

Study of the physical and thermal properties of composite polymers of high density polyethylene (HDPE) -porcelain obtained by experimental approach.

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ARTICLE INFO

Article History:

Received : 20/08/2018

Accepted : 10/10/2018

Key Words:

Composite polymers;
Tri-layer coating;
Mechanical properties.

ABSTRACT/RESUME

Abstract: Composites of HDPE-porcelain with an average diameter of 51.961 μm have been developed. Experimental results, tensile tests show that the elongation at break and the tensile strength increase in the presence of the porcelain particles in the HDPE more precisely in the material (80wt. % HDPE + 20wt. % porcelain). The DSC results show a melting temperature shift of HDPE to high temperatures in the presence of porcelain particles up to 128.37°C for the 20wt. % porcelain sample. The heat treatment at 1380°C of the porcelain powder which was used as reinforcement in the HDPE matrix, the mullite needles form a network within the vitreous phase, the porosity decreases by the reduction of the black voids observed in the MEB images.

I. Introduction

For many years, there has been a strong interest in the development of polymeric coatings with specific properties. The cost of the raw material, the ease of forming the parts, the weight and the recycling are also parameters to be taken into account. It is of interest to be able to synergistically combine the intrinsic properties of the inorganic component (thermal stability, stiffness, etc.) with those of the polymer (flexibility, dielectric properties, ductility, ease of use, etc) [1]; [2]. Moreover, if the size of the inorganic domains becomes nanometric, the final properties of the organic / inorganic (O / I) hybrid material, which could also be called nanocomposite, are greatly improved due to a greater specific inorganic surface developed to interact with the polymer phase [3, 5]. Steel pipes for the transport and distribution of buried gases may be exposed to ground corrosion attacks that damage grids. Thus, our industrial partner seeks to protect these pipes efficiently by combining the HDPE with the third

layer of protection used externally and which is in contact with the ground with a ceramic material in the form of porcelain powder [6]; [7]. This substance is reliable over time and can withstand external environmental degradation. The choice was made for the tri-layer assembly successively composed of an epoxy layer (Ep), a layer of a mixture of ethylene, butyl acrylate (EBA) and anhydride Maleic acid (MA), and a layer of high density Polyethylene (HDPE). The final properties of nanocomposites depend on several parameters, including the nature, shape, size and rate of the particles inserted and the interface between the polymer and the nanoparticles.

II. Materials and methods

The polymer used, HDPE, is a polymer widely used in industry and also the simplest thermoplastic polymer because of its structure. The choice of porcelain particles is rather for its affordable cost using porcelain waste by recycling. The samples are prepared by the incorporation of porcelain particles

in a polymer matrix. The choice fell on the three-layer assembly each of these layers provides a particular function within the collage:

- The epoxy layer essentially serves as adhesion primer.
- The EBA layer has an adhesive function that allows it to optimize the mutual bonding of materials on both sides of the assembly (Ep and HDPE).
- The layer of HDPE, deposited in thick compared to other materials, provides the function of thick protective coating.



Figure 1. Three-layer coating on a metal substrate

II.1. Sample formulation and development

Mixtures of different proportions (0 to 40wt. %) of porcelain particles are produced in an internal mixer (Brabender, Volume max = 35 cm³). After melting the HDPE at 180°C., in the mixer, the porcelain particles are added in powder form. The torque applied by the mixer rotors as a function of time was recorded. After finishing inserting the particles into the HDPE and arriving at a constant torque, the mixer continues to run for about 10 minutes to ensure homogeneous mixing. The mixtures thus obtained are removed from the mixer, ground and injected in a press (ARBORG, 35 tons, screws with a diameter of 18 mm) in the form of a tensile test specimen. The photo below shows a traction test piece obtained after injection.



Figure 2. Tensile test sample of a HDPE-porcelain mixture after shaping in the injection molding machine

Table 1 shows the different formulations taken into consideration in our study. The aim is to study the influence of porcelain on the behavior of the material.

Table 1. Numbering of samples according to different formulations tested

Raw materials	Percentages by mass (wt. %)			
PEHD (Matrix) (wt. %)	100	90	80	60
Porcelain (reinforcement) (wt. %)	0	10	20	40

II.2. Characterization of samples

The mechanical, thermal and structural properties of the samples are characterized by different techniques: Particle Size Analysis, Differential Scanning Calorimetry (DSC), Scanning Electron Microscopy (MEB). The tensile tests are carried out on a tensile machine (Zwick / Roell Z050 (reference Z2.5KN with a tensile speed of 1mm/min at ambient temperature).

III. Results and discussion

III.1. Particle size analysis results

Material stability, chemical reactivity, opacity, fluidity and strength are affected by the size and characteristics of the particles in them.

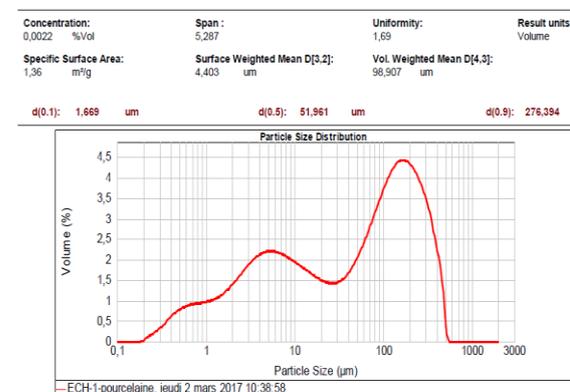


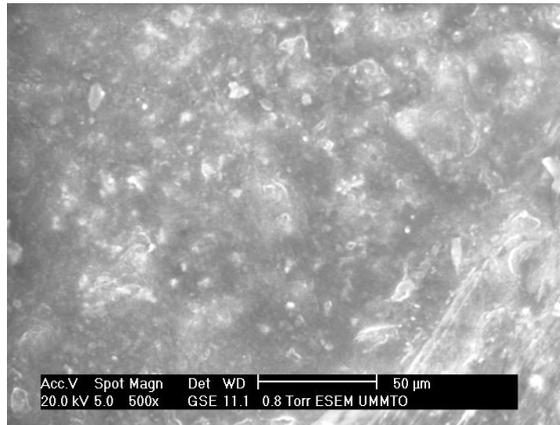
Figure 3. Granulometric distribution of porcelain powder

The porcelain powder used in our case as reinforcement in the HDPE polymer was dried at 105°C, crushed, ground and sieved using a vibration sieve column using different series of sieves. The material was sieved to dryness on a series of Afnor type sieve with dimensions are: 125, 100 and 63µm. All analyzes were carried out with the powdered clay material of particle size less than or equal to 100 µm. Figure 3 shows the particle size curve of the porcelain powder. The analysis shows a bimodal distribution with two maximum points, which are centered at about 5 µm and 170 µm.

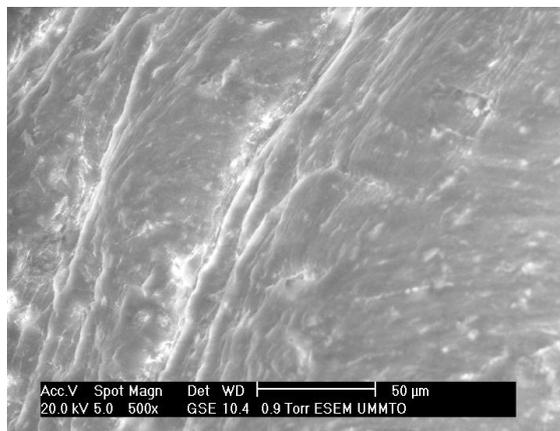
Reducing the size of the porcelain particles can be done in different ways. This powder has larger grain sizes in the practice of ceramics, different types of equipment are used to reduce the particle size depending on the type of material, the initial particle size distribution and the desired final distribution [8]. In the field of ceramics, there is a reduction in size by the grinding process. The particle size of the majority of the volume in general is considered interesting. This size and distribution allows the final powder from the raw material mix to have a greater free surface energy and, consequently, influences the densification of the final product [9].

III.2. Structural properties

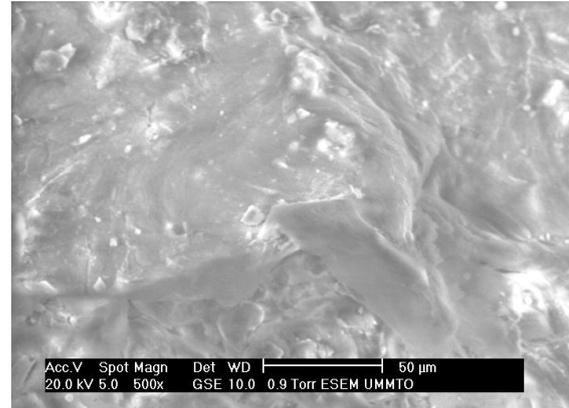
The MEB images below show the microstructure obtained with the various porcelain mixtures (0.10, 20 and 40wt. %) incorporated in the HDPE polymer. The MEB photographs presented were made on samples formed by pressing. The structures of the composites seem to depend on the conditions of the mixing applied and the rate of fillers.



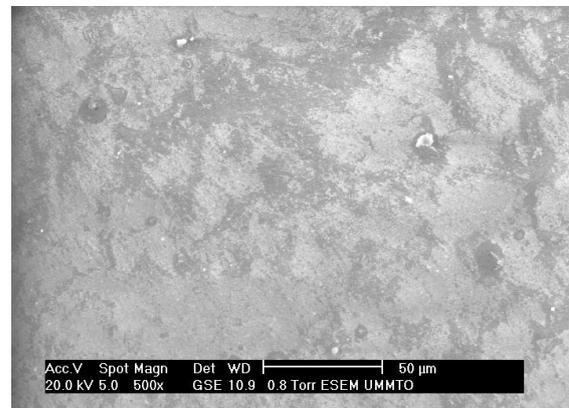
(a)



(b)



(c)



(d)

Figure 4. Microstructure of porcelain bodies: (a) 100% PEHD+0% porcelain; (b) 90% PEHD+10% porcelain; (c) 80% PEHD+ 20% porcelain; (d) 60% PEHD+40% porcelain.

After a heat treatment at 1380°C of the porcelain material which has been used as reinforcement by recycling in a polymer matrix HDPE, the vitreous phase is important, the kaolin sheets which is one of the raw materials of the porcelain material no longer exist and the sizes of the mullite crystals are very large in the 20wt. % porcelain sample [10]. On the other hand, we observe intergranular pores represented in black background on the images 0 and 40wt. % in porcelain. Note the existence of large porcelain particles having geometric shapes and other very small which appear as a powder in the sample 40wt. % porcelain and this because of the phenomenon of flocculation and the poor dispersion of raw materials in the solid. These large particles probably formed during mixing in the extruder. Indeed, the mullite phase decreases and the total porosity of the material also decreases, according to the work of [11], [12] heat treated at 1380°C of the porcelain material, the mullite needles form a network within the vitreous phase, the porosity decreases by the decrease of the black voids observed in the SEM 10 and 20wt. %

porcelain images. These observations are in perfect agreement with the measurements of the density; the increase of the density causes a reduction of the pores.

The figures collect MEB photographs taken at different levels of less rigid porcelain, and rigid different formulations. We carried out a more detailed microscopic study of the effect of the existence of porcelain in the HDPE polymer. Microscopic observations sectional samples of platelets indicate that the material has still suffered internal changes. The platelets have been hardened after adding porcelain

III.3. Thermal properties

The HDPE curves obtained correspond to the variation of heat flux as a function of temperature. The shape of the melting peak depends directly on the temperature keeping time. Table 2 gives the melting temperatures of the systems studied. For the blank sample; we find that the melting temperature is of the order of 103.87°C. They also have an offset of HDPE melting temperature to high temperatures in the presence of porcelain particles, as shown in the thermograms below figure 5. We are interested in this study much more at the melting temperatures of the samples.

Table 2. Melting temperature for the different samples

Melting temperature DSC (°C)	Percentage of porcelain incorporation into PEHD (wt. %)
103.38	0
108.94	10
128.37	20
81.50	40

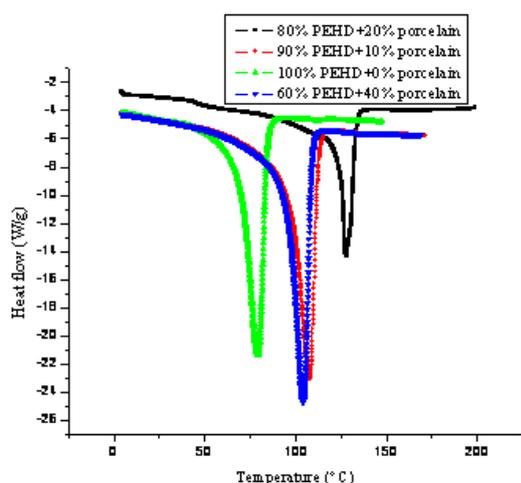


Figure 5. DSC thermograms obtained on composite HDPE-porcelain

The results show that there is indeed an effect of reinforcing the rigidity of the samples in the

presence of porcelain particles according to, indicating the addition of additives such as clays, Al₂O₃, lithium oxide, etc [13], in the polymer materials leads to the increase of the melting temperature (increase of the thermal force), porcelain is classified among refractory materials, during a heat treatment, phyllosilicates lose their structure in sheet, and when the temperatures are sufficiently high (> 1000°C), after cooling, they consist of a mixture of a vitreous phase and crystals, typically mullite and cristobalite [14], [15]. The final powder obtained by mixing the raw materials has a greater free surface energy, the thermal properties improve and, therefore, an increase in the melting temperature and densification of the product 20wt. % porcelain compared to other materials (0, 10 and 40wt. % porcelain).

III.4. Mechanical properties

The tests are carried out at ambient temperature (about 23°C.) and a speed of 100 mm / min. The stress-strain curve, up to the rupture of the specimen, indicates the different values of the mechanical properties of the material such as the Young's modulus, the yield stress, the stress and elongation at break. We performed four test pieces: HDPE (0, 10, 20 and 40wt. % porcelain), the results are shown in figure 6.

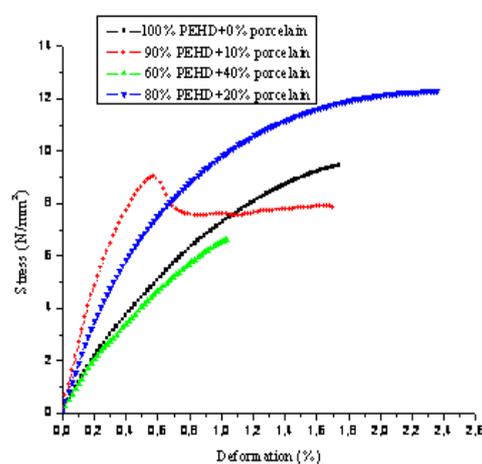


Figure 6. Stress deformation curve for HDPE samples with (0, 10, 20, 40wt. % of porcelain)

The yield stress (tensile strength) and the Young's modulus of the samples obtained at different percentages of porcelain are shown in Table 3. An increase in the threshold stress and Young's modulus is observed for the samples: 10, 20 and 40% of porcelain compared to 0% HDPE of porcelain as shown in table 3. The tensile curves obtained with a speed of 1 mm/min show that the elongation at break increases in the presence of porcelain particles in HDPE more precisely in the

material (80wt. % HDPE + 20wt. % porcelain), although the maximum stress also increases in the elastic domain. When the rate of the porcelain powder is higher, the dispersion becomes difficult and promotes the entanglement of strings that have a character flexible. The addition of the nanocharge of clay seems to induce an antagonistic effect in decreasing the breaking stress of the nanocomposites. This behavior could be generated by the presence of clay aggregates within the matrix, which caused its early break,

Table 3. Values of mechanical quantities: tensile strength and modulus of Young

Quantities mechanical Samples	Tensile strength « R _{tr} » (N/mm ²)	Young's module « E » (N/mm ²)
100wt. % PEHD + 0wt. % porcelain	8.44	9.33
90wt. % PEHD + 10wt. % porcelain	9.07	20.00
80wt. % PEHD + 20wt. % porcelain	12.30	32.00
60wt. % PEHD + 40wt. % porcelain	6.70	10.00

There is thus an effect of reinforcing the rigidity of the composites in the presence of the porcelain particles. Porcelain increases the density and tensile strength that will increase the strength of the coating [14], [16] [17]. Indeed, we notice that the composite module is even higher than the rate of the porcelain powder increases, because by its presence with a rather important rate up to 20wt. %, the matrix induces a certain rigidity which resulted in the decrease of the deformation at the break.

IV. Conclusion

In this work, composite polymers were developed by incorporating porcelain ceramic particles having a mean diameter of 51.961 μm into a HDPE matrix. Subsequently, the influence of the presence of porcelain particle on the mechanical, thermal and structural properties of composite polymers has been studied experimentally. From the stress-strain curve of the virgin material, in particular the stress at the plasticity threshold, the breaking stress and the elongation at break were analyzed as a function of the incorporation of the porcelain powder used as reinforcement in a polymer matrix HDPE. The different values of the mechanical properties of the material such as the Young's modulus, the threshold stress, and the breaking stress increase with respect

to the addition of porcelain. On the other hand, the elongation at break increases sharply which results in a gain of ductility of the material which is related to the phenomenon of rigidity which is accompanied by a strong attachment of the chains. The DSC thermograms allowed access to the melting temperature (top position of the exothermic peak). The results show a sharp increase in melting temperature with the addition rate of porcelain, which comes from the porcelain firing temperature which 1380°C. Incorporation of different amounts of treated porcelain powder at different sintering temperatures has led to the appearance of a mullite phase which is technically important and appears better in the image of the 20wt. % porcelain material. Simulations are in progress for intermediate cases where the interaction between the ceramic particles and HDPE is modified through the HDPE-ceramic interface.

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Please cite this Article as:

Boussak H., Chemani H., Study of the physical and thermal properties of composite polymers of high density polyethylene (HDPE) –porcelain obtained by experimental approach, ***Algerian J. Env. Sc. Technology*, 4:3 (2018) 24-29**