



## CALCIUM CARBONATE, KAOLIN AND SILICA FILLED POLYPROPYLENE/POLYSTYRENE BLENDS: INFLUENCE OF FREE QUENCHING

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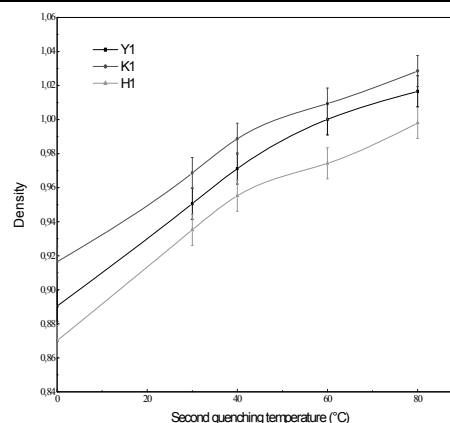
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In this study, the effects of free quenching on thermal, mechanical and morphological behaviors (properties) of blend-composites, namely polypropylene/polystyrene/calcium carbonate (PP/PS/CaCO<sub>3</sub>), polypropylene/polystyrene/ Kaolin (PP/PS/kaolin) and polypropylene/polystyrene/silica (PP/PS/SiO<sub>2</sub>) have been investigated.

Three different thermal treatments are used: A first free quenching from the melt state to air, a second free quenching from 130°C to various temperatures (0, 40, 80 °C and in air) and finally an annealing. The results showed that a quenching at 0°C according to the second treatment gives better results and a correlation between the mechanical and thermal properties is observed.



### INTRODUCTION

Once a thermoplastic polymer has been synthesized, it must be submitted to processing to obtain a finished product. This processing includes different variables, which influence its final characteristics, the most important of them are the thermal history acquired by the polymer during the solidification process.<sup>1</sup> Therefore, the effect of thermal history on polymers should be studied in detail, since the degree of crystallinity reached by these polymers depends on this history, the degree of crystallinity in turn governs the mechanical properties.<sup>2-5</sup>

Polypropylene (PP) is one of the most important commodity polymers which is widely used in

technical applications. Because of its good processability, relatively high mechanical properties, great recyclability and low cost, it has found a wide range of applications in household goods, packaging and automotive industry.<sup>6-11</sup> While its main disadvantage is its adequate (weak) mechanical properties, namely the impact strength, one of the conventional methods for overcoming this drawback is blending it with other polymers like polystyrene.<sup>12-16</sup> The polystyrene (PS) with its various applications is one of the most widely used commodity polymer materials.<sup>17-20</sup> For many years, PS has been very successfully used, it has the advantages of being clear hard easily processed and of low cost.<sup>19,21</sup>

The generation of residual stresses (RS) is also known to improve Izod impact strength of

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amorphous polymers like polycarbonate.<sup>22</sup> The study of the effect of residual stresses (RS) induced by free quenching on the mechanical behaviour of semi-crystalline polymers or blends semi-crystalline / amorphous polymers is a way to understand the relationship between the microstructure and the macroscopic properties of polymer materials such as polypropylene/ polystyrene blend and its composites. Residual stresses (RS) in inorganic and polymeric glasses have received a large attention in the past because of their technological importance. In amorphous polymers, the (RS) are in fact, totally attributed to volumetric fluctuations associated to the relaxation kinetics at the glass transition temperature ( $T_g$ ).<sup>23-25</sup> Many attempts have been made to determine the influence of residual stresses on properties of molded polymers.<sup>26-28</sup> During processing, the residual stresses are introduced by nearly all techniques used for polymer manufacturing. For example, the variations of the injection molding conditions may produce changes in molecular orientation. In order to avoid a contribution of flow induced orientation, free quenching experiments have been used by several investigators on semi-crystalline and amorphous polymers.<sup>27-32</sup>

Blendcomposites: polypropylene/polystyrene/calciumcarbonate (PP/ PS/  $\text{CaCO}_3$ ), polypropylene/polystyrene/ kaolin(PP/PS/Kaolin), polypropylene/polystyrene/Silica(PP/PS/ $\text{SiO}_2$ ) have been investigated in this study, the matrix being a blend of two incompatible polymers.<sup>10-13</sup> The blend compatibilized with styrene-ethylene-butadiene-styrene copolymer (SEBS)<sup>14,24</sup> was thermally treated (by annealing and free-quenching).The aim will to define the suitable conditions after heat treatment by various characterizations (thermal, mechanical, physical and morphological) carried out to determine the properties of the produced material. Many researches involve the crystallization of polymers, but few have studied the development of crystallization in presence of reinforcement and thermal treatment as in the present case of blend-composites. The addition of  $\text{CaCO}_3$ , kaolin or  $\text{SiO}_2$  modifies the properties of our material and its thermal treatment (annealing and free-quenching) can be one way of effective methods for blend-composites (PP/PS/ $\text{CaCO}_3$ ), (PP/PS/kaolin) and (PP/PS/  $\text{SiO}_2$ ) that can be used in various applications. It is important to explore the ways to prevent or minimize loss of toughness to reduce the sensitivity to the presence of notches and consequently, the loss of impact strength. To improve the impact strength of (PP/PS/ $\text{CaCO}_3$ ), (PP/PS/kaolin) and (PP/PS/ $\text{SiO}_2$ ),the effect of the first quenching from the melt state and the second

quenching from 130°C have been investigated, in addition to mechanical characterization, physical, morphological and thermal properties were measured and presented in this paper.

The objective of this work is to investigate the effect of the second quenching temperature on the mechanical, physical, thermal and morphological properties of (PP/PS/ $\text{CaCO}_3$ ), (PP/PS/kaolin) and (PP/PS/  $\text{SiO}_2$ ) blend-composites, and also to try to improve some of these properties. Therefore, the purpose of the present study is to give extensive experimental results that may lead to a better understanding of (RS) and their relation with some properties of these blend-composites.

## EXPERIMENTAL

### 1. Materials

Polypropylene PP was supplied by Basell Polyolefins under the name Koylene SS35N.It is a commercial granular Isotactic Homopropylene and its melt temperature is about 165°C.

Cristal polystyrene (PS) is a commercial PS, LG/GPPS, 15NFI witch was supplied by HAMTECH.

Styrene-ethylene-butadiene-styrene (SEBS) block copolymer, also called Kraton G 1650, was provided by Kraton Polymers, Germany.

Calcium carbonate ( $\text{CaCO}_3$ ) used was provided by the Tunisian Society of Industrial Calcium Carbonate (STICC), sold under the designation of Hermacarb 2FT it is a treated micronized filler of high purity and whiteness.

Kaolin was provided by SOLKA, complexe of kaolins of EL MILIA ALGERIA, under the designation of ECH: Kaolin KT2 as a treated filler.

Silica ( $\text{SiO}_2$ ) was provided by Rhodia, sold under the designation of ZMP1165, is a treated micronizedfiller of high purity and whiteness.

### 2. Preparation of samples and the first quench procedure

PP, PS,  $\text{CaCO}_3$ , kaolin and  $\text{SiO}_2$  were dried in an oven at 90°C for 24 hours to ensure removal of moisture before use. Weight compositions used include PP/PS/SEBS 50/50/5(this formulation is called B1), inorganic fillers ( $\text{CaCO}_3$ , kaolin or  $\text{SiO}_2$ ) at (5, 1.5 and 2.5 phr) respectively were added and then samples were prepared by melt mixing in a brabender plasticorder at a rotor speed of 60 rpm, at 190°C.

The blend was granulated into pellets by using a granulator. After that, pellets were put into the mold and pressed under 25 bars during 8 minutes at molding temperature of 190°C. The samples were then immediately quenched from moulding temperature into air during 15 minutes, this step was named "first quench".

### 3. Second quench procedure

The second quench procedure was carried out only for samples initially molded at 190°C. These samples were heated again in an oven at 130°C for 2 hrs for specimens (20min for films) and were immediately quenched a second time in water baths at different temperatures (0, 40, 80°C) and in air for 15 min. This procedure was named "second quench". All specimens included in this study are listed in Table 1.

*Table 1*  
List of samples and their abbreviations

Samples	Quenching temperature (°C)	Abbreviation
B1 /5 CaCO <sub>3</sub>	Reference	Y1
B1 /5 CaCO <sub>3</sub>	0	Y1.0
B1 /5 CaCO <sub>3</sub>	40	Y1.40
B1 /5 CaCO <sub>3</sub>	80	Y1.80
B1 /5 CaCO <sub>3</sub>	Air	Y1.air
B1 /5 CaCO <sub>3</sub>	Annealing	Y1.Annealed
B1 /1.5 kaolin	Reference	K1
B1 /1.5 kaolin	0	K1.0
B1 /1.5 kaolin	40	K1.40
B1 /1.5 kaolin	80	K1.80
B1 /1.5 kaolin	Air	K1.air
B1 /1.5 kaolin	Annealing	K1.Annealed
B1 /2.5 SiO <sub>2</sub>	Reference	H1
B1 /2.5 SiO <sub>2</sub>	0	H1.0
B1 /2.5 SiO <sub>2</sub>	40	H1.40
B1 /2.5 SiO <sub>2</sub>	80	H1.80
B1 /2.5 SiO <sub>2</sub>	Air	H1.air
B1 /2.5 SiO <sub>2</sub>	Annealing	H1.Annealed

#### 4. Annealed samples

Finally, to prepare a reference sample, an annealing is performed. Annealed samples were prepared using samples first quenched in air. These samples were heated again at 130°C for 2 hrs for specimens (20 min for films) and finally slowly cooled in the oven until room temperature (25°C) at a rate of about 5°C min<sup>-1</sup>. These samples were named “annealed samples”.

/min. The crystallinity fraction,  $\chi_c$ , was calculated by measuring the enthalpy of melting  $\Delta H_m$  and cold crystallization  $\Delta H_{cf}$  from the heating curves using equation (1), where  $\Delta H_{m0}$  corresponds to 209 J/g and refers to the enthalpy of 100% crystalline PP.<sup>33</sup>

$$\chi_c = (\Delta H_m - \Delta H_{cf}) / \Delta H_{m0} \quad (1)$$

## CHARACTERIZATION

### 1. Thermal Characterization:

#### Differential scanning calorimetry (DSC)

The thermal behavior of blend composites (B1/5CaCO<sub>3</sub>), (B1/1.5Kaolin), (B1/2.5SiO<sub>2</sub>) was analyzed in a PE NELSON Differential Scanning Calorimeter (DSC) model 1022 instrument in an inert N<sub>2</sub> atmosphere. Samples of (4-7 mg) were heated from 40°C to 300°C with a speed of 10°C

### 2. Morphological Characterization: Wide Angle X-ray Diffraction

The wide-angle X-ray scattering (WAXS) study was carried out with a Seifert (Ahrensburg, Germany) diffractometer working in the reflection mode. The experimental conditions were as follows: Ni-filtered Cu Ka radiation with 40 kV and 35 mA, angular range  $2\theta = 5-35^\circ$  and a scan rate: 0.05°/s. The degree of crystallinity WAXS of all samples was derived

from the ratio of the area corresponding to the crystalline peaks to the total area of the diffractogram.

### 3. Mechanical characterizations

#### 3.1. Microhardness measurements (H)

The microhardness (H) was measured at the room temperature by using a Leitz (Wetzlar, Germany) micro indentation equipped with a diamond indenter on square base. The value of (H) was diverted from the residual region thrown by penetration. Eight indentations were performed on the surface of each sample, and the average value was used for the data plot.

#### 3.2. Notched Izod impact strength ( $a_k$ )

Izod impact strength properties were determined at room temperature with a CEAST 6546/000 machine provided with a 15 J pendulum according to ASTM D 256-73, and using (63'12.7'3) mm<sup>3</sup> specimens. The latter were molded with a notch radius of 0.5 mm. The radius was chosen so that the tip of the notch was located in the residual compressive zone. At least, five specimens were tested and the average value was used for the data plot.

### 4. Physical characterization: Density measurement

Density was measured with a pycnometer by weighting the substance, usually in the liquid state, placed in the device and filled in the pycnometer to a mark on its neck or to the upper edge of a capillary tube, corresponding to the nominal volume of the pycnometer. The major advantages of the pycnometric method in determining the density are high accuracy of measurement (to 10<sup>-5</sup> g/cm<sup>3</sup>). At least, five specimens were tested and the average value was used for the data plot.

## RESULTS AND DISCUSSION

Several studies indicated that residual compressive stresses (RS) improve Izod impact strength by suppressing.<sup>32</sup> Moreover, the

presence of compressive stresses reduces the sensitivity to fatigue crack initiation at the surface, cracks, and flaws.<sup>33</sup> Rouabah *et al.*<sup>25, 35</sup> studied the effect of quenching process on mechanical and thermal properties of polycarbonate (PC). They found that a larger ductility can be obtained by quenching the samples from the melt state to the lowest temperature (0 °C). Akay and Ozden<sup>36</sup> showed that heat distortion temperature (HDT) and Izod impact strength were also sensitive to the (RS). In other respects, Siegmann *et al.*,<sup>29</sup> without calculating the change in free volume speculated that the increase in (HDT) was related to the decrease of free volume as a result of relaxation of thermal stresses and molecular orientation.

### The differential scanning calorimetry (DSC)

Fig. 1 (a, b and c) illustrates the thermograms of the blend-composites: (B1/5CaCO<sub>3</sub>), (B1/1.5 Kaolin) and (B1/2.5SiO<sub>2</sub>) respectively as a function of the second quenching temperature. Thermal tests can give very different results, depending on the variation of the second quenching temperature and filler. When varying the second quenching temperature or filler, thus varying T<sub>g</sub>, T<sub>m</sub>, ΔH<sub>m</sub> and the crystallinity degree (χ<sub>c</sub>). The crystallinity values calculated from the thermograms of the samples are listed in Table 2 (3<sup>rd</sup> column). It was found that in all blend-composites quenched, an increase in crystallization was observed, and a sensible increase in the melt temperature T<sub>m</sub> of these blend-composites as a function of second quenching temperature, varying from 159°C for Y1 to 166°C for Y1.80, from 162°C for K1 to 165°C for K1.80, and from 160°C for H1 to 165°C for H1.80, when varying the second quenching temperature from 25°C to 80°C.

The thermograms in Fig.1 (a, b and c) show that the highest amount of crystallinity is obtained with annealed composites and specimens second quenched at 80°C and in subsequently, higher than the other amounts of crystallinity of specimens quenched at various temperatures.

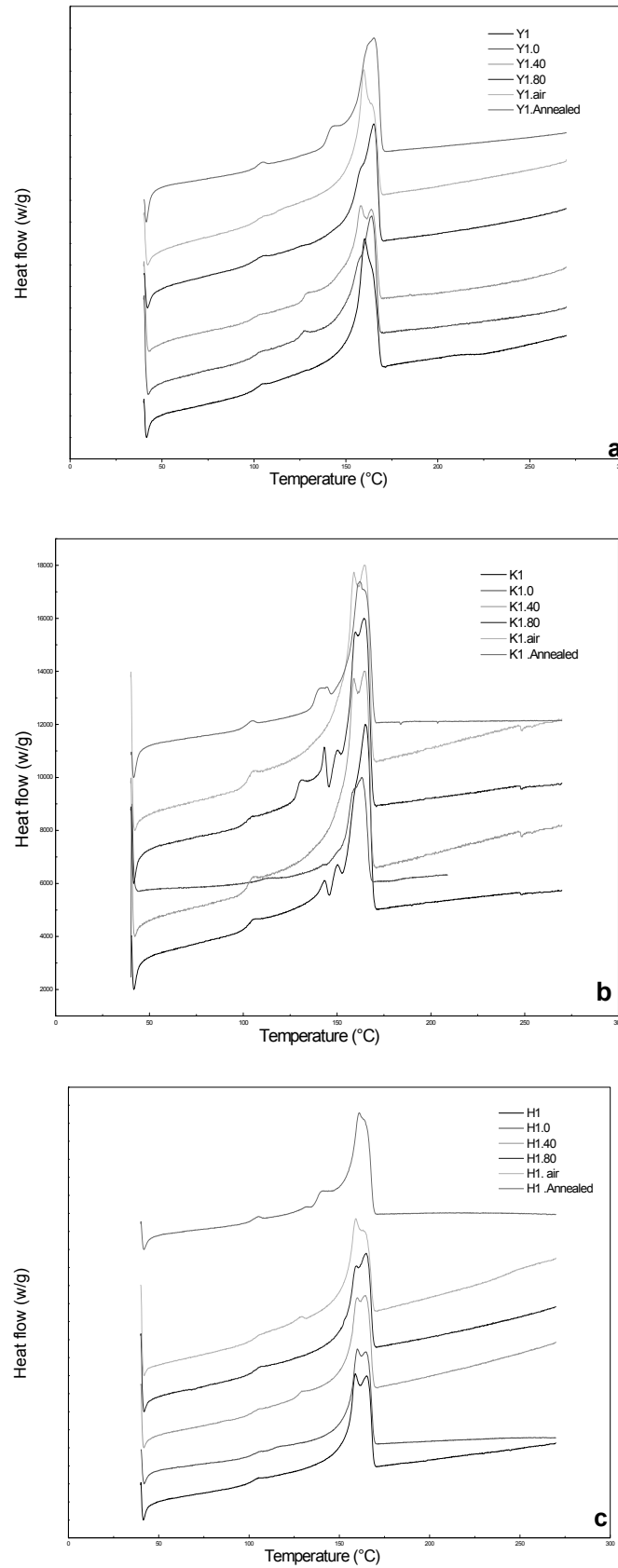


Fig. 1 – Differential Scanning Calorimeter DSC (a,b and c) of blend- composites (B1/CaCO<sub>3</sub>), (B1/Kaolin) and (B1/SiO<sub>2</sub>) respectively.

### Wide-angle X-ray scattering

Fig. 2 (a, b and c) shows the wide-angle X-ray diffractograms of (B1/5CaCO<sub>3</sub>), (B1/1.5Kaolin), and (B1/2.5SiO<sub>2</sub>) respectively, in first free quench from the melt state to air and in a second free quenching from 130°C to various temperatures (0, 40, 80 °C), in air and finally an annealing. The degrees of crystallinity are reported in Table 2 (2<sup>nd</sup> column), showing that increasing the quenching temperature leads to higher levels of crystallinity. The wide-angle X-ray scattering diffractograms of all the studied samples, for the same formulation are quite similar but with different intensity and thus crystallinity. At higher quenching temperatures, an increase in crystallinity is observed so of order parameter, indicating that at high temperatures, different effects are operative and nonuniform crystallinity profiles can be produced in the material, or more than one physical crystalline structure can be developed on the walls of the blend-composites. Consequently, these parameters influence crystallization kinetics, transition temperature (T<sub>g</sub>), melt temperature (T<sub>m</sub>), and crystallization temperature (T<sub>c</sub>) and the degree and type of crystallization.<sup>37</sup> Few researchers have given importance to correlation between properties and structure, and specifically, the thermal treatment effect on the crystallization of the blend-composite components.

The crystallinity values calculated from the diffractograms and thermograms of the samples are listed in Table 2.

### Microhardness measurements (H)

Microhardness (H) in Fig. 3 is noticed to gradually grow with the increase in the second quenching temperature, that may be due to the increase of order or the crystallinity degree within the material<sup>38</sup>, which is in correlation with the results found in differential scanning calorimetry (DSC) and wide-angle X-ray scattering (RX). It has also been observed that a low second quenching temperature decreases microhardness.

The effects of free quenching on the structure and micromechanical properties of (B1/5CaCO<sub>3</sub>), (B1/1.5 Kaolin) and (B1/2.5SiO<sub>2</sub>) blend-composites were investigated. The lowest second quenching temperature provokes a decrease in this mechanical property.

### Impact strength

Fig. 4 shows the notched Izod impact strength ( $a_k$ ) as a function of second quenching temperature of (B1/5CaCO<sub>3</sub>), (B1/1.5Kaolin) and (B1/2.5SiO<sub>2</sub>) blend-composites. The Izod impact strength

reaches maximum values for the second quenching temperature of 0°C. On the other hand, one can notice that Izod impact strength ( $a_k$ ) values decrease with the increase of the second quenching temperature. For example, for notched specimens, Izod impact strength ( $a_k$ ) decreases from 6 kJ/m<sup>2</sup> to 3.600 kJ/m<sup>2</sup> for quenching temperatures ranging between 0°C and 80°C. These variations are more prominent for the annealed samples Y1, K1 and H1 (3.34 kJ/m<sup>2</sup>, 3, 12 kJ/m<sup>2</sup> and 3, 72 kJ/m<sup>2</sup>) respectively. Generally, the impact strength of semi-crystalline polymers depends on the crystalline phase that develops during heat treatment. There is a drop in the impact strength with increasing crystallinity.

The notched Izod impact strength as a function of second quenching temperature, is presented in Figure 4. The highest notched Izod impact strength is obtained at second quenching temperature of 0 °C, which is also related to a good ductility under these conditions. The results show a drastic decrease of the notched Izod impact strength values as the second quenching temperature increases. As the impact strength can be correlated with the ability of polymer chains to execute segmental motion and hence dissipate the energy associated to crack propagation, an increase in free volume may also be responsible of the increase of impact strength. The evolution of crystallinity may be due to the evolution of order in the blend-composite, which gradually grows up with the increase in the second quenching temperature probably due to the decrease of the molecular mobility of the matrix blend. With the various thermal treatments, the highest amount of crystallinity is obtained with annealed composites and specimens second quenched at 80°C.

### Density (d)

The density changes have even been reported as a result of various thermal treatments in polystyrene and polycarbonate.<sup>39,40</sup> The density variations as a function of second quenching temperature are shown in Fig. 5, where the density increases clearly as the second quenching temperature increases. In the case of faster cooling, which corresponds to 0°C second quenching temperature, the macromolecular chains have less time to reorganize. This induces an increase of the free volume and therefore, a lower density. According to Van Krevelen<sup>41</sup>, the density (d) is correlated to Young's modulus (E). As Young's modulus is, on turn, correlated to the crystallinity degree, this means that samples having a lower density have also a lower crystallinity degree.

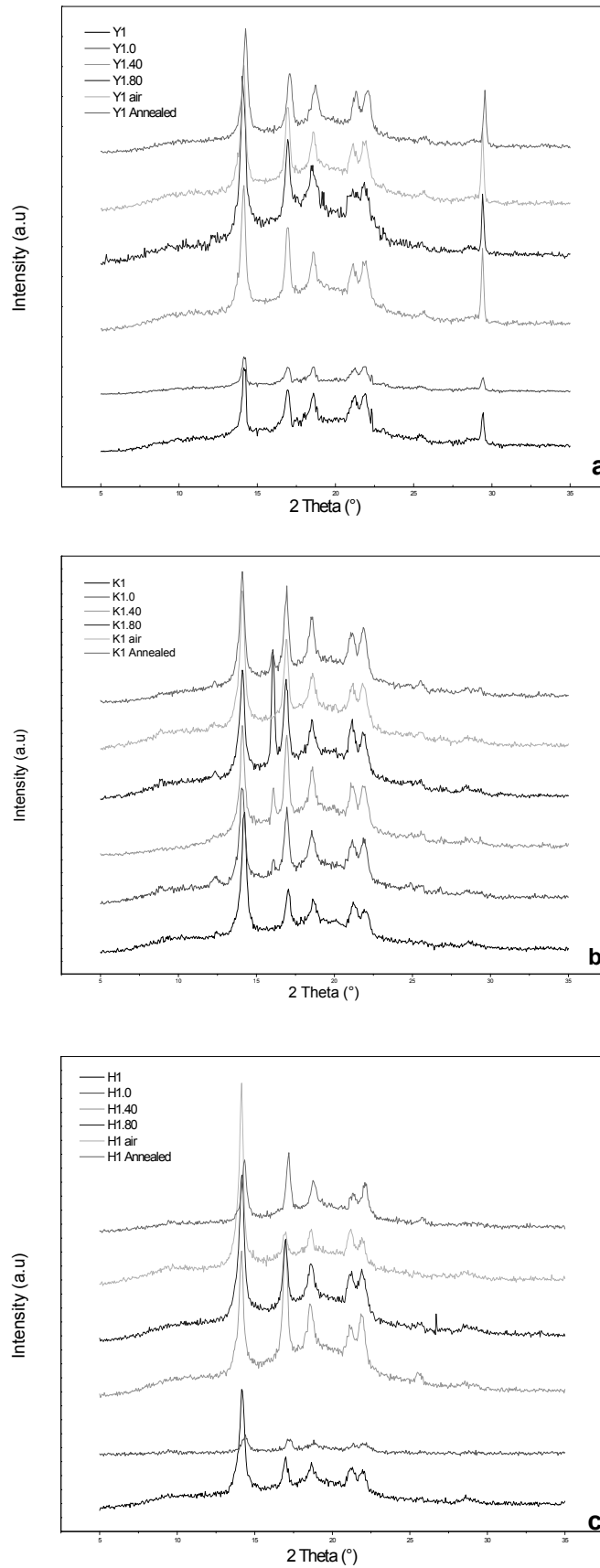


Fig. 2 – Wide angle X-ray scattering (WAXS) diffractograms (a,b and c) of blend-composites (B1/CaCO<sub>3</sub>), (B1/Kaolin) and (B1/SiO<sub>2</sub>) respectively.

Table 2

Wide angle X-ray scattering (WAXS) and (DSC) crystallinity		
Samples	Cristallinity(RX)	Cristallinity(DSC)
Y1	48,34	49,18
Y1.0	42,15	45,54
Y1.40	52,13	51,75
Y1.80	53,20	54,32
Y1.air	47,92	49,82
Y1.Annealed	54,14	56,40
K1	48,35	47,60
K1.0	46,25	44,82
K1.40	52,74	53,46
K1.80	56,83	51,28
K1.air	53,72	56,35
K1.Annealed	56,26	57,34
H1	46,05	46,52
H1.0	41,47	42,76
H1.40	50,26	49,14
H1.80	53,78	52,85
H1.air	50,91	48,87
H1.Annealed	50,53	51,42

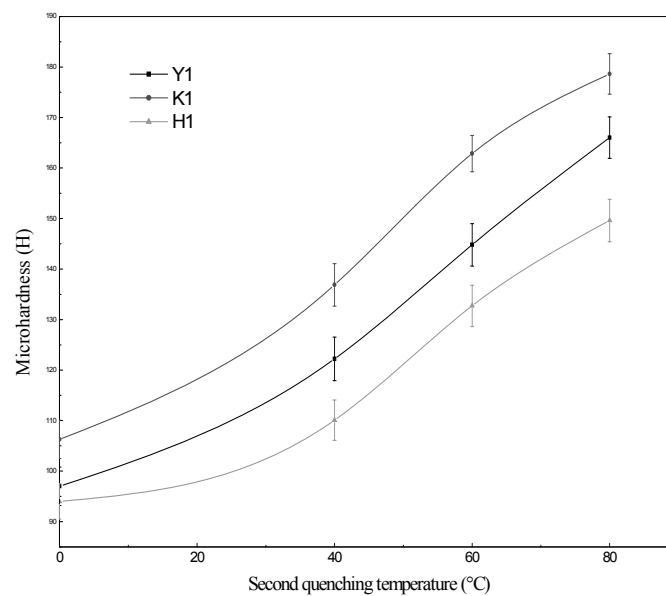


Fig. 3 – Microhardness (H) of blend-composites (B1/CaCO<sub>3</sub>), (B1/Kaolin) and (B1/SiO<sub>2</sub>) respectively as a function of second quenching temperature.



As observed in Fig. 5, the fast quenching ( $80^{\circ}\text{C}$ ) tends to increase the density (and crystallinity). It is important to note that the samples second quenched at  $0^{\circ}\text{C}$  have consistently slightly lower density (and crystallinity) than the samples second quenched in air and at  $40^{\circ}\text{C}$ . On the other hand, the annealing leads to a significant increase in the density of (B1/5CaCO<sub>3</sub>), (B1/1.5Kaolin) and (B1/2.5SiO<sub>2</sub>) blend-composites (1,0240, 1,0381 and 1,012) respectively. Compared to the quenched material, this is largely explained by the significant gain of crystallinity associated to this

heat treatment. However, during annealing, thermal residual stresses are more or less limited, probably due to the origin of structural stresses which dominate, where the values of microhardness and density are higher compared to those of quenched samples. Quenched samples at high temperatures (at  $80^{\circ}\text{C}$ ) are consistently more rigid than those from rapid quenching (at  $0^{\circ}\text{C}$ ). This is mainly due to different stresses of structural origin that probably increase the degree of crystallinity.

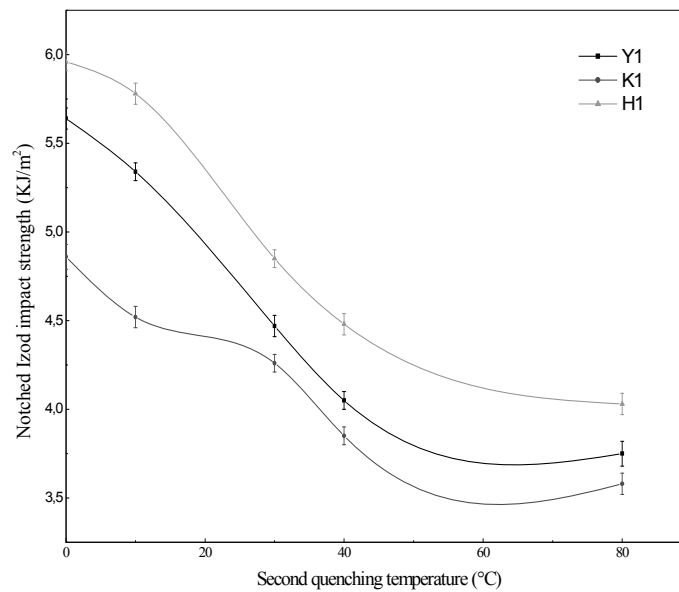


Fig. 4 – Notched Izod impact strength of blend-composites (B1/CaCO<sub>3</sub>), (B1/Kaolin) and (B1/SiO<sub>2</sub>) respectively as a function of second quenching temperature.

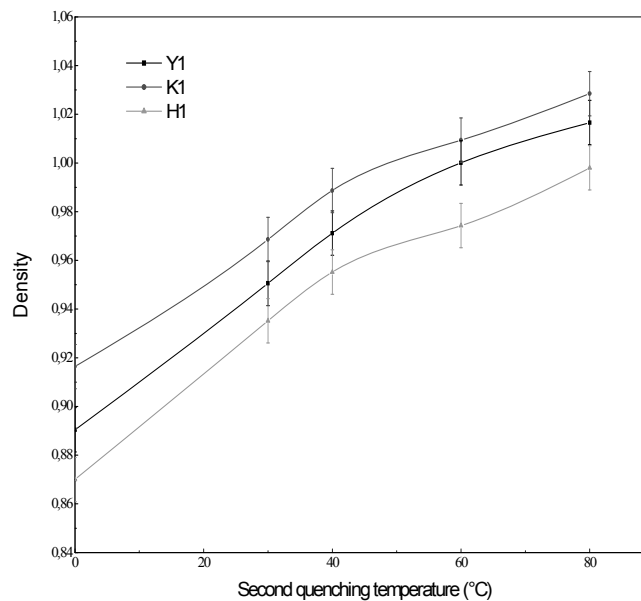


Fig. 5 – Density of of blend-composites (B1/CaCO<sub>3</sub>), (B1/Kaolin) and (B1/SiO<sub>2</sub>) respectively as a function of second quenching temperature.

## CONCLUSION

Thermal treatment affects the ((B1/5CaCO<sub>3</sub>), (B1/1.5Kaolin) and (B1/2.5SiO<sub>2</sub>) blend-composites properties by affecting their crystallinity. Results show the development of the crystallinity induced by slow quench and annealing.

The influence of heat treatment (quenching and annealing) clearly appear in all the properties studied. The properties of the same samples obtained after second quenching at 0°C (corresponding to rapid quenching) are very distinct from properties corresponding to slow quench (at 80°C) and in air.

It was shown that a larger ductility can be obtained by quenching the samples at the lowest temperature (0°C) from 130°C. A faster cooling, corresponding to the lower quenching temperature, generates probably more residual stresses and free volume. The latter increases Izod impact strength but decreases microhardness and density. The effect of quenching process on mechanical, physical, morphological and thermal properties of (B1/ 5CaCO<sub>3</sub>), (B1/1.5Kaolin) and (B1/2.5SiO<sub>2</sub>) blend-composites were investigated via impact, microhardness, physical and thermal measurements. The minimum density reached after a second quench from 130°C to 0°C was associated to the presence of more free volume, which leads to a higher molecular mobility. This explains the increase of Izod impact strength, and the decrease of microhardness and crystallinity degree ( $\chi_c$ ). Materials with identical composition can show several properties such as  $T_m, \Delta H_m, H_a, k_d$  and crystallinity degree ( $\chi_c$ ). These properties depend on the material microstructure, which in turn can be determined by the thermal treatment.

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