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# Aging of the composite material in different environments

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**Abstract**

***The present work concerns the study of aging a new biodegradable material by mixing the poly lactic acid (PLA), thermoplastic biodegradable, with a low density in the presence of compatibilist PE-GMA in a different environment. The sample formulations LDPE/PLA/PE-GMA (20/80/5; 0/100/0; 100/0/0) are tested in soil and water over time. The aged samples are characterized (by mass loss, pH, and infrared spectroscopy). The results show that the mass loss of blend LDPE/PLA/PE-GMA is 24% after 5 weeks in water at 60°C. The pH value decreases over time. A negligible amount of mass loss in the soil is observed with a pH of 5 and ambient temperature. All results are confirmed by analysis of Infrared Spectroscopy.***

***Keywords:*** *chemical hydrolysis, low-density polyethylene, polylactic acid, soil*

## I. Introduction

Polylactic acid (PLA) is the most important biodegradable polyester with wide applicability in many fields, such as biomedical. PLA has good biocompatibility, and good physical properties such as high strength and high modulus, however, PLA is very brittle [1]. PLA is commonly used as a component of biodegradable materials in pharmaceutical, biomedical, and environmental applications [2]. Density Polyethylene is one of the current thermoplastics that has a higher elongation at break than PLA but is not degradable for this reason, biodegradable polymers like polylactic acid, acorn-based polymer, have been the subject of many studies during the past decade[3].
The increasing manufacturing of green materials in recent years is related to the preoccupation with environmental protection. For that reason, researchers have been oriented toward the elaboration of new environment-friendly biocomposite materials either based on polymers originating from petroleum such as no biodegradable PE, PP, or the plant world (PLA, PBAT…).
Among the biodegradable polymers, polylactic acid (PLA) is increasingly used because it is a biodegradable, biocompatible polymer that can be produced from renewable resources, has low cost, high specific strength, and can substitute polymers derived from petroleum (PP, PE, etc) [4-7]. Protecting the environment means reducing the waste generated by the significant consumption of common thermoplastics such as polyethylene and polypropylene, 40% of which is intended for packaging. To this end, attention has been focused on biodegradable polymers derived from renewable resources such as polylactic acid (PLA). The development of new materials, based on polymer blends, presents an economic and environmental advantage.
An alternative is to minimize the amount of non-degradable polymer by biodegradable substitutes, the goal is to have new materials, both biodegradable and with an affordable price.
As part of this study, we are interested in the behavior of a material PEBD/PLA/PE-GMA, (20/80/5), (100/0/0), and 0/100/0) in soil and water.

## II. Material and methods

We used samples already developed see [8]. The samples are cut into a square shape measuring (1cm x 1cm) numbered and steamed at 105°C until constant weight then they are weighed.
Provide sufficient detail to allow the work to be reproduced. Methods already published should be indicated by a reference: only relevant modifications should be described.
Figure 1:


**Figure 1:** Samples of material

The used soil is taken from the public dump of Mikla town of Tizi Ouzou Algeria. It is dried in the open air for 10 days, crushed, and sifted. We've determined soil composition and characteristics. The working method is illustrated in Figure 2.

### II.1 Hydrolysis

Aging by hydrolysis of samples in distilled water was studied at different temperatures: 45°C and 60°C. The samples of the

LDPE/PLA/PEG-GMA formulations (20/80/5; 0/100/0; 100/0/0) thus prepared are placed in glass jars each filled with 150 ml of distilled water pH= 7.8 well closed at the chosen temperatures, the whole is immersed in the water bath. The duration of aging is spread over 5 and 4 weeks respectively. Three samples were taken each week.

**II.2 Biodegradation of material in soil**

Aging in the soil is studied in different environments. The samples of the LDPE/PLA/PEG-GMA formulations (20/80/5; 0/100/0; 100/0/0) thus prepared are buried in the ground:

- Anaerobic at room temperature,
- Aerobic at room temperature,
- Thermal at a temperature of 60°C.

Watering is done with distilled water with a pH equal to 7.8 and acidic tap water with a pH equal to 5 in the three studied cases above. Aging took place over 2 months:

Three samples were taken every 15 days for each formulation.

Water is added to each container to compensate for the water loss and maintain a fixed humidity level.

The evaluation of biological and hydrothermal aging was determined by the evolution of:

- The loss of mass, as a function of time.
- pH of the medium as a function of time,
- Characterization of spectral FTIR of samples.

**II.3 Mesure de la perte de masse (gravimétrie)**

Après le lavage ; les échantillons, ont été séchés dans une étuve à température de 105°C puis pesés, plusieurs fois afin d'obtenir une masse constante.

La perte de masse a été calculée pour chaque échantillon à l'aide d'une balance analytique de; selon la formule suivante:

$$\textit{Perte de masse (\%) : } \Delta m = \frac{mt-m0}{m0}\,.100\% \qquad (1)$$

*m0*: the initial mass of the sample in grams

*mt*: the mass after aging in grams.

*Δm*: the loss of mass which gives an indication of the rate of degradation.

**II.4 Evolution of pH**

For hydrolysis, the pH of the water is read directly from a pH meter.

**II.4 Spectral and microscopic analysis**

To confirm the chemical degradation we analyzed the samples before and after chemical, and biological degradation.

(Fourier transform infrared spectroscopy) FTIR JASCO model FT/IR 4100 type A OMNIC.

## III. Results and discussion

**III.1 Hydrolysis**

The results of the loss weight of polymers and blend are given in Figure 2,3.

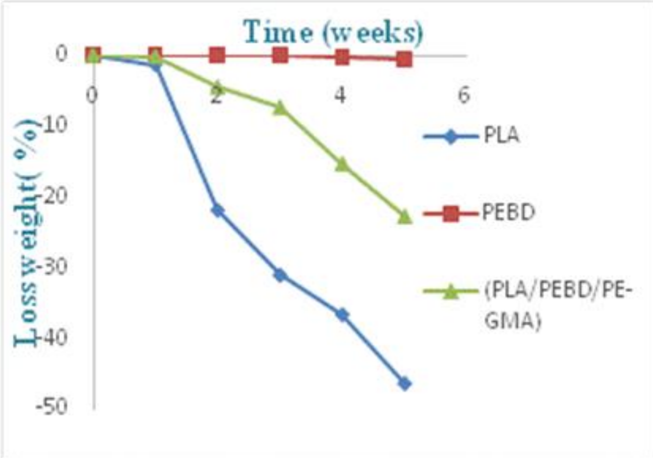


**Figure 2:** Lost weight in water distilled at 60°C

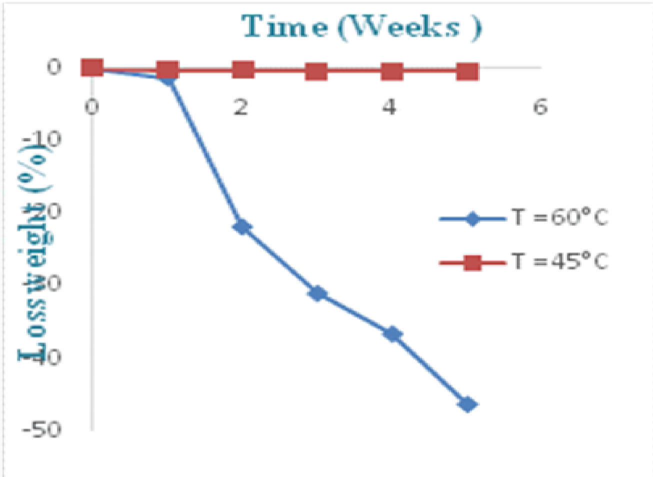


**Figure 3:** Loss weight of PLA in water distilled at 60°C and 45°C

The results obtained show the rate of mass loss at 60 °C is much greater than that at 45 °C. The hydrothermal degradation of the LDPE/PLA/PE-GMA mixture occurred but remained lower than that of PLA.

According to Figures 4,5 and 6, there is a strong decrease in pH at 60°C compared to that recorded at 45°C, moreover, the diminution in pH for PLA alone is faster and with a higher rate than that of the mixture. This decrease is explained by the migration of lactic acid from PLA. The incorporation of PLA into LDPE promotes the degradation of the produced material. PLA can absorb water, which results in the hydrolysis of ester bonds and causes the breakage of macromolecular chains [9] which confirms the loss of mass of the material

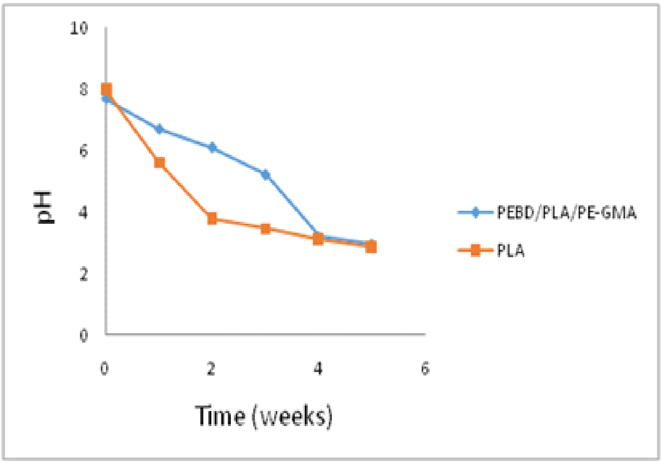


**Figure 4.** Evolution of pH of distilled water as a function of the aging time at temperatures 60°C.

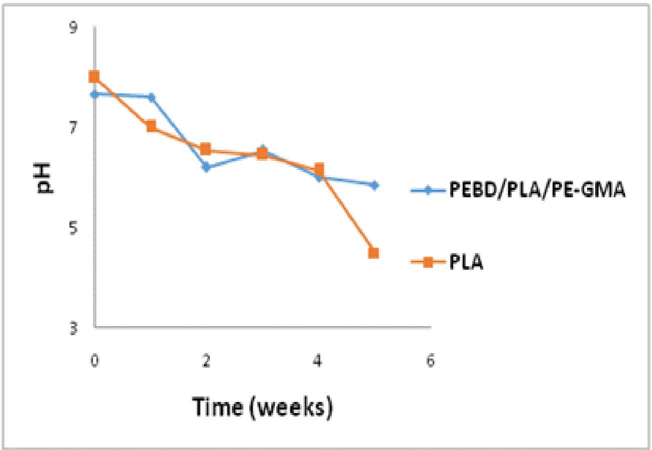


**Figure 5.** Evolution of pH of distilled water as a function of the aging time at temperatures 45°C.

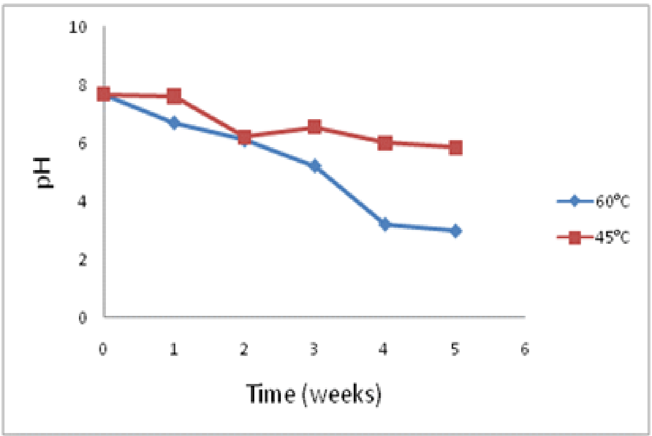


**Figure 6.** Evolution of pH of distilled water as a function of the aging time of the blend LDPE/PLA/PE-GMA (20/80/5) at different temperatures

### III.2 Biodegradation

Soil texture is given in Table 1.

According to the results obtained, the soil has a balanced texture. Sand allows water and air to infiltrate, while clay promotes water retention

**Table1.** Composition du sol

| Analyse granulométrique | | | | |
|---|---|---|---|---|
| Argile (%) | **Limon Fin (%)** | **Limon. Grossier (%)** | **Sable Fin (%)** | **Sable Grossier (%)** |
| *26.15* | ***11.55*** | ***20.75*** | ***18.40*** | ***23.15*** |

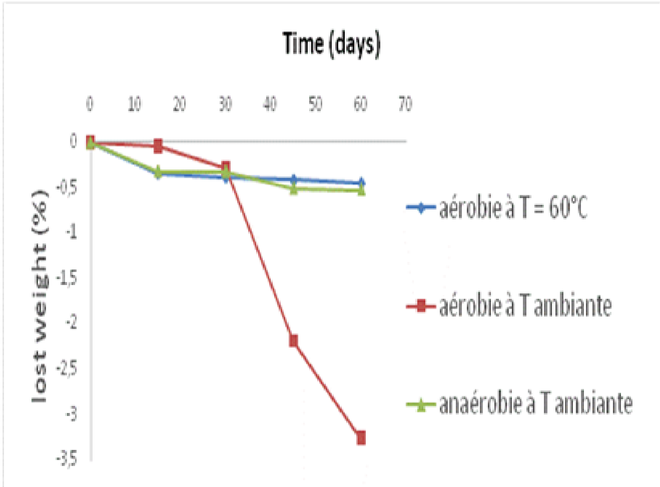


**Figure 7.** Loss of mass LDPE / PLA / PE-GMA (20/80/5) as a function of the aging time in soil, watered with acidulated tap water pH = 5

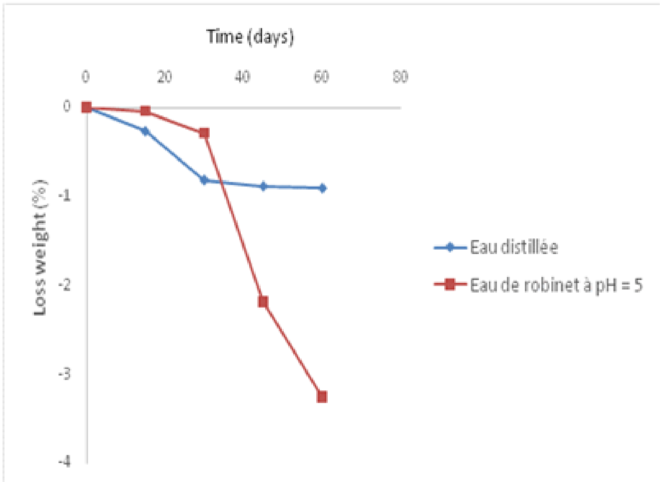


**Figure 8.** Loss weight of LDPE/PLA/PE-GMA aerobic soil at room temperature

The obtained results (Figure 7,8) show that the aerobic environment promotes the degradation of the LDPE/PLA/PE-GMA material (20/80/5). We note a mass loss of 3.5% and 0.4% in aerobiosis and anaerobiosis respectively in the presence of acidulated tap water at a pH of 5. But the degradation remains very low. These mass losses remain insignificant, they agree with the study carried out by Karamanlioglu et al [10] which showed an absence of degradation of PLA in the soil after twelve months at a temperature of 25 °C. Degradation of PLA in soil is difficult at low temperatures [11]. The temperature influences the degradation of PLA (figure 3) these results are consistent with the literature [12]

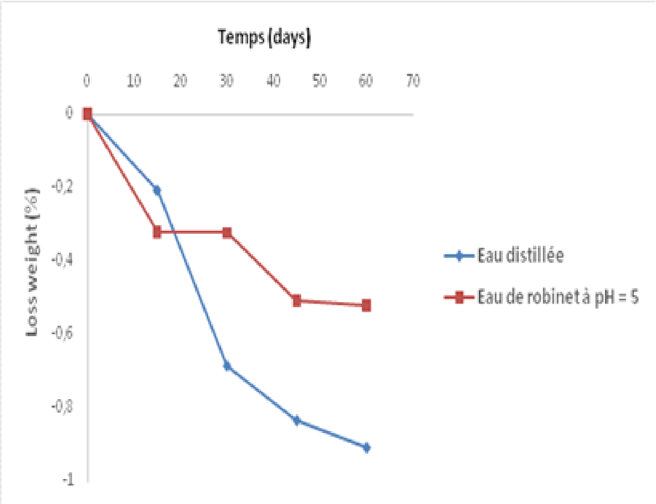


**Figure 9.** Loss weight of LDPE/PLA/PE-GMA anaerobic soil at room Temperature

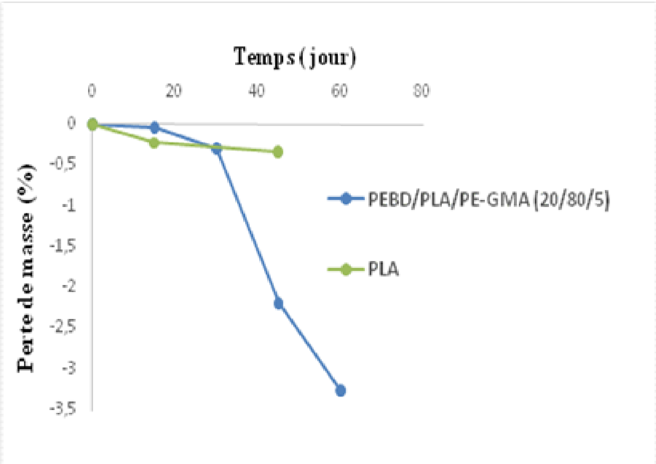


**Figure 10.** Loss weight of LDPE/PLA/PE-GMA and PLA aerobic soil at room T watered with acidulated tap water at pH = 5

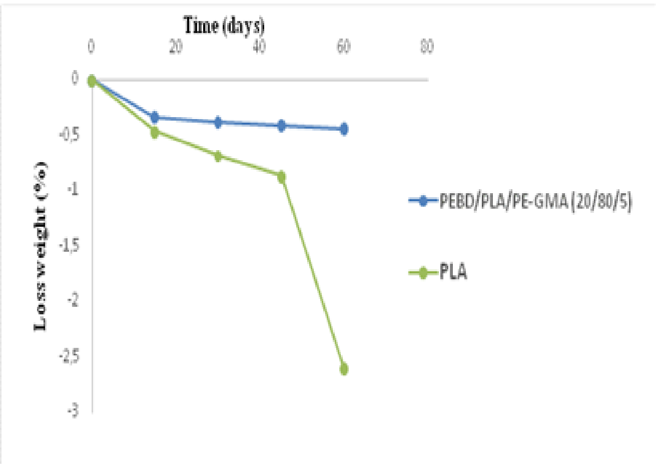


**Figure 11.** Loss weight of LDPE/PLA/PE-GMA and PLA aerobic soil at T 60°C watered with acidulated tap water at pH = 5

The results of the mass loss illustrated by Figure 9,10 and 11 show that the high temperature promotes the degradation of PLA but this loss remains insignificant.

### III.2 FTIR spectra

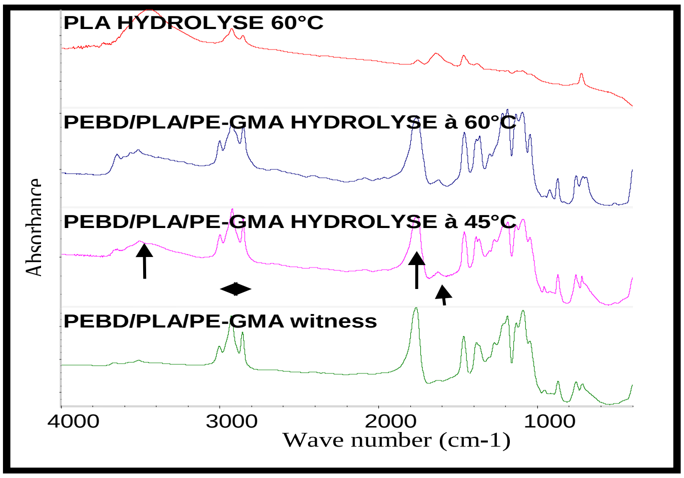


**Figure 12.** FTIR spectra of the different various material hydrolysis

The Figure 12. shows the LDPE / PLA / PE-GMA spectrum (20/80/5) before and after hydrolysis. The results show an increase in the intensity of the symmetrical and asymmetric CH bands (2850-3000 $cm^{-1}$), a broadband intensity increase between 3100-3500 $cm^{-1}$ attributed to the hydrogen bond formation between the hydroxyl group and carboxyl groups, which is confirmed by the displacement of the carbonyl band from 1762 to 1758 $cm^{-1}$. The appearance of a band at 1618 $cm^{-1}$ corresponds to the presence of water enclosed in the cracks thus formed after degradation. The increase of the band to 920 $cm^{-1}$ corresponds to the elongation of the O-H bond of the carboxylic acids.

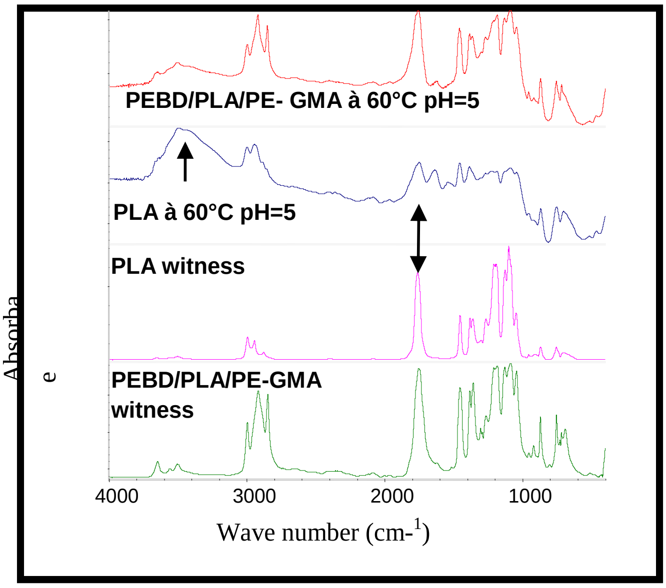


**Figure 13.** FTIR spectra of the various materials buried in the soil aerobic.

The spectra of the mixture before and after aging are in Figure 13. show the appearance of a wide band at 3505 $cm^{-1}$ and that at 1620 $cm^{-1}$ whereas the rest of the bands do not signal any specific change, however, changes in l The intensity of the absorption of the 1757 $cm^{-1}$ and 2850-2997 $cm^{-1}$ bands thus confirming the low mass loss obtained.

## IV. Conclusions

The hydrolysis of the LDPE/PLA/PE-GMA and PLA mixture takes place at a temperature above its glass transition temperature. Therefore, the pH value decreases greater at 60 ° C. than at 45 ° C.

Degradation of PLA in soil is difficult at low. The temperature of 60 ° C. does not contribute to the degradation in the soil of the LDPE / PLA / PE-GMA material (20/80/5) in both cases: watered by distilled water or acid tap water at pH = 5. These mass losses remain insignificant.

The aerobic environment at room temperature is more favorable but the biodegradation process remains weak.

The results show that the PLA incorporation in PEBD promotes the hydrothermal of material.

The FTIR analysis shows the disappearance and appearance of bands which confirm the mass loss. The structural changes were investigated by FTIR spectroscopy. The material thus produced is much more marked by hydrolysis.

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