow index is a simple parameter to obtain, and a very useful one which gives us an indication of a polymer's viscosity, branching rate, free volume between chains and degree of crystallinity. Fig. 7 shows the variation in melt index at a temperature of 200°C under a 2.16 kg load for the various HDPE/filler composites.

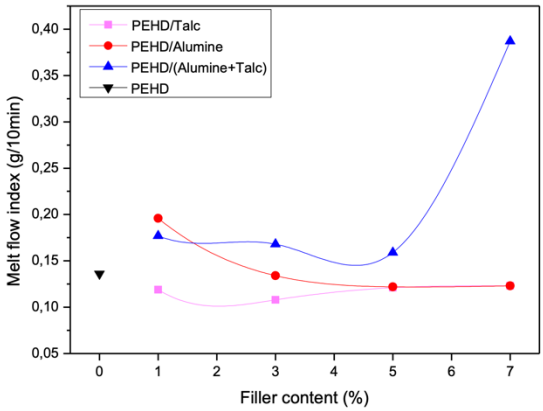

Fig. 7. Variations in the melt flow index of HDPE / filler composites as a function of filler ratio

Fig. 7 shows that the incorporation of talc causes a decrease in the melt flow index of the HDPE/Talc composite, and that the melt flow index value decreases with increasing alumina content, resulting in an increase in viscosity. The composite with binary fillers, on the other hand, has the highest melt flow index value. This is due to the fine particle size obtained by grinding during the preparation of the mixture of the two fillers, which facilitates the orientation of the flow and therefore favors the flow (melt flow) of the material [22]. The good particle dispersion in this case remarkably reduces the Van Der Waals forces between the polymer chains (polymer-polymer interactions), which facilitates the movement of the polymer chains and consequently facilitates flow, with a reduction in HDPE viscosity [25].

4. Conclusions

The aim of this work was to develop high-density polyethylene/(alumina-talc)-based composites, in order to study the effect of such mineral fillers on the properties of the polymers. By way of comparison, HDPE/alumina and HDPE/talc composites were also prepared and studied. From the various results obtained, it can be concluded that:

- Infrared results showed the appearance of new peaks characteristic of the mineral, which proves the presence of alumina and talc in the matrix (HDPE).

- The results of the impact test showed that the impact resistance of the composites decreased with increasing mineral loading due to the formation of particle aggregates in the matrix. The alumina-based composites showed good impact resistance talc-based composites, with the best impact resistance coming from composites containing a 5% mixture of the two fillers (synergism).
- On the other hand, the results of the tensile test showed an increase in stress at break only for composites with 1% talc and 3% mixtures of the two fillers, and a decrease in deformation at break of the composites as a function of the filler ratio, but there was some improvement in modulus of elasticity, implying that the incorporation of fillers imparts a certain stiffness to HDPE.
- HDPE/talc/Alumina co-filler composites combined good properties of both (synergism effect) in comparison with HDPE/talc and HDPE/alumina ones. The excellent dispersion of the filler's particles in HDPE matrix as well as the efficient treatment (dry co-mixing) lead to the enhancement of the mechanical PVC properties.
- Measurement of the melt flow index of various composite formulations showed that the incorporation of a mixture of two fillers resulted in an increase in the melt flow index and hence a reduction in composite viscosity represented by the HDPE/(alumina + talc) composite at 7% of the filler ratio, confirming the results of the mechanical tests that binary-filler composites offer good filler dispersion thanks to the fine particle size obtained by the grinding performed to prepare the mixture of two fillers. Mechanical test results showed that fine particle size has a significant effect on improving mechanical properties.

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