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Perfection of a composite material reinforced by a natural filler modified by a gamma irradiation treatment

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Abstract

Olive residue flour (ORF) were irradiated at doses of 10, 25, 50 and 70 (kGy) and incorporated in Poly (vinyl chloride) (PVC) in different ratios of 10/90 and 20/80 (wt ORF %/PVC wt%) for the preparation of composites. Mechanical behaviors of those composites such as tensile strength and elongation at break have been, assessed. The composite samples prepared with the untreated filler present a decrease of elongation and tensile strength. This decrease is, attributed to the low interfacial adhesion between the filler, which have a strong affinity for water (hydrophilic strength) and the PVC hydrophobic surface. However, the composite filled with ORF treated with gamma irradiation exhibit higher elongation and tensile strength than those of un-irradiated composites. This is, attributed to the decrease in the hydrophilicity of the olive residue flour after gamma-irradiation treatment. The results of the mechanical behavior shown by the structure morphology observation indicate an improvement of the ORF dispersion as seen through Scanning Electron Microscopy. The water-absorption test of different composites shows that irradiated filler composites present less water uptake than those of unirradiated composite.

Keywords: *Poly (vinyl chloride), wood flour, wood plastic composite, γ-radiation*

I. Introduction

Lignocellulosic fibers are, used as fillers or reinforcements in composites with thermoplastic matrices for industrial applications. In fact, hydrophilic character of these lignocellulosic materials disrupts the preparation of such materials based a thermoplastic matrix with a hydrophobic nature giving poor interfacial compatibility, which causes poor mechanical properties.

The industry of olive oil Algerian produces about 200,000 tons of solid waste and the amount tends to increase each year [1]. Chemically, olive residue flour constitutes the cellulose, hemicellulose and lignin and in order to reduce the hydrophilicity of the flour to improve the addition force to the thermoplastic matrix, it is necessary to perform a structural modification of this surface. Several approaches have been studied, Djidjelli, H.et al. have carried out the plasticization of olive residue flour by benzyl chloride to improve some thermal and physicochemical properties of composite PVC/olive residue flour [2]. Boukerrou A. et al. have decreased hydrophobicity of olive residue flour by silanization of the latter [3]. Another technique, which is simpler and less expensive based on the gamma irradiation of composites.

This technique, has been used by several researchers in indeed, Khan M. A. et al. have studied the effect of the filler and the matrix treatment by gamma radiation on the properties of composite [4]. Cemmi A. et al. have succeeded to decrease the wettability of the cellulose by gamma irradiation [5].

In this work, olive residue flour has been irradiated with gamma radiation at doses of 10, 25, 50, 60 and 70 (kGy) and the composites based on polyvinyl chloride (PVC) loaded with 10 and 20% in weight of filler treated and untreated were prepared. The mechanical and morphological characteristics of PVC composites/FGO were studied and the rate of water absorption was determined.

II. Material and methods

II.1. Materials

Olive residue flour was used as filler having a diameter of around 100 (µm), it was collected from Bejaia, Algeria. ORF was dried at 100 °C in a vacuum oven for 24h prior to the preparation of the composites. The matrix in this work was based on PVC type 3000H produced by CIRES (USA). The polymer has the following physical characteristics: Kw value: 72; powder, density: from 0.48 to 0.56 (g.cm-3). The additives used in the preparation of the various formulations are dioctyl phtalate (DOP) as a plastizer with a viscosity range from 75-80 CSt, a molecular weight of 390 (g.mol-1), a boiling temperature of 231°C. Ca/Zn type Lankromark LC486 produced by Akros Chemicals, and stearic acid were also used as stabilizer and lubricant respectively. The mass composition of the different formulation is, summarized in Table 1.

Table 1. Mass composition of formulation for PVC/ Olive residue flour

Compounds	Resin of PVC	Plasticizer	Ca/Zn	Stearic acid
F0 (grams)	100	30	2	0.6
F10(grams)	90	30	2	0.6
F20(grams)	80	30	2	0.6

II.2. Pretreatments of olive residue flour

The Algerian, olive residue flour was, subjected to several pretreatments: washing with hot water to eliminate pulp, drying under ambient conditions for 48 h, and then drying in an oven at 105 °C for 24 hours. The powder was then subjected to crushing and finally sifting to obtain a flour of size smaller than 100 (µm). The flour of olive residue obtained was then washed with acetone in a soxhlet extractor for 24 hours to eliminate any contamination or impurity.

II.3. Irradiation treatment

ORF was treated with γ-rays from a 60Co source in air at room temperature at doses of 10, 25, 50 and 70 kGy using a dose rate of 48 (kGy/h).

II.4. Sample preparation

The mass composition of the pure PVC (F0), PVC/olive residue (F10) and (F20) formulations is represented in Table1. Sheets with a thickness about 1mm are obtained by a calendaring process at 160 °C and a residence time of 15 min.

III.Characterization of composites
III.1. Mechanical properties of composites

Specimens for mechanical testing were prepared in accordance with standard CEI 60811-1-1. , type "H" have the following dimensions Figure1.Tests were carried out using a tensile testing machine of type: BTC-FR 2.5TN.D09, at room temperature. The tensile speed was 100 (mm/min). The reported data are the average of five successful tests.

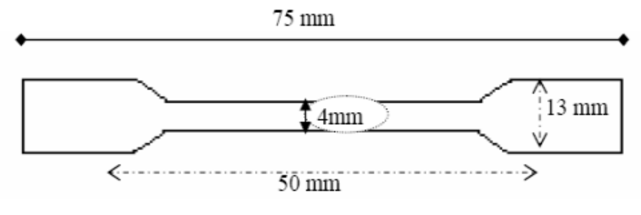

Figure 1. The "H" type specimen

III.2. Surface morphology

Scanning electron microscopy (SEM) was, used to examine qualitatively the dispersion of lignocellulosic fibers in the PVC matrix. The surface of the composites was, examined with a Philips scanning electron microscope.

III.3. Water-absorption tests

Water absorption of composite was, examined by immersion of sheet sample of 1mm thickness in distilled water at room temperature for 24 h, periodically measuring the change in weight of the sample. After weighing, the WA of the samples was, calculated as eq. (1).

$$W_A(\%) = \left(\frac{W_t - W_0}{W_0} \times 100\right) \qquad eq.1$$

Where W_0 and W_t are the weights of the sample before and after immersion in distilled water, respectively

IV.Results and discussion
IV.1. Effect of untreated ORF, on mechanical and morphology properties of composites (Tensile properties)

The values of elongation at break, tensile strength and the Young's modulus for the composites were, measured and the results were, summarized in Table 2. It is, observed that the tensile strength and elongation at break decreases. These results were in accordance with much work [6-8], that has attributed this decrease to the low interfacial adhesion between the filler and the polymer matrix. Wood materials have a strong affinity for water (hydrophilic strength) which creates an incompatibility interface between it and the PVC hydrophobic surface. The deterioration of the elongation at break is also partially due to the incorporation of the rigid flour into the matrix of the PVC, which reduces the mobility of the polymeric chains and facilitates the break of the specimen at low stress.

The incorporation of the ORF into the matrix significantly improves the Young's modulus compared to that for PVC alone. This can be, explained by the fact that the particles of ORF have a rigid character leading to a high resistance to deformation.

Table 2. Stress (σ) and the elongation at break (ε) values for F0, F10 and F20

Compounds	F0	F10	F20
Strength at break σ (MPa)	24.24±3.70	20.60±2.23	16.91±2.77
Elongation at break ε (%)	124.27±2.63	74±1.26	43.82±03.12
Young's modulus E (MPa)	189,41±1,90	192±1,05	193±2,83

Effect of ORF on morphology of composites

The morphological of the fracture surfaces of F0, F10 and F20 composites investigated by SEM are, shown in Figure 2.

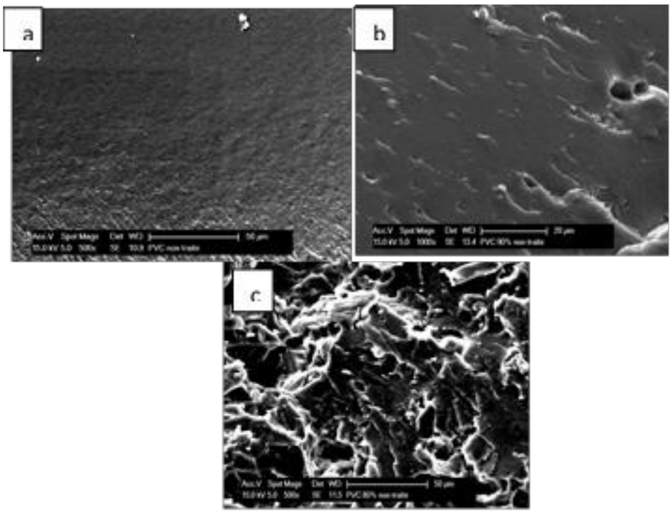

Figure 2. Scanning electron micrograph of: (a) F0, (b) F10, (c) F20

Figure 2a shows the SEM image of freeze-fractured F0 sample. A uniform structure of PVC is, displayed. Observing Figure 2b and Figure 2c corresponding to the morphological of the fracture surfaces of F10 and F20 composite we notice a rough morphology and many gaps and voids. This indicates poor interfacial adhesion that reveals the low affinity between the polymer matrix and the ORF fiber. Poor interfacial adhesion seems to facilitate debonding of the fiber [9].

Effect of gamma radiation on the mechanical properties of ORF in PVC composites
Tensile properties

ORF was, treated with γ-rays at doses of 10, 25, 50, and 70 (kGy) and incorporated in PVC to enhance the composites properties. Figure 3.a and Fig.3.b show the tensile strength and elongation at break as a function of radiation doses of composites.

For F10 composite, the results shown that the strength at break increases at dose of 10 (kGy), after that, it remains almost constant by increasing the radiation dose to 70 (kGy). The elongation at break increases with increasing irradiation dose. For F20 composite, the tensile strength increases by increasing the radiation dose to 70(kGy) and the elongation at break increases from 10 to 50 (kGy) dose and after that the value of elongation at break decreases up to70 (kGy) dose, but it remains higher than that of untreated composite.

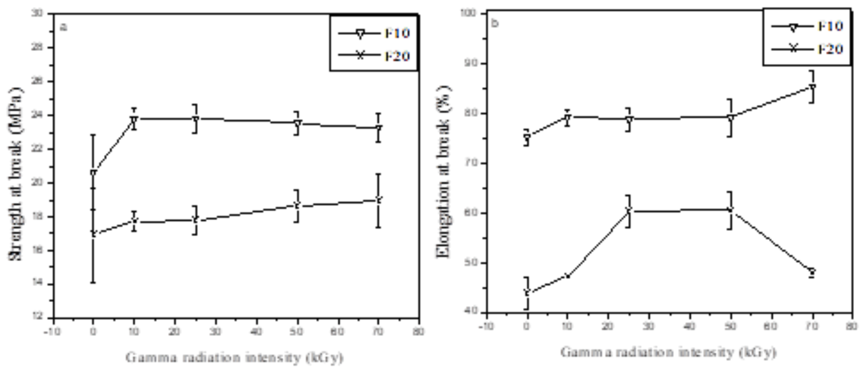

Figure 3. Effect of gamma radiation dose on: (a) tensile strength, (b) elongation at break of composites

The best mechanical properties were obtained for F10 composite irradiated at 70 kGy, we found that the tensile strength = 23.83 MPa, and elongation at break = 85.42%.

The improvements of tensile strength and elongation at break doses may be due to the intercross-linking between the neighboring cellulose molecules that occurs under gamma exposure [10, 11].

IV.2. Morphology

To verify the results of the tensile test, we have analyzed various samples by SEM and the micrographs of the fracture surfaces of F10 before and after treatment at doses of 10 and 70 (kGy) doses are, shown in Figure 4.

Where F10 10kGy: PVC filled with 10% of ORF irradiated at dose of 10 (kGy), and F10 70kGy: PVC filled with 10% of ORF irradiated at dose of 70 (kGy).

Dispersion of the filler in the un-irradiated composites is, presented in Figure 4a. Some cavities are to be, seen where the filler has been pulled-out. The presence of these cavities means that the interfacial bonding between the filler and the matrix polymer is poor and weak.

This is due to the presence of the polar, hydroxyl groups on the surface of lingo-cellulose, the major component of the flour of olive residue, which resulted in agglomeration and the poor adhesion with the non-polar PVC [12-14]. Figure 4b and Fig.4c show the micrographs of the fracture surfaces of irradiated composites at doses of 10 and 70 kGy respectively. Fewer cavities are presents compared the un-irradiated composite surface. This indicates that the interfacial bonding between filler and matrix is generally, improved after gamma irradiation. The best result was, obtained with the composite irradiated with a dose of 70 (KGy).

In addition, cracks between the filler and the matrix are apparent in these figures. The cracks and pores left from pulled-out filler can easily be, seen. Under high, energy radiation ionized and excited molecules are, formed. The polymer may undergo scission to be broken into smaller fragments or else cross-linking, or the molecules may be linked together into large molecule. In fact, the free radicals thus produced may react to change structure of the polymer and alter the physical properties of the materials [10, 11].

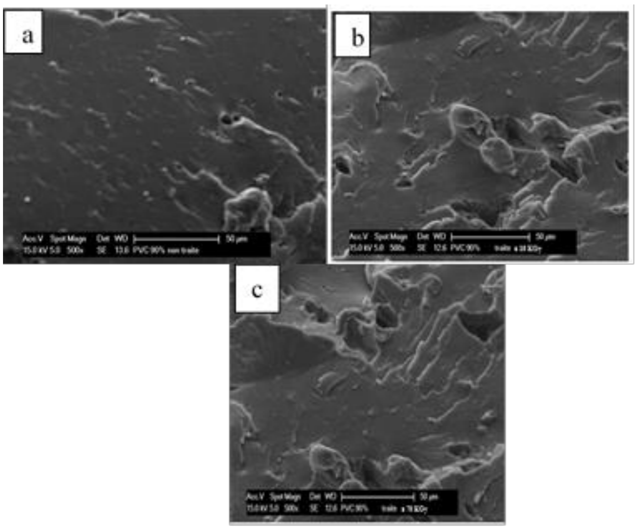

Figure 4. Scanning electron micrograph of a: F10, b: F10 10kGy, c: F10 70kGy

IV.3. Water absorption

The results of water absorption are, shown in Figure 5. It is, recognized that the absorption of water by different composites is largely dependent on the availability of free-OH groups on the surface of reinforcing fibers. Therefore, it is evident in the figure that WA (%) increases with an increase in filler loading. With an increase in filler loading, the number of hydroxyl groups in the composites increases, which consequently increases the WA. The water absorption results demonstrated that the composites filled with gamma irradiated ORF absorbs less amount of water. A lower absorption of water by the composite indicates that more OH groups of cellulose are, blocked from their interaction with PVC matrix. It is, also believed that –OH groups of cellulose molecules in the composites are mutually bonded or cross-linked due the effect of γ-radiation [10].

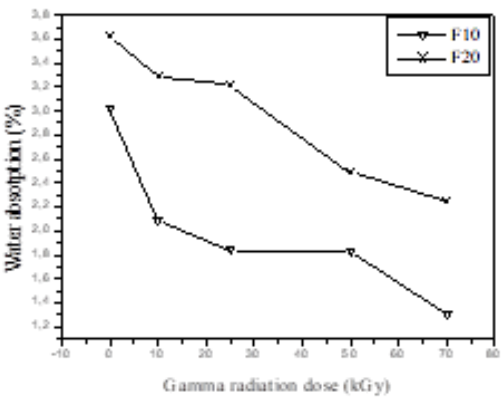

Figure 5. Water absorption of the various composites

V. Conclusion

The purpose of this study was to investigate the effect of ORF and gamma irradiation on mechanical and morphological properties of a composite PVC/olive residue flour. The presence of olive residue flour with hydrophilic nature in the hydrophobic PVC matrix, leads to a material characterized by a heterogeneous dispersion and a weak interfacial adhesion. Gamma radiation treatment at low doses improve the mechanical performance of the ORF reinforced PVC composite, and a lower absorption of water by the composite filled with gamma irradiated ORF was observed. The improvement interaction between flour of olive residue and PVC by radiation treatment is, also verified from the SEM of fracture surface of the composite, by the reduction in the cavities on the surface.

Define abbreviations that are not standard in this field in a footnote to be placed on the first page of the article. Such abbreviations that are unavoidable in the abstract must be defined at their first mention there, as well as in the footnote. Ensure consistency of abbreviations throughout the article.

Conflict of interest. The authors report no conflict of interest.

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