**The answers for the reviewer A:**

1. **Page 3, line 90: unclear more Detaille how CEC of montmorillonite was determined using ammonium-ions.**

Yes, I agree.

The following details are now added in the section “*Bentonite Treatments“*:

The cation exchange capacity (CEC) of the sodium montmorillonite (MMT-Na+) was determined by saturated this MMT-Na+ by NH4+ using ammonium acetate (NH4OAc) 1 mol/L at pH=7. Excess salt was removed by washing with an ethanol-water mixture. Next, the NH4+ was displaced by K+, which was applied as a 10% KCl solution at pH=2.5. The amount of NH4+, representing the CEC, it is measured by distillation and titration (Leij and Dane, 1989).

We pipette 10 ml of aqueous phase recovered at the end of NH4+ displacement by K+, and put it into the distillation unit. Distilled is put into a conical flask containing 10 ml of 30% NaOH solution in 2% boric acid with an indicator. Keep distilling until the receiving solution raise to 50 ml in volume. This last (distillate) is tittered with 0.1 M HCl until the color just about to change from green to light red.35

The CEC of the sample is obtained from the following equation (Eq 1):

CEC= = mmol/ g (1)

Where;

CEC = the cation exchange capacity of the sample expressed with mmol/g of MMT-Na+;

= the concentration of Hydrochloric acid with moles/l;

= the volume of Hydrochloric acid with l.

We obtained a CEC value of 1 mmol/g. The grafting of the ODA into MMT is effected using a 2-CEC ratio (ODA/MMT) as follows: 10-2 moles of the ODA were introduced in 1 L of 0.01N HCl at 80°C under mechanical stirring. After three hours, 5 g of MMT-Na+ was added.

The previous Bentonite Treatments part was:

*Bentonite Treatments*

The purification of the BNT consists in the elimination of all the impurities like quartz and calcite, then extract the MMT, by applying various physicochemical treatments.

In order to extract MMT and remove the quartz particles present in the BNT, we sediment the suspension (BNT/water, 10% Wt) during 70 hours; the aqueous solution was centrifuged in order to recover the MMT. The recovered MMT was treated with 0.1M HCl (10 g of MMT immersed in 250 ml of 0.1M HCl) at room temperature for 4 hours. Followed by H2O2 (10V) treatment; when 10 g of MMT dispersed in 500 ml of H2O2 for 24 hours tracked by heating at 70°C for 30 min.

In order to obtain uniform MMT surface, a cation exchange is necessary.  In our case, we proceed as follows: 10 g of the purified MMT are dispersed in 500 ml of NaCl solution 0.5 N at 70°C for 4 hours. The operation is repeated three times to reach the saturation of the MMT by the sodium cations and the obtention of sodium montmorillonite MMT-Na+. Finally, in the aim to modify the hydrophilic nature of the MMT-Na+, alkylammonium ions are used ODA.

The cation exchange capacity (CEC) of the MMT-Na+ was determined by saturated this MMT-Na+ by NH4+ using ammonium acetate (NH4OAc) 1 mol/L at pH=7. Excess salt was removed by washing with an ethanol-water mixture. Next, the NH4+ was displaced by K+, which was applied as a 10% KCl solution at pH=2.5. The amount of NH4+, representing the CEC 34. We obtained a value of 1 mmol/g CEC.

The grafting of the ODA into MMT is effected using a 2-CEC ration (ODA/MMT) as follows: 10-2 moles of the ODA were introduced in 1 L of 0.01N HCl at 80°C under mechanical stirring. After three hours, 5 g of MMT-Na+ was added. After 20 hours of cation exchange, the suspension was filtered and washed with distilled water at 80°C. In order to eliminate the physisorbed alkylammonium, the MMT was washed after removal of the chloride ions with a water/ethanol mixture, previously heated to 70°C. When the suspension is completely filtered, dried at 85°C, crushed and sifted it is stored in a desiccator.

The corrected Bentonite Treatments part is after:

*Bentonite Treatments*

The purification of the BNT consists in the elimination of all the impurities like quartz and calcite, then extract the MMT, by applying various physicochemical treatments.

In order to extract MMT and remove the quartz particles present in the BNT, we sediment the suspension (BNT/water, 10% Wt) during 70 hours; the aqueous solution was centrifuged in order to recover the MMT. The recovered MMT was treated with 0.1M HCl (10 g of MMT immersed in 250 ml of 0.1M HCl) at room temperature for 4 hours. Followed by H2O2 (10V) treatment; when 10 g of MMT dispersed in 500 ml of H2O2 for 24 hours tracked by heating at 70°C for 30 min.

In order to obtain uniform MMT surface, a cation exchange is necessary.  In our case, we proceed as follows: 10 g of the purified MMT are dispersed in 500 ml of NaCl solution 0.5 N at 70°C for 4 hours. The operation is repeated three times to reach the saturation of the MMT by the sodium cations and the obtention of sodium montmorillonite MMT-Na+. Finally, in the aim to modify the hydrophilic nature of the MMT-Na+, alkylammonium ions are used ODA.

The cation exchange capacity (CEC) of the MMT-Na+ was determined by saturated this MMT-Na+ by NH4+ using ammonium acetate (NH4OAc) 1 mol/L at pH=7. Excess salt was removed by washing with an ethanol-water mixture. Next, the NH4+ was displaced by K+, which was applied as a 10% KCl solution at pH=2.5 34.

We pipette 10 ml of aqueous phase recovered at the end of NH4+ displacement by K+, and put it into the distillation unit. Distilled is put into a conical flask containing 10 ml of 30% NaOH solution in 2% boric acid with an indicator. Keep distilling until the receiving solution raise to 50 ml in volume.This last (distillate) is tittered with 0.1 M HCl until the color just about to change from green to light red.35

The CEC of the sample is obtained from the following equation (Eq 1):

CEC= = mmol/ g (1)

Where;

CEC = the cation exchange capacity of the sample expressed with mmol/g of MMT-Na+;

= the concentration of Hydrochloric acid with moles/l;

= the volume of Hydrochloric acid with l.

We obtained a CEC value of 1 mmol/g. The grafting of the ODA into MMT is effected using a 2-CEC ratio (ODA/MMT) as follows: 10-2 moles of the ODA were introduced in 1 L of 0.01N HCl at 80°C under mechanical stirring. After three hours, 5 g of MMT-Na+ was added. After 20 hours of cation exchange, the suspension was filtered and washed with distilled water at 80°C. In order to eliminate the physisorbed alkylammonium ions, the MMT was washed after removal of the chloride ions with previously heated water/ethanol mixture (≈70°C). When the suspension is completely filtered, dried at 85 °C, crushed and sifted it is stored in a desiccator.

1. **Page 3, ligne 71, Added the Evaluation of flame retardancy in Characterization  
   method and give it in the Experimental part.**

Yes , I agree.

These details are now added into “Characterization method” section:

The test of the flammability of the polyurethane foam sample is evaluated according to the standard UL 94, used by American laboratories. This evaluation is done according to the UL 94 HB (*Horizontal Burning*) and to UL 94V (*Vertical Burning).* We used this test for the prepared polyurethane foam samples that have 127×12.7×12.7 mm as dimensions. Marks are indicated on the sample at 25.4 (1 ") mm and 101.6 mm (3").

The previous Characterization method was:

Fourier transform infrared (FTIR) spectra were recorded between 400 and 4000 cm-1 from KBr pellets on an infrared Fourier transform spectrometer (Shimadzu type 8400 S). X-Ray powder diffraction patterns were recorded on a PANalytical X'Pert PRO diffractometer fitted with CuK*α* radiation (*λ* = 1.5404 nm) at 40 kV and 40 mA in the 4–60° 2*θ* region. The surface morphologies of samples were investigated via scanning electron microscopy (SEM) (FEI pattern Quanta 600 at 15 kV), in order to observe the evolution of the structure of the cell and their sizes after incorporation OMMT. For the preparation of the (SEM) samples, the sample was cut and coated with a thin gold layer, using a Cressington 108 auto sputter coater sputter, with a current intensity of 40 mA, for 30 seconds. The tensile testing of the rigid polyurethane foams was carried out on an LLOYD instrument TM, with a load cell of 20 KN. The crosshead speed was set to 5 mm/min. Samples were cut to Polyhedron dimensions 127 × 10 × 9 mm, the size between the detectors is 50 mm.

The corrected Material and Characterization method is after:

Fourier transform infrared (FTIR) spectra were recorded between 400 and 4000 cm-1 from KBr pellets on an infrared Fourier transform spectrometer (Shimadzu type 8400 S). X-Ray powder diffraction patterns were recorded on a PANalytical X'Pert PRO diffractometer fitted with CuK*α* radiation (*λ* = 1.5404 nm) at 40 kV and 40 mA in the 4–60° 2*θ* region. The surface morphologies of samples were investigated via scanning electron microscopy (SEM) (FEI pattern Quanta 600 at 15 kV), in order to observe the evolution of the structure of the cell and their sizes after incorporation OMMT. For the preparation of the (SEM) samples, the sample was cut and coated with a thin gold layer, using a Cressington 108 auto sputter coater sputter, with a current intensity of 40 mA, for 30 seconds. The tensile testing of the rigid polyurethane foams was carried out on an LLOYD instrument TM, with a load cell of 20 KN. The crosshead speed was set to 5 mm/min. Samples were cut to Polyhedron dimensions 127 × 10 × 9 mm, the size between the detectors is 50 mm.

The test of the flammability of the polyurethane foam sample is evaluated according to the standard UL 94, used by American laboratories. This evaluation is done according to the UL 94 HB (*Horizontal Burning*) and to UL 94V (*Vertical Burning).* We used this test for the prepared polyurethane foam samples that have 127×12.7×12.7 mm as dimensions. Marks are indicated on the sample at 25.4 (1 ") mm and 101.6 mm (3").

1. **Page 3, ligne 71, The description of density determination for prepared foams**

Yes , I agree

These lines was added in the section of “characterization method”;

“The bulk density of the samples foams was estimated by directly measuring the weight and volume of the polyhedron foams”.

The previous Characterization method was:

Characterization method

Fourier transform infrared (FTIR) spectra were recorded between 400 and 4000 cm-1 from KBr pellets on an infrared Fourier transform spectrometer (Shimadzu type 8400 S). X-Ray powder diffraction patterns were recorded on a PANalytical X'Pert PRO diffractometer fitted with CuK*α* radiation (*λ* = 1.5404 nm) at 40 kV and 40 mA in the 4–60° 2*θ* region. The surface morphologies of samples were investigated via scanning electron microscopy (SEM) (FEI pattern Quanta 600 at 15 kV), in order to observe the evolution of the structure of the cell and their sizes after incorporation OMMT. For the preparation of the (SEM) samples, the sample was cut and coated with a thin gold layer, using a Cressington 108 auto sputter coater sputter, with a current intensity of 40 mA, for 30 seconds. The tensile testing of the rigid polyurethane foams was carried out on an LLOYD instrument TM, with a load cell of 20 KN. The crosshead speed was set to 5 mm/min. Samples were cut to Polyhedron dimensions 127 × 10 × 9 mm, the size between the detectors is 50 mm.

The test of the flammability of the polyurethane foam sample is evaluated according to the standard UL 94, used by American laboratories. This evaluation is done according to the UL 94 HB (*Horizontal Burning*) and to UL 94V (*Vertical Burning).* We used this test for the prepared polyurethane foam samples that have 127×12.7×12.7 mm as dimensions. Marks are indicated on the sample at 25.4 (1 ") mm and 101.6 mm (3").

The corrected Characterization method is after:

Characterization method

Fourier transform infrared (FTIR) spectra were recorded between 400 and 4000 cm-1 from KBr pellets on an infrared Fourier transform spectrometer (Shimadzu type 8400 S). X-Ray powder diffraction patterns were recorded on a PANalytical X'Pert PRO diffractometer fitted with CuK*α* radiation (*λ* = 1.5404 nm) at 40 kV and 40 mA in the 4–60° 2θregion. The surface morphologies of samples were investigated via scanning electron microscopy (SEM) (FEI pattern Quanta 600 at 15 kV), in order to observe the evolution of the structure of the cell and their sizes after incorporation OMMT. For the preparation of the (SEM) samples, the sample was cut and coated with a thin gold layer, using a Cressington 108 auto sputter coater sputter, with a current intensity of 40 mA, for 30 seconds. The tensile testing of the rigid polyurethane foams was carried out on an LLOYD instrument TM, with a load cell of 20 KN. The crosshead speed was set to 5 mm/min. Samples were cut to polyhedron dimensions 127 × 10 × 9 mm, the size between the detectors is 50 mm. The bulk density of the samples foams was estimated by directly measuring the weight and volume of the polyhedron foams.

The test of the flammability of the polyurethane foam sample is evaluated according to the standard UL 94, used by American laboratories. This evaluation is done according to the UL 94 HB (*Horizontal Burning*) and to UL 94V (*Vertical Burning).* We used this test for the prepared polyurethane foam samples that have 127×12.7×12.7 mm of dimensions. Marks are indicated on the sample at 25.4 (1 ") mm and 101.6 mm (3").

1. **Page 7, lines 164-166: X-ray diffraction pattern of the fondry BNT, MMT-Na+ and OMMT.**

Yes, I agree,

The d001-value is calculated from the position of (001) reflection using Braggs equation, and this peak was not determinated for OMMT, it is shifted to 2θ values lower than 4°. The distance calculated using braggs equation represente basal spacing not interlayer distance.

The previous text was:

The increase in the interlayer distance between layers of montmorillonite, after the organophilic modification. The distance between layers of the montmorillonite shifted until reaching 29.3 Å, whereas this distance was only 10 Å for the untreated fondry BNT.

This distance between clay layers is calculated by using the Bragg equation as shown below (Eq 3):

(3)

The foundry BNT shows the characteristic peak at 2 = 8.91, corresponding to the interlayer distance (d001-spacing) of 9.91 Å. For the MMT-Na, the peak of the montmorillonite is found at 2 = 6.27, corresponding to the interlayer distance (d001-spacing) of 14.06 Å. This increase in d001-spacing of MMT-Na+ was previously found by D. E. Kherroub and al 41. It is likely be attributed to the aggressive ion exchange of the calcium cations (Ca2+ = 100 pm) by sodium cations (Na+= 102 pm), which is bigger than calcium cations. Almost similar results were found in the work of El Achaby et al, reported on the change of MMT-Na by 2-(1-hydroxyethyl)-1,3-dihexadecyl-benzimidazolium bromide (Bz), the interlayer distances have increased from 14.06 Å to 29 Å.38

The corrected text is after:

The increase in the basal spacing d001, after the organophilic modification. This distance is greater than d001-29.3 Å for the OMMT, where this distance was only d001-10 Å for the untreated foundry BNT.

The d001-value is calculated from the position of (001) reflection using Braggs equation as shown below (Eq 4):

(4)

The foundry BNT shows the characteristic peak at 2θ = 8.91°, corresponding to d001-spacing of 9.91 Å. For the MMT-Na+, the peak of the montmorillonite is found at 2θ = 6.27°, corresponding to d001-spacing of 14.06 Å. This increase in d001-spacing of MMT-Na+ also reported by Kherroub and al 42, can be attributed to the aggressive ion exchange of the calcium cations (Ca2+ = 100 pm) by sodium cations (Na+= 102 pm). Similar results were found in the work of El Achaby et al 39, reported on the change of MMT-Na+ by 2-(1-hydroxyethyl)-1,3-dihexadecyl-benzimidazolium bromide (Bz), where the distance increased from 14.06 Å to 29 Å.

1. **d001-spacing of organo-montmorillonite**

Yes, the (001) reflection corresponding to d001-spacing superior than 2.93 nm is not visible; this peak is shifted to lower 2θ values than 4°. This correction is performed in the answer for the question 4.

1. **Insert the answer to question 8 from the previous revision in the  
   manuscript.**

Yes, I agree, a sentence was added in the corrected abstract.

We used only 2 %  of OMMT to prepare PUR4 composite because the optimal proprieties of PUR 3 composites was obtained with 2% of OMMT as first step for formulation optimization, this can be due to the best distribution of the OMMT.

The previous text was:

In this work, we propose new method of elaboration nanostructured foam polyurethane/ Organo-modified Montmorillonite (PUR/ OMMT) elaborated by in situ polymerization using a twin-screw extruder in contrarotative mode combined with reaction injection moulding (RIM) process with an appropriate montage. The blended polyols, copolymer polyol (CPP) were included between the OMMT layers via the twin-screw extruder. The formulation of the PUR and the inter-foliar distance in the montmorillonite (MMT) both have been optimized. The effect of some parameters such as MMT content and catalyst was also undertaken. The synthesized materials (OMMT, PUR and PUR/ OMMT) were characterized by different methods; Fourier transform infrared spectroscopy FTIR spectroscopy, X-ray diffraction (XRD) and Scanning electron microscope (SEM) for only PUR 3/ OMMT. The evaluation tests like flammability according to UL 94 and the tensile test were carried out. In view of the obtained results, the PUR 4 (r == 1.15 with 0.97% of diamino-1,2 propane) foam admits mechanical and flame-retardant properties better than the PUR 3 (r == 1.15) foam. However, the PUR 4 + 2% OMMT formula exhibits the most delayed flame diffusion and pronounced stiffness.

The corrected text is after:

In this work, we propose new method of elaboration nanostructured foam polyurethane/ Organo-modified Montmorillonite (PUR/ OMMT) elaborated by in situ polymerization using a twin-screw extruder in contrarotative mode combined with reaction injection moulding (RIM) process with an appropriate montage. The blended polyols, copolymer polyol (CPP) were included between the OMMT layers via the twin-screw extruder. The formulation of the PUR and the inter-foliar distance in the montmorillonite (MMT) both have been optimized. The effect of some parameters such as MMT content and catalyst was also undertaken. The synthesized materials (OMMT, PUR and PUR/ OMMT) were characterized by different methods; Fourier transform infrared spectroscopy FTIR spectroscopy, X-ray diffraction (XRD) and Scanning electron microscope (SEM) for only PUR 3/ OMMT. The evaluation tests like flammability according to UL 94 and the tensile test were carried out. In view of the obtained results, Based on the results obtained, composite foams of PUR 3+ OMMT, PUR 3+ 2% OMMT foam shows optimal results, for that only 2% OMMT was used for PUR 4 (r == 1.15 with 0.97% of diamino-1,2 propane). the PUR 4 foam admits mechanical and flame-retardant properties better than the PUR 3 (r == 1.15) foam. However, the PUR 4 + 2% OMMT formula exhibits the most delayed flame diffusion and pronounced stiffness.

1. **Page , ligne , Added the tensile strength at break (MPa) and elongation at break (%) in Table IV and presente the stress-strain curves for composite and explaine how the stiffness is determinated.**

Yes, I agree, in the corrected tensile propriety we added the tensile strength and elongation at break for all composites in table IV. The fig.8 and fig.9 was added for the variation for the stress-strain curves and strength-elongation curves.

We mean by stiffness the rigidity for this material for this we remplaced the stiffness by rigidity. Foam rigidity was determined from the gradient of the initial linear portion of the strength- elongation curve, it is calculated using the equation below.

R=

R: rigidity for this materiel (N/m)

: The difference strength in linear part of cuve.

allongement in the linear part.

The previous part of “Tensile properties” was:

In order to estimate the tensile properties of rigid foams polyurethanes, we carried out this test on five patterns of PUR(s), the dimensions of each one are 127 × 10 × 9 mm for each formulation, and the size between the detectors is 50 mm. with crosshead speed was set to 5 mm/min. The apparatus records the strain stress according to the elongation. At the end of the test, the device also records the Young's modulus, stiffness, breaking strength and maximum force used by the apparatus. The results obtained in this test for the various samples studied of PUR(s) foams are listed in Table IV.

TABLE IV. Results of the tensile test.

|  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- |
| **Formule** | **Density**  **(kg/dm3)** | **Young's modulus**  **(MPa)** | **Stiffness**  **(N/m)** | **Strength at break (N)** | **Maximale**  **Strength (N)** |
| **PUR 3** | 0.4000 ± 0.0091 | 226.6 ± 23.1 | 407860 ± 4300 | 187 ± 2 | 191,1 ± 3 |
| **PUR 3 + 2% OMMT** | 0.1700 ± 0.0031 | 674.1 ± 3.3 | 2224715 ± 5432 | 40 ± 8 | 65 ± 5 |
| **PUR 3 + 3% OMMT** | 0.2070 ± 0.0054 | 122.1 ± 91.1 | 163660 ± 4321 | 51 ± 7 | 65 ± 7 |
| **PUR 3 + 5% OMMT** | 0.2610 ± 0.0092 | 170.4 ± 6.4 | 299833 ± 3329 | 63 ± 3 | 72 ± 4 |
| **PUR 3 + 7% OMMT** | 0.2170 ± 0.0045 | 148.0 ± 5.2 | 275746 ± 2341 | 43 ± 4 | 47 ± 3 |
| **PUR 3 + 10% OMMT** | 0.2550 ± 0.0041 | 379.8 ± 3.3 | 620325 ± 985 | 48 ± 6 | 66 ± 5 |
| **PUR 4** | 0.5340 ± 0.0033 | 458.9 ± 1.2 | 807595 ± 929 | 315 ± 3 | 433 ± 9 |
| **PUR 4 + 2% OMMT** | 0.2080 ± 0.0021 | 345.4 ± 5.1 | 592830 ± 3351 | 122 ± 5 | 100 ± 8 |

There is a proportional relationship between the added clay content and the density from the calculated apparent densities of the prepared materials. This density decreases sharply when we added OMMT and increases according to the rate of OMMT added. It decreases by 57, 48, 34, 45 and 36% for 2, 3, 5, 7 and 10% of OMMT respectively. This decrease in density is due to the pattern cells formed by the foam, it is noted that the foam with 2% of montmorillonite is formed with open cells that contain other cells.

Moreover, we have an increase in stiffness with 445% and young’s modulus 197% for only the composite PUR 3 charged with 2% by weight of OMMT, despite the opening cells , and just an increase in young’s modulus when we added 10% for OMMT and an increase in stiffness with 52% for the composite charged with 7%. This increase is probably due to closed cells for this composite. The other composites PUR 3 foam display a decrease in the young’s modulus and stiffness proprieties. The mechanical properties is interfered by the presence of clay nanoparticules by the perturbation of the structure of the polyurethane foam leading to reduced mechanical properties and an increase when we have a homogeneous hydrogen bond.

It is important to report that the stiffness and young modules for PUR 4 is twice as big as PUR 3, it is explained by the increase the presence to the hydrogen bond, which increase with the presence of the diamino-1,2propane. The proprieties of PUR 4 decrease when we added 2% of the OMMT. The density, the stiffness and the Young’s modulus for PUR 4 decrease by 61, 26 and 24% respectively. This decrease is probably due to perturbation of the hydrogen bond formed between the PUR, the diamino-1,2 propane and OMMT. However, PUR 4 formula has optimum mechanical properties, compared to the other samples.

The corrected part of “Tensile properties” is after:

In order to estimate the tensile properties of rigid foams polyurethanes, five patterns of these lasts were tested. The apparatus records the stress according to the strain. At the end of the test, the device also records the Young's modulus, rigidity of the material, tensile strength and the elongation at break, strength at breaking and maximum strength used by the apparatus. The results obtained in this test for the various samples studied of PUR(s) foams are listed in Table IV, the stress-strain and the strength-elongation curves for composites is shown in Fig. 8 and Fig. 9 respectively.

TABLE IV. Results of the tensile test.

|  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- |
| **Formule** | **Apparent Density**  **(kg/dm3)** | **Young's modulus (MPa)** | **Rigidity**  **(KN/m)** | **Tensile strength at break (MPa)** | **Elongation at break (%)** | **Strength at break (N)** | **Maximale**  **Strength (N)** |
| **PUR 3** | 0.4000 ± 0.0091 | 226.6 ± 23.1 | 407.86 ± 4.30 | 0.525 ± 0.005 | 3.03 ± 0.10 | 212 ± 2 | 224,1 ± 3 |
| **PUR 3 + 2% OMMT** | 0.1700 ± 0.0031 | 674.1 ± 3.3 | 2224.72 ± 5.43 | 0.531 ± 0.020 | 1.15 ± 0.12 | 20 ± 8 | 24 ± 5 |
| **PUR 3 + 3% OMMT** | 0.2070 ± 0.0054 | 122.1 ± 91.1 | 163.66 ± 4.32 | 0.210 ± 0.018 | 1.03 ± 0.09 | 20 ± 7 | 42 ± 7 |
| **PUR 3 + 5% OMMT** | 0.2610 ± 0.0092 | 162.4 ± 6.4 | 299.83 ± 3.33 | 0.188 ± 0.007 | 4.96 ± 0.17 | 17 ± 3 | 27 ± 4 |
| **PUR 3 + 7% OMMT** | 0.2170 ± 0.0045 | 148.0 ± 5.2 | 275.75 ± 2.34 | 0.385 ± 0.010 | 1.63 ± 0.12 | 36 ± 4 | 46 ± 3 |
| **PUR 3 + 10% OMMT** | 0.2550 ± 0.0041 | 379.8 ± 3.3 | 620.33 ± 1.90 | 0.625 ± 0.015 | 1.95 ± 0.07 | 26 ± 6 | 29 ± 5 |
| **PUR 4** | 0.5340 ± 0.0033 | 458.9 ± 1.2 | 807.60 ± 1.93 | 2.546 ± 0.007 | 4.63 ± 0.18 | 224 ± 3 | 234 ± 9 |
| **PUR 4 + 2% OMMT** | 0.2080 ± 0.0021 | 345.4 ± 5.1 | 592.83 ± 3.35 | 0.730 ± 0.013 | 4.09 ± 0.20 | 122 ± 5 | 126 ± 8 |

It can be readily seen from TABLE IV that there is a proportional relationship between the added clay content and the apparent density of the prepared materials. This density decreases sharply when we added OMMT and increases with the increase of the OMMT rate added. This decrease in density is due to the pattern cells formed by the foam, it is noticed that the foam with 2% of OMMT is formed with open cells that contain other cells (presented in the SEM micrographs section). Moreover, we have an increase in rigidity by 445% and in young’s modulus by 197% for only the composite PUR 3 charged with 2% OMMT despite the opening cells, similar result was found by Saha et al.46 An increase only in rigidity by 52% and young’s modulus by 67%, were recorded For PUR 3 + 7% OMMT and PUR 3 + 10% OMMT, respectively.

From the values ​​of young’s modulus, there is an increase of this last only for PUR 3 + 2% OMMT and PUR 3 + 10% OMMT compared to PUR 3. This increase is probably due to closed cells for this composite. The other composites PUR 3 foam display a decrease in the young’s modulus and rigidity. This can be due to the perturbation of the polyurethane foams structure in the presence of clay nanoparticles, which disrupt hydrogen bonds.

C:\Users\pc\Downloads\Graph1.tif

Fig. 8.Tensile stress–strain curves for PUR composites.

C:\Users\pc\Downloads\Graph2.tif

Fig. 9. Tensile Strength–Elongation curves for PUR composites.

It is important to report that the rigidity and young modules for PUR 4 is twice as big as PUR 3, this can be explained by the strengthens of hydrogen bond presence, which increase with the presence of the diamino-1,2 propane. The mechanical proprieties of PUR 4 were degraded where we added 2% of the OMMT. Where, the density, the rigidity and the Young’s modulus decrease by 61, 26 and 24%, respectively. This decrease is probably due to perturbation of the hydrogen bond formed between the PUR, the diamino-1,2 propane and OMMT. However, PUR 4 formula presents optimal mechanical properties, compared to the other samples.

1. **Page 14, line 286, XRD analysis of clays and composites**

During the XRD analysis, we used the range of 2θ, higher that 4° for all samples clays and composites. The slight peak would appear towards an angle of 2θ of 4° for PUR 3+ 3% OMMT foam, indicating that the OMMT layers are partially exfoliated.

The previous commentary was:

The polyurethane foams prepared by E.A. Moawed45,W. Yang et al 46, H. Lian et al 47 and H. Liu et al 48 have the same shape and peak characteristic as that elaborated in this work.

The X-ray diffraction for the all samples of PUR 3 charged and uncharged foams show Broad diffuse peaks centered at 2θ=20° for, this peak show the amorphous character of the polyurethane foam. This peak can be correlated with the (110) reflection plane from PUR chains with an inter-chain d-spacing of 0.442 nm. 49

We notice a shifting of the peak characterizing the interlayer distance of the clay after polymerization for an angle 2θ less than 3 degrees. Which indicate that the layers of the OMMT are partially exfoliated for PUR 3+ 3% OMMT. This result is in good agreement with those obtained by Zheng et al8 in the work reported on the preparation of polyurethane/montmorillonite nanocomposite with a phosphorus compound.

The corrected commentary is after:

The X-ray diffraction pattern of the samples PUR 3 and PUR 3/OMMT at different rate was presented in Fig. 10. The polyurethane foams prepared by Moawed47,Yang et al 48, Lian et al 49 and Liu et al 50 have the same shape and peak characteristic as that elaborated in this work.

The X-ray diffraction for all PUR 3 loaded and unloaded foam samples shows large peak centered at 2θ=20°, which were not influenced by the addition of montmorillonite, this peak shows on the one hand the amorphous character of the polyurethane foam and on the other hand it indicates that the layers of OMMT are probably exfoliated. This peak characterizing the PUR chains is at the reflection plane (110) corresponds to a d-spacing between the chains of 0.442 nm.51

Nevertheless, there is a slight peak would appear towards an angle of 2θ=4° for PUR 3+ 3% OMMT foam, which indicates that the layers of OMMT are partially exfoliated. This result is in agreement with those obtained by Zheng et al8 in the reported work on the preparation of a polyurethane/montmorillonite nanocomposite with a phosphorus compound.

1. **Page 17, line 326, Add the magnification for SEM analysis instead of scale bar**

Yes, I agree, the scale bar was replaced by the magnification.

The previous SEM analysis was:

|  |  |
| --- | --- |
|  |  |
| *PUR 3 +2% OMMT* | *PUR 3 + 3% OMMT* |
|  |  |
| *PUR 3 +5% OMMT* | *PUR 3 +7% OMMT* |

Fig 9. SEM micrographs of nanocomposite foams PUR 3 + % OMMT at 200µm (300X ).

|  |  |
| --- | --- |
| **PUR 3 + 2 % OMMT 100 um** | **PUR 3 + 3 % OMMT 100 um** |
| *PUR 3 +2% OMMT* | *PUR 3 + 3% OMMT* |
| **PUR 3 + 5 % OMMT 100 um** | **PUR 3 + 7 % OMMT 100 um** |
| *PUR 3 +5% OMMT* | *PUR 3 +7% OMMT* |

Fig 10. SEM micrographs of nanocomposite foams PUR 3 + % OMMT at 100µm (600 X).

The corrected SEM analysis is after:

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| C:\Users\pc\AppData\Local\Temp\Rar$DRa0.467\1a.jpg | C:\Users\pc\AppData\Local\Temp\Rar$DRa0.702\1b.jpg |
| C:\Users\pc\AppData\Local\Temp\Rar$DRa0.438\1c.jpg | C:\Users\pc\AppData\Local\Temp\Rar$DRa0.231\1d.jpg |
| *C:\Users\pc\AppData\Local\Temp\Rar$DRa0.689\1e.jpg* | *C:\Users\pc\AppData\Local\Temp\Rar$DRa0.104\1f.jpg* |

Fig. 12. SEM micrographs of nanocomposite foams (a) *PUR 3 +2% OMMT* (b) *PUR 3 + 3% OMMT* (c) *PUR 3 + 5% OMMT* (d) *PUR 3 + 7% OMMT (e)* *PUR 3 + 10% OMMT* (f) *PUR 4 + 2% OMMT* (300X ).

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| C:\Users\pc\AppData\Local\Temp\Rar$DRa0.414\2c.jpg | C:\Users\pc\AppData\Local\Temp\Rar$DRa0.184\2d.jpg |
| *C:\Users\pc\AppData\Local\Temp\Rar$DRa0.248\2e.jpg* | *C:\Users\pc\AppData\Local\Temp\Rar$DRa0.245\2f.jpg* |

Fig. 13. SEM micrographs of nanocomposite foams (a) *PUR 3 +2% OMMT* (b) *PUR 3 + 3% OMMT* (c) *PUR 3 + 5% OMMT* (d) *PUR 3 + 7% OMMT (e)* *PUR 3 + 10% OMMT* (f) *PUR 4 + 2% OMMT* (600X ).

1. **Page 16, line 320, Added the SEM micrographs of sample PUR 4 + 2% OMMT**

Yes, I agree,

The SEM micrographs of PUR 3 + 10% OMMT and PUR 4 + 2% OMMT was added in the answer of the previous question. The commentary was ameliored in order to elucidate the effect of used catalyst system on pore size.

The previous commentary was:

The surface morphologies of samples (PUR 3 and PUR3/OMMT) were investigated via scanning electron microscopy (SEM) (FEI pattern Quanta 600 at 15 kV). We selected the samples PUR 3 + 2% OMMT, PUR 3 + 3% OMMT, PUR 3 + 5% OMMT and PUR 3+ 7% OMMT, to follow the variation of the structure of the poor and their sizes after incorporation OMMT. As shown in Fig. 9 and 10, each sample was observed at 200 and 100 μm, respectively.

PUR 3 + 7% OMMT formulation instituted organized and closed cells who have regular size. The shape of the alveolus is orbicular, with an estimated diameter of 100 μm \* 100 μm. On the other hand, PUR 3 + 2% OMMT, PUR 3 + 3% OMMT and PUR 3 + 5% OMMT formulations instituted open cells, with micro-alveolus cells. The size of the micro-alveolus was regular and their diameter was estimated to be 100 μm \* 100 μm, for PUR 3 + 2% OMMT foam, 60 μm \* 60 μm for PUR 3 + 3% OMMT foam and 50 μm \*40 µm for the PUR 3 + 5% OMMT foam. We conclude that the size of the alveolus decreases as a function of the increase in the rate of OMMT content in the foam. The size of the cells confirms the variation in the density, which increases as a function of the clay content added.

Jahanmardi et al.50 observed the same phenomenon, in other works a decrease in size and an increase in the number/area of ​​the alveoli were explained by the barrier effect of the clay. They have necessarily reduction of the nucleation energy.

The corrected commentary is after:

The surface morphologies of PUR composite foams samples were investigated via scanning electron microscopy (SEM). We selected the samples PUR 3 + 2% OMMT, PUR 3 + 3% OMMT, PUR 3 + 5% OMMT, PUR 3 + 7% OMMT, PUR 3 + 10% OMMT and PUR 4 + 2% OMMT to understand the effect of OMMT incorporation and the presence of diamino-1,2 propane as catalyst. As shown in Fig. 11 and 12, each sample was observed at 300 and 600 X, respectively.

PUR 3 + 7% OMMT, PUR 3 + 10% OMMT and PUR 4 + 2% OMMT formulations instituted organized, closed and regular size cells. The shape of the alveolus is orbicular, with an estimated diameter of 100 μm 100 μm, 100 μm 50 μm and 250 μm 80 μm, respectively. On the other hand, PUR 3 + 2% OMMT, PUR 3 + 3% OMMT and PUR 3 + 5% OMMT formulations instituted open cells with micro-alveolus. The size of this lasts is regular and their diameter was estimated to be 100 μm 100 μm for PUR 3 + 2% OMMT foam, 60 μm 60 μm for PUR 3 + 3% OMMT foam and 50 μm 40 µm for the PUR 3 + 5% OMMT foam.

We can conclude that cell size decreases with increasing the OMMT content in the foams. Starting from the 7% rate of OMMT, the cell structure becomes closed and becomes thicker. The size of the cells confirms the change in density, which increases with the added clay content. On the other hand, we can notice the influence of the diamino 1,2-propane catalyst which gave thick and closed cells after being opened for the foam load with 2% OMMT.

Jahanmardi et al.52 observed the same phenomenon, where a decrease in size and an increase in the number/area of ​​the alveoli were explained by the barrier effect of the clay which leads to the reduction of the nucleation energy.

1. **Page 3, line 66 and Page 15, line 317:**

Yes, I agree

Chlorure de potassium was remplaced by Potassium Chloride and pour was remplaced by for.

The previous text was:

Diamino-1,2-propane (C3H10N2, >98%), sodium chloride (NaCl, 99%), [Ammonium acetate (NH4 OAc, C2H7NO2, ≥98%](https://www.sigmaaldrich.com/catalog/product/sigma/a1542?lang=en&region=US)) and chlorure de potassium (KCl, 99%) was purchased from [Sigma-Aldrich](http://www.sigmaaldrich.com/catalog/product/aldrich/773204).

The corrected text is after:

Diamino-1,2-propane (C3H10N2, >98%), sodium chloride (NaCl, 99%), [Ammonium acetate (NH4 OAc, C2H7NO2, ≥98%](https://www.sigmaaldrich.com/catalog/product/sigma/a1542?lang=en&region=US)) and ‎ Potassium chloride (KCl, 99%) was purchased from [Sigma-Aldrich](http://www.sigmaaldrich.com/catalog/product/aldrich/773204).

The previous text was:

Which indicate that the layers of the OMMT are partially exfoliated pour PUR 3+ 3% OMMT.

The corrected text is after:

Which indicate that the layers of the OMMT are partially exfoliated for PUR 3+ 3% OMMT.

1. English should be significantly improved throughout the text.

Yes, I agree: in the revised manuscript, we tried to improve the English quality in the all manuscript.