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Study on Mechanical Properties and Material Distribution of Sandwich Plaques Molded by Co-injection

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Abstract

In the sandwich injection molding process (co-injection), two different polymer melts are sequentially injected into a mold to form a part with a skin/core structure. Sandwich molding can be used for recycling, improving barrier and electrical properties or producing parts with tailored mechanical properties.

In this study the evaluation of flexural modulus and impact strength of co-injected plaques have been investigated. Virgin and short glass fiber reinforced (10 and 40%) polypropylene were used in six different combinations of sandwiched layers. The skin and core thicknesses were measured by optical microscopy and used to calculate the theoretical flexural modulus, which was compared to the experimentally measured modulus. Fiber orientation states were also observed by scanning electronic microscopy (SEM) at some specific locations and their effect on mechanical properties discussed. The experimental results indicate that an important improvement in transverse modulus, near the gate, is obtained when the virgin polypropylene (PP) is used as a skin and 40 % short glass fiber polypropylene (PP40) as core. When both skin and core are made of PP40, the flexural moduli are slightly higher than conventionally injected PP40.

INTRODUCTION

The co-injection molding process (or sandwich injection molding) has been in use since the early 70's and was first invented by Garner and Oxley of ICI [1, 2]. This process consists of injecting sequentially and/ or simultaneously two or more polymers in the same mold. The first polymer melt entering the mold forms the skin material and the second constitutes the core resulting in a sandwich structure.

In the past decade, interest in this process has increased and a lot of efforts have been made to develop it and to meet the requirements of the plastic industry. The principal interest in this technology is the possibility to combine skin and core materials having different physical and chemical properties. For instance, by using a fibre reinforced polymer for the core and a virgin polymer with a good surface aspect for the skin, one can obtain interesting mechanical properties and keep the aesthetic properties of the part. In some cases, the co-injection process can be an important alternative to one-shot conventional injection by using cheaper recycled plastics but with still good physical properties as the core material while the product outer skin is made of an appropriate virgin plastic [3, 4].

Few investigations on the mechanical properties of sandwich injection molding are reported in the literature. Donavan et al. [4] measured Gardner impact strength on sandwich plaques containing two different grades of ABS. It was concluded that the impact was mainly determined by the skin material. Tomari et al. [5] studied bending properties of specimens cut from sandwich-molded plates using polyamide (PA) as a skin and polycarbonate (PC) as a core. Their results clearly show that the skin material to a large extent determine the flexural properties. In fact, similarly to conventional injection molding, the skin material in the sandwich molding is exposed to higher shear rate and steeper temperature gradients which leads to increased levels of orientation and consequently improved properties. Hamada, et al. [6] studied flexural properties of different sandwich structures containing virgin, short and long fiber reinforced PA and PC and found that flexural stiffness was strongly influenced by fiber orientation produced during processing. Selden [7] investigated the effect of molding parameters on material distribution and mechanical properties of co-injection molded plates using an experimental design plan. The plates were molded with a polyamide 6 as skin and 20% glass fibre reinforced polybutylene-terephthalate (PBTP). The statistical analysis showed that the mechanical properties such as flexural and impact strength have a high correlation with the skin/core distribution.

The main objective of this paper is to study primarily the distribution of flexural and impact properties in a sandwich injection molded plaques obtained by using a virgin and short glass fiber reinforced polypropylene as skin and core materials. The fiber orientations were observed by SEM at the skin-core interface of some samples and the skin thickness measured by optical microscopy. Their effects on mechanical properties are also discussed.

EXPERIMENTAL

Injection molding was done on a 150 ton Engel co-injection machine. This machine is equipped with two injection units oriented at an angle of 90 ° to each other. The diameter of the horizontal screw (injection unit 1) is 40 mm and the diameter of the vertical screw (injection unit 2) is 30 mm. The two injection units can be operated independently, allowing the press to be used for both conventional and co-injection molding.

The materials used in this study were virgin polypropylene (RTP 0199 natural), 10% and 40% short glass fiber (SGF) reinforced polypropylene (RTP 0199 A and RTP 0199 D respectively). These three materials are respectively designated as PP, PP10 and PP40. The sandwich combinations considered are as follows: PP/PP, PP/PP10, PP/PP40, PP40/PP, PP40/PP10, and PP40/PP40. In this nomenclature, the first material constitutes the skin while the second designates the core. The viscosity versus shear rate curves for the three materials (PP, PP10 and PP40) at 200 °C are shown in Fig. 1.

The geometry of the mold cavity used in this study is shown in Fig.2. It consists of two symmetric rectangular plaques having lateral dimensions 75×100 mm and a 3 mm thickness. A semicircular gate, 8 mm in diameter was located at the center of the longer side of each rectangle.

The machine control setting data for injection and co-injection molding are shown in Table 1. The co-injection molding was performed in the following sequence: a given amount of skin material is first injected in the cavity by injection unit 1, the total amount of the core material is then injected by injection unit 2; finally a much smaller amount of skin is injected to seal the gate. The core material was colored black with a 1% master batch to facilitate identification of the interface between the core and the skin within the plaques. Using injection unit 1, conventional injection moldings were made from the three base materials: PP, 10% SGF reinforced PP and 40% SGF reinforced PP

To carry out the flexural tests, specimens were cut from the co-injection plaques both parallel and perpendicular to the flow direction. Two specimens were cut at distances of 12 mm and 51 mm from the gate respectively and had dimensions of 100×12×3 mm, while two other specimens were cut at distances of 12 mm and 44 mm from the lateral edge of the plaque, with dimensions of 75×12×3 mm. All the specimens are labeled according to the nomenclature shown in Figure 3 a, b. The flexural tests were performed using a standard three point-bending fixture mounted on an Instron universal test machine, in accordance with ASTM-D790. The support span was 50 mm long and the crosshead speed of loading was 2 mm/s. All measurements were done at ambient temperature of 23 °C.

The flexural tests were carried out on the six co-injection combinations described earlier and on specimen bars made of the three base materials cut from conventionally injection-molded plaques.

Impact tests were conducted in an instrumented falling weight impact Dynatup machine. Two samples having a square shape (42 x 42 mm) were cut from the molded plates in the central zone and labeled G (near the gate) and E (far from the gate) as shown in Fig. 3d. These samples were laid flat with a clamping on a circular support containing a central hole with a diameter of 40 mm. The tests were performed at a test speed of 3m/s, using a hemispherical head with a diameter of 12.7 mm and a 1.18 kg anvil. For each test the force at peak (maximum load) and the total energy were determined. Five specimens were tested for each value of impact strength.

Measurements of the skin and core thicknesses for each specimen were done on cross sections cut along and transverse to the flow at positions SG, SC, SE and SL (Fig. 3c). These positions were chosen because of their proximity to the zone subjected to the flexural load. An optical microscope Bausch & Lomb equipped with a CCD video camera was used to acquire cross section images, which were subsequently analyzed using the Visilog image analysis software.

Samples for Scanning Electron Microscopy were obtained by breaking specimens cooled in liquid nitrogen. The samples were mounted on a supporting plate and the sections in the broken zone were then coated with a gold-palladium alloy using a sputtering technique. A JEOL JSM 6100 instrument was used to view the microstructure at varying magnifications.

RESULTS AND DISCUSSION

Flexural properties

The results of the transverse flexural modulus for the virgin polypropylene PP, the short fiber reinforced composites PP10 and PP40 and the three sandwich combinations where PP constitutes the skin are shown in Fig.4. First, it can be seen in this figure that the fiber reinforced polypropylenes PP10 and PP40 have higher flexural modulus than the virgin polypropylene as expected. These three materials are used here for reference. For the co-injected samples, we also can see an increase in the flexural modulus as the fiber content of the core is increased. For these combinations the improvement is more pronounced for the samples cut near the gate (TG) than those cut close to the end of the plaque (TE). Note that for PP/PP40, the modulus of specimen cut near the gate (3552 MPa) is almost equivalent to the moduli of the specimen obtained by conventional injection molding of PP40 (3876 MPa). The advantage of using co-injection in this case would be to achieve a better surface appearance than 40 % glass fiber reinforced PP plaque injected conventionally, while maintaining similar flexural properties.

For the samples cut in the flow direction and where PP constitutes the skin (Fig. 5), we do not observe very significant differences in the longitudinal flexural moduli between the locations close to the edge (LL) and at the center of the plaque (LC). However the samples with 40 % reinforced core show an improvement of about 66 and 63 % respectively at the locations LL and LC with respect to combination PP/PP10 at the same locations. Note also that the flexural modulus of samples LL, LC and TE are of the same order of magnitude (see Fig. 4 and 5).

Figure 6 presents the transverse flexural modulus for PP, PP10, PP40 and the three sandwich combinations where PP40 is used as skin. In this case the effect of the reinforcing fibers in the skin layer on the flexural modulus is higher but we do not observe significant differences between TG and TE. We can observe also that for the combination PP40/PP40 at location TG the modulus is higher than conventionally injected PP40. It is believed that this improvement is due to a more efficient use of the fibers within the layered microstructure, such as the presence of a sub layer of flow-oriented fibers just below the skin/core interface.

In the flow direction, similar observations can be made in the case of sandwich structures where PP40 is used as skin as shown in Fig.7. Note that as in Fig.6, the combination PP40/PP40 at location LL and LC shows also higher modulus than PP40.

Optical microscopy

The ratio of the skin to part thickness for the six sandwich combinations are shown in Fig. 8-9. Where PP is used as skin (Fig.8), we can observe that the thickness at position SE increases with the fiber content of the core. Since the viscosity of the core increases with the fiber content, it is expected that the flow length of the core will decrease resulting in a thinner skin at the gate and a thicker skin at the front. The effect of the viscosity on the flow length of the core has been shown by simulation work done by Ilinca and Hétu [8]. Thicknesses at location SC and SL have about the same magnitude as thickness SE and this might explain the similar values obtained for the flexural moduli of specimens LC, LL and TE (see Fig.4 and 5). Comparing Fig.4 and 8, it can be concluded that for the combination PP/PP40, the flexural modulus is directly related to the skin thickness fraction: the lower it is (as in position SG), the higher is the flexural modulus (as for specimen TG). In this specific case, the flexural modulus is almost equivalent to that of specimen with 40% PP reinforced conventionally injection molded.

Stiffness prediction

The calculation of the flexural modulus of the co-injection specimens was done using the composite beam theory. Assuming perfect adhesion at the skin/core interface so that both skin and core deform simultaneously, the flexural modulus can be calculated as [6]:

$$E = (E_s I_s + E_c I_c) / I \quad (1)$$

where E_s, E_c are bending moduli of skin and core materials respectively, I the second moment of inertia of the sandwich part, and I_s, I_c are second moments of inertia of skin and core materials respectively.

I_s and I_c can be calculated with equations (2) and (3):

$$I_s = b \left(\frac{t_s^3}{6} + t_s \frac{(t_s + t_c)^2}{2} \right) \quad (2)$$

$$I_c = b \left(\frac{t_c^3}{12} \right) \quad (3)$$

where b is specimen width, t_s and t_c are thickness of skin and core respectively.

Equation (1) was applied at locations TG, TE, LC and LL, where the flexural moduli of the skin and the core material were previously measured on the sample obtained by conventional injection molding at the above-mentioned locations. In doing this, the effective local fiber orientation is, to some extent, taking into account. Thicknesses t_s and t_c used in this equation are measured at positions SG, SE, SL and SC (see fig. 3c) for the sandwich parts tested at locations TG, TE, LL and LC respectively. The choice of these positions was dictated by the fact that they support most of the load during the bending test. As an example, to calculate the flexural modulus of the sandwich combination PP/PP40 at location TG, we took the bending moduli of conventionally injected PP (skin) and PP40 (core) at the same location and we used the thicknesses of PP and P40 at position SG from the co-injected sample.

The calculated flexural moduli using equations (1), (2) and (3) are shown in Tables 2 and 3. It is seen that for all combinations, the calculated values compare reasonably well with experimental values. The highest discrepancies were observed for the two combinations PP/PP40 and PP40/PP at the same location LC. This seems to indicate some limitations in using equation (1) due to the complex behavior of the glass fiber in sandwich co-injected moldings (fiber orientation and spatial distribution), especially when either the core or the skin is highly reinforced.

Impact tests

The results from the impacts tests on single polymer plates and sandwich molded parts at locations G and E are presented in Figures 10 to 13. For the sandwich structure where PP constitutes the skin (Fig. 10), the only improvement in the strength is observed for the combination PP/PP40 at location G. For the combination where PP40 constitutes the skin (Fig. 11) the impact strength increase with the increase of the fiber content in the core. When a fiber reinforced polymer is added an improvement of impact load and energy is always observed. Analysis of Figures 10 and 11 indicates that if the skin is substantially reinforced, then the core contribution to the sustained impact force is more significant.

In general similar conclusions can be drawn from Fig. 12 and 13 when we consider the total energy.

Scanning electronic microscopy

Some of the fractured molded parts and sandwich structures were observed with a scanning electron microscope (SEM) to examine the fiber orientation distribution. Samples PP40, PP/PP40 and PP40/PP were chosen for microscopic observations of the fractured surfaces. The selection of PP/PP40 was based on the fact that this combination presented comparatively higher mechanical properties near the gate. Figure 14a,b shows SEM microphotographs of sample LC broken at the center for the specimen

PP40. The change in fiber orientation from the skin region (Fig.14 a) to core region (Fig. 14b) is quiet complex. It is well known that in fiber reinforced injection molded part the fiber orientation distribution can vary from position to position and through the thickness of the part [9-10]. We can see in the skin region that the fibers are mostly oriented in flow direction due to maximum shear stress. Close to the wall, a thin layer of randomly oriented fibers can also be seen. This random orientation of the fibers in this layer can be explained by the dynamics of the fountain flow of the melt front [11].

Figures 15a, b show SEM pictures of sample TG broken at the center for a specimen PP/PP40. It has been observed that the fiber orientation at the skin/core interface (Fig. 15a) is preferentially in the flow direction. Holes were observed where the fibers were pulled out either perpendicular or at some angle to the cracked surface of the matrix. In the central region of the core (Fig. 15b), most of the fibers lie at an angle to the flow. The dislodging of fibers lying parallel or near parallel to the cracked surface resulted in long furrows.

Similarly, for the sample LC broken at 18 mm from the end of the part PP/PP40, the fibers at the interface (Fig. 16a) are seen to be also oriented in the flow direction, while in the central region of the core the fibers are lying transverse to the flow.

CONCLUSION

This study was concerned with the spatial evaluation of the flexural modulus and impact strength of co-injected plaques using virgin and glass fiber reinforced polypropylene as skin and core materials. The main conclusions that can be drawn from this work are as follows:

-For the co-injected plaques where virgin polypropylene is used as skin, the flexural modulus in the transverse direction is much higher near than far from the gate and especially for the combination PP/PP40.

- For the combination where PP40 is used as skin, there are no significant differences in the flexural moduli between specimen of same direction but different location, except that the PP40/PP40 presented a higher modulus compared to conventional injection molded PP40. This improvement is due to more efficient use of the fibers within the layered microstructure.

-The flexural modulus of co-injected parts can be predicted analytically by a simplified composite beam theory, using the measured moduli and thickness of the individual skin and core materials.

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Table 1: Molding conditions

Parameter	Conventional injection Injection unit # 1	Co-injection molding	
		Injection unit #1 (Skin)	Injection unit # 2 (Core)
Cylinder temperature, °C	200		
Nozzle temperature, °C	200		
Mold temperature, °C	42		
Cooling time, s	25		
Holding time, s	5		---
Injection speed, mm/s	45	30	60
Stroke, mm	45	16	38

Table 2: Calculated flexural moduli (in GPa) for sandwich structures (PP as skin)

Combination	TG			TE			LL			LC		
	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp}/ E_{cal}
PP/PP	1.475	1.352	1.09	1.328	1.422	0.93	1.343	1.381	1.01	1.350	1.350	1.00
PP/PP10	1.976	1.771	1.11	1.610	1.573	1.02	1.453	1.437	1.01	1.356	1.466	0.92
PP/PP40	3.552	2.902	1.21	1.935	1.886	1.02	2.200	1.967	1.11	2.125	2.741	0.77

Table 3: Calculated flexural moduli (in GPa) for sandwich structures (PP40 as skin)

Combination	TG			TE			LL			LC		
	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp} / E_{cal}	E_{exp}	E_{cal}	E_{exp}/ E_{cal}
PP40/PP	2.851	2.749	1.03	3.218	3.214	1.00	4.113	3.726	1.10	4.202	3.405	1.23
PP40/PP10	3.269	3.177	1.02	3.380	3.401	0.99	4.228	3.487	1.21	3.971	3.497	1.13
PP40/PP40	4.132	3.876	1.06	3.804	3.732	1.01	5.175	4.806	1.07	4.901	4.385	1.11

Figure captions

Figure 1: Shear viscosity of materials at 200 °C

Figure 2: Co-injected plaques dimensions

Figure 3: Specimen cut from molded plaques for: a) transverse flexural tests (TG and TE), b) longitudinal flexural tests (LL and LC), c) skin thickness measurements at position SG, SE, SC and SL, d) impacts tests (G and E). All dimensions are in mm.

Figure 4: Transverse flexural modulus (PP as skin)

Figure 5: Longitudinal flexural modulus (PP as skin)

Figure 6: Transverse flexural modulus (PP40 as skin)

Figure 7: Longitudinal flexural modulus (PP40 as skin)

Figure 8: Skin to part thickness ratio (PP as skin)

Figure 9: Skin to part thickness ratio (PP40 as skin)

Figure 10: Maximum load versus materials (PP as skin)

Figure 11: Maximum load versus materials (PP as skin)

Figure 12: Total energy versus materials (PP as skin)

Figure 13: Total energy versus materials (PP40 as skin)

Figure 14: SEM pictures of specimen PP40 taken from the sample LC, a) Skin region, b) Core region

Figure 15: SEM pictures of specimen PP/PP40 taken from sample TG broken at the center, a) Interface skin/core, b) Central region of the core.

Figure 16: SEM pictures of specimen PP/PP40 taken from sample TG broken at 18 mm from the end of the part PP/PP40, a) Interface skin/core, b) central region of the core.

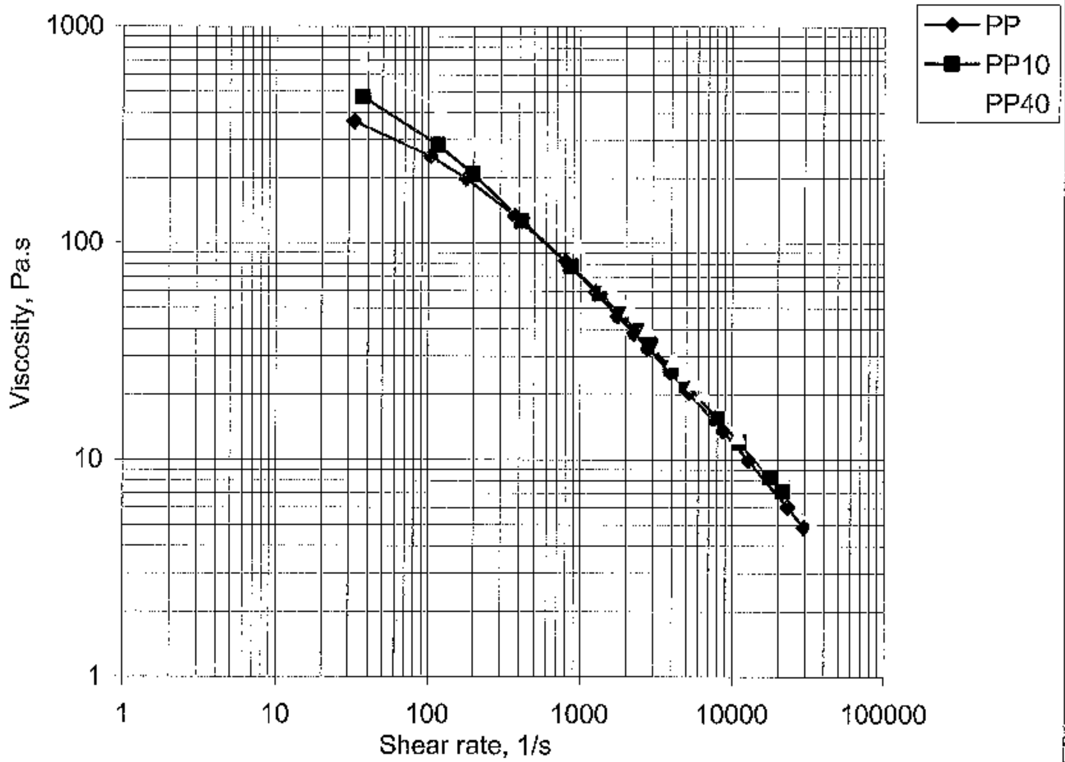


Fig.1. Shear viscosity of materials at 200 °C

Ait-Messaoud, Fig. 1

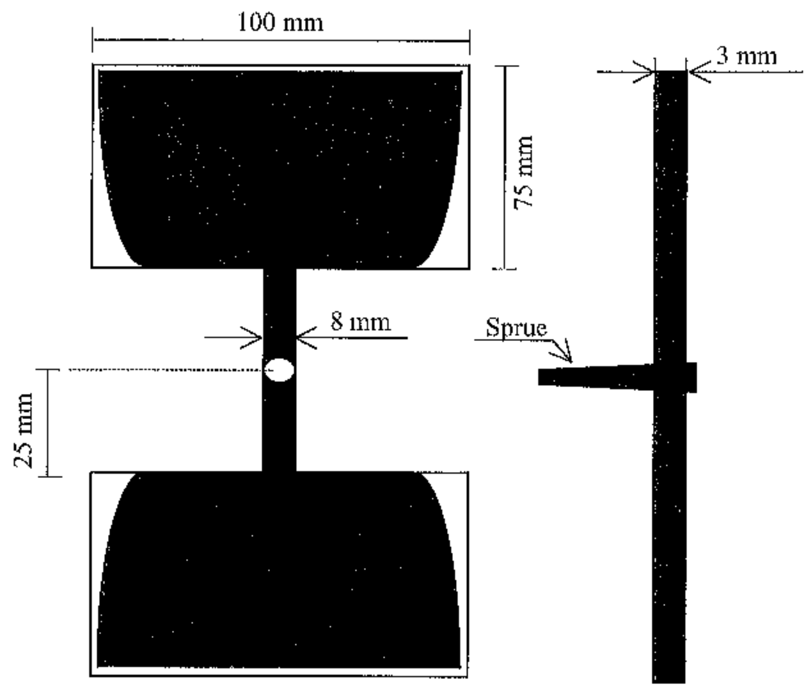


Fig. 2. Co-injected plaque dimensions

Ait-Messaoud, Fig. 2

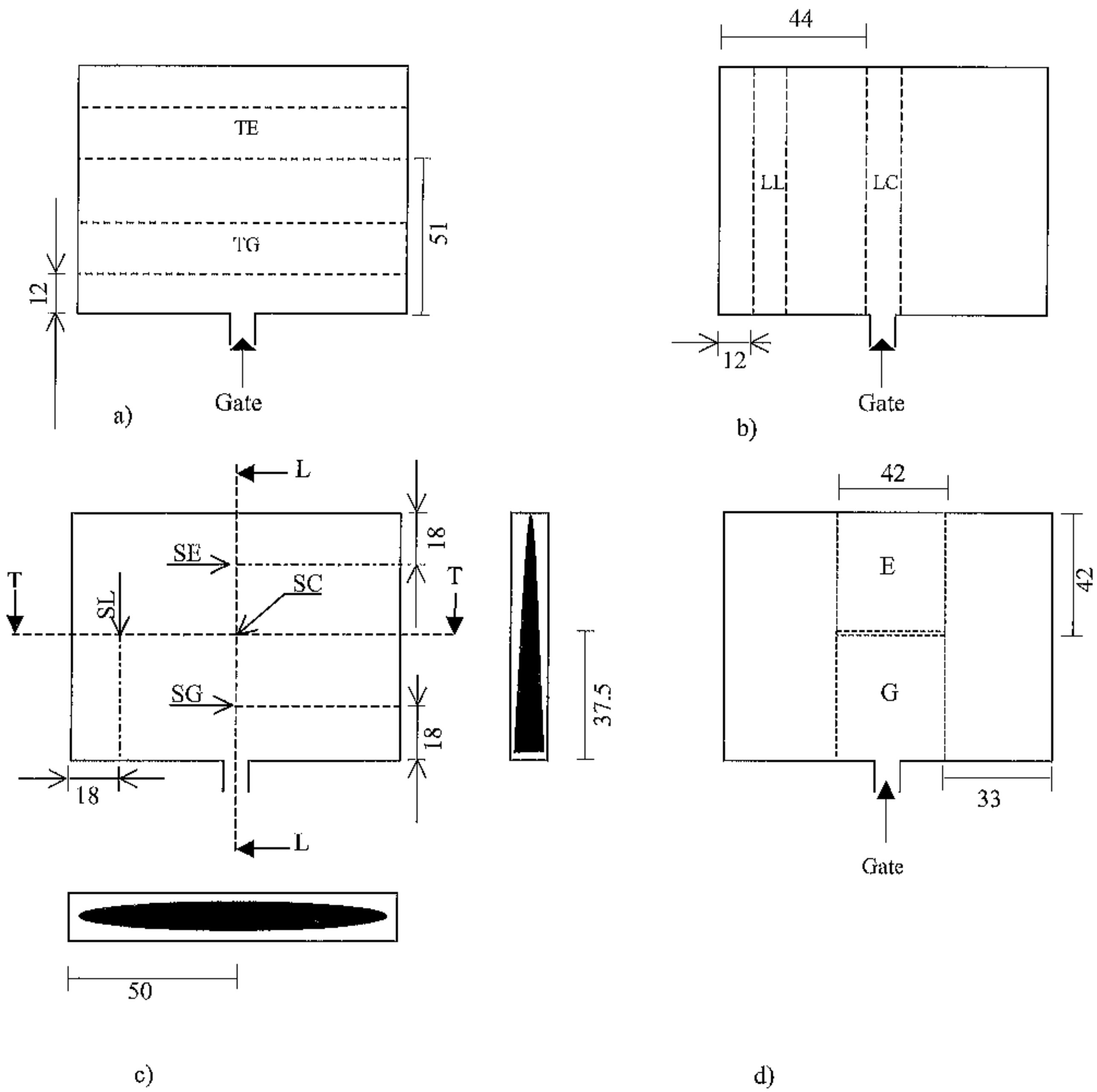


Fig.3. Specimen cut from molded plaques for: a) transverse flexural tests (TG and TE), b) longitudinal flexural tests (LL and LC), c) skin thickness measurements at position SG, SE, SC and SL, d) impact tests (G and E).
All dimensions are in mm

Ait-Messaoud, Fig. 3

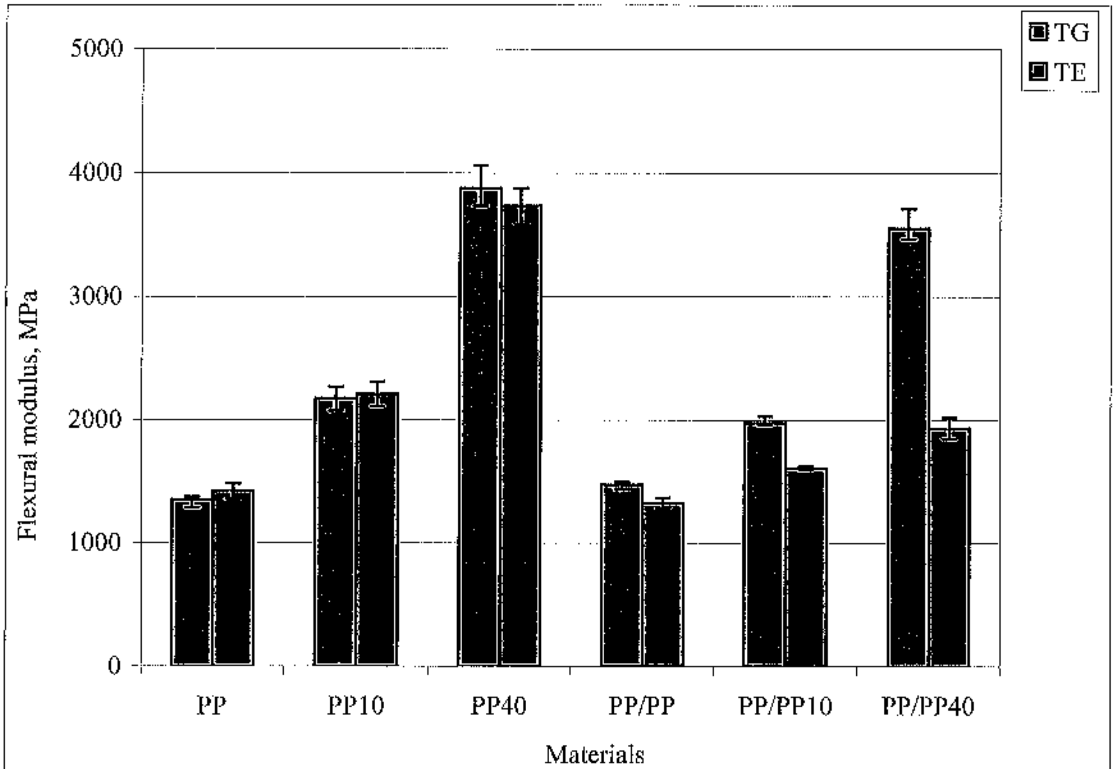


Fig. 4. Transverse flexural modulus (PP as skin)

Ait-Messaoud, Fig. 4

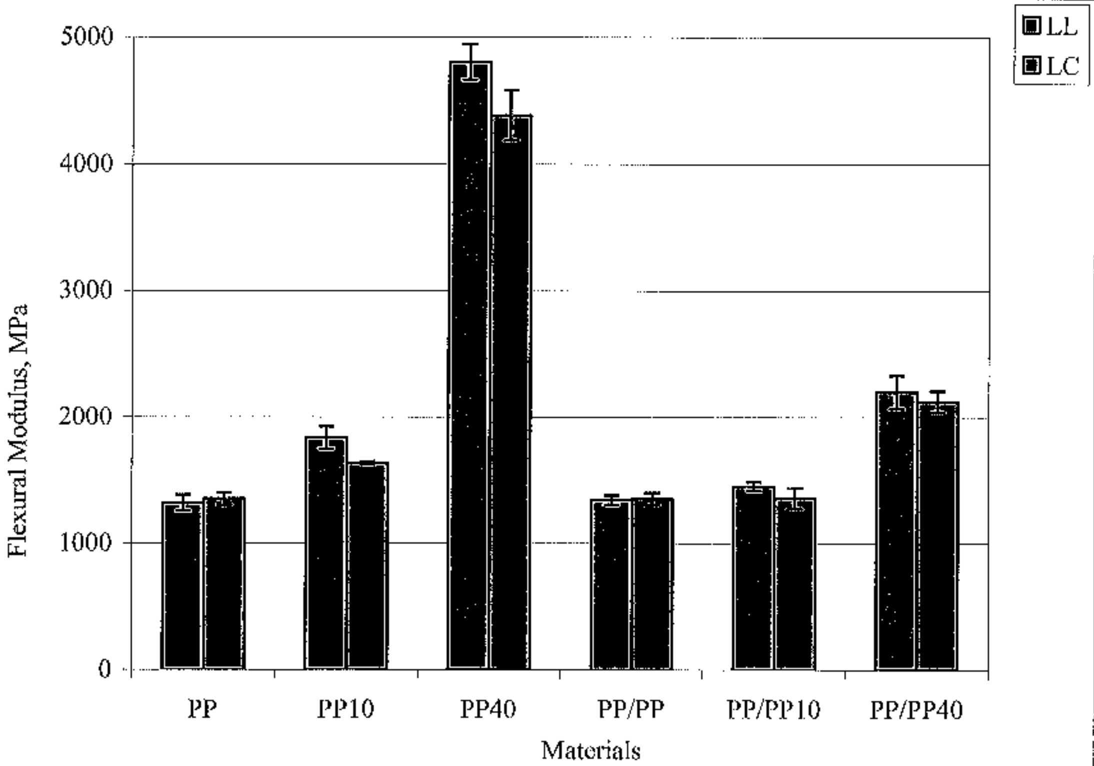


Fig. 5. Longitudinal flexural modulus (PP as skin)

Ait-Messaoud, Fig. 5

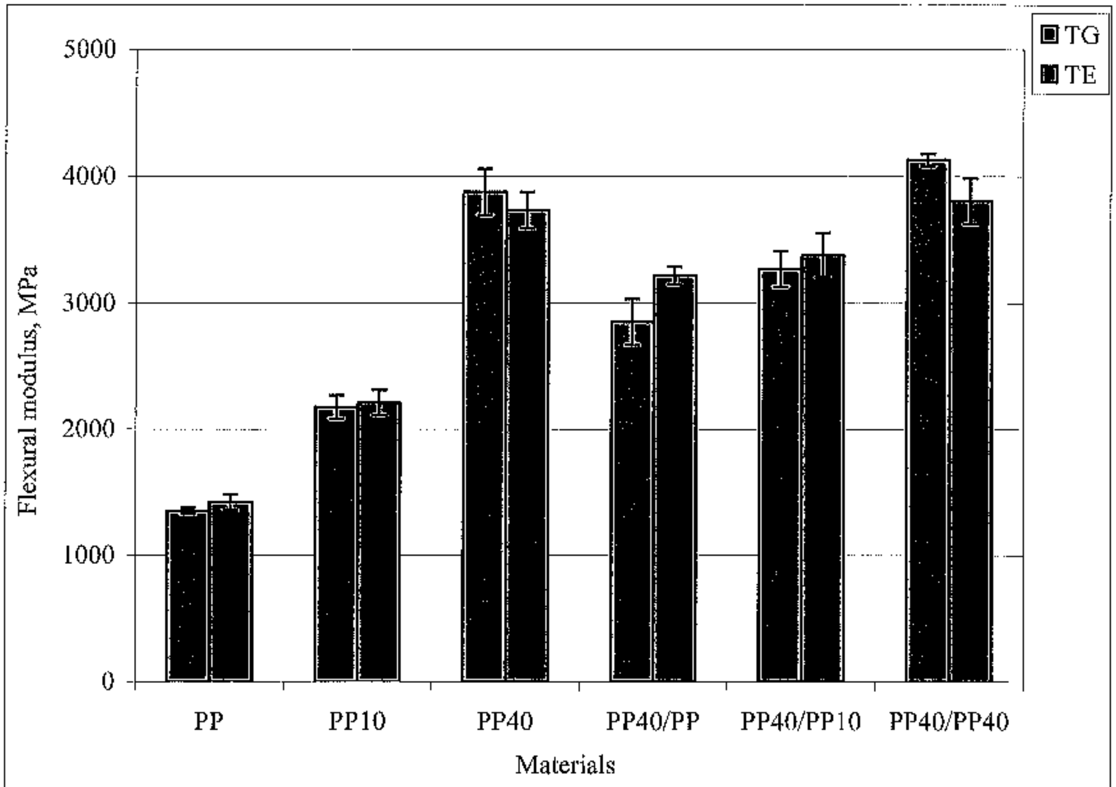


Fig. 6. Transverse flexural modulus(PP40 as skin)

Ait-Messaoud, Fig. 6

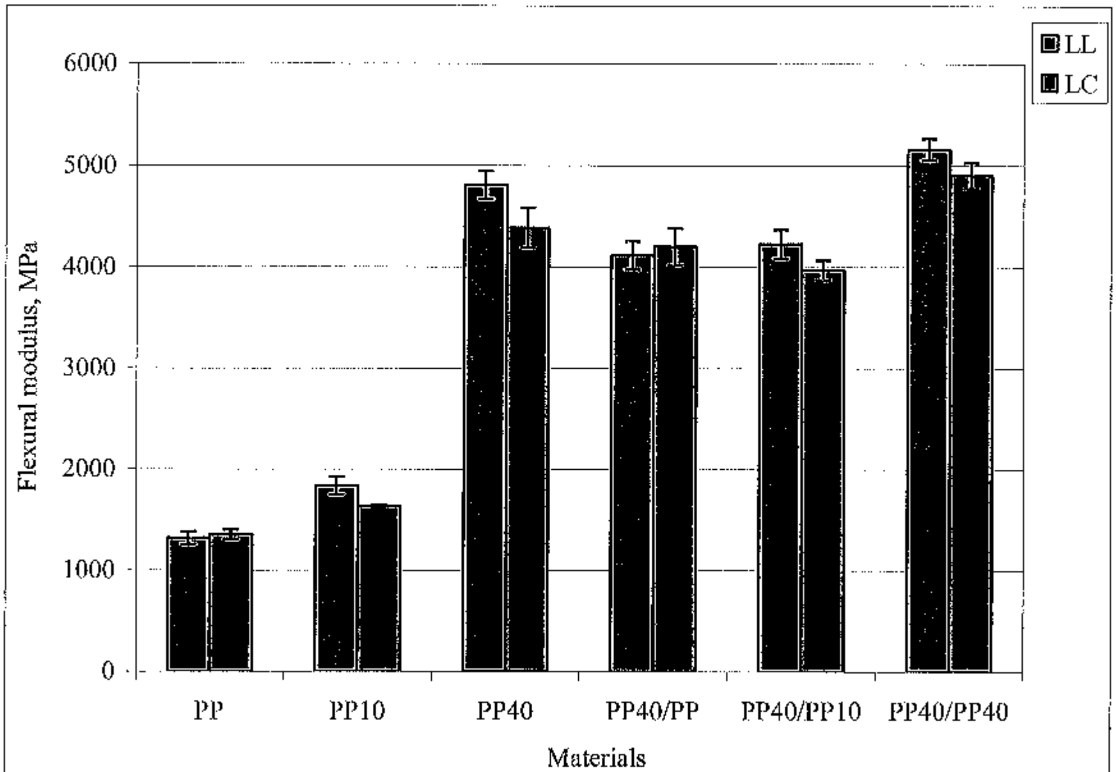


Fig. 7. Longitudinal flexural modulus (PP40 as skin)

Ait-Messaoud, Fig. 7

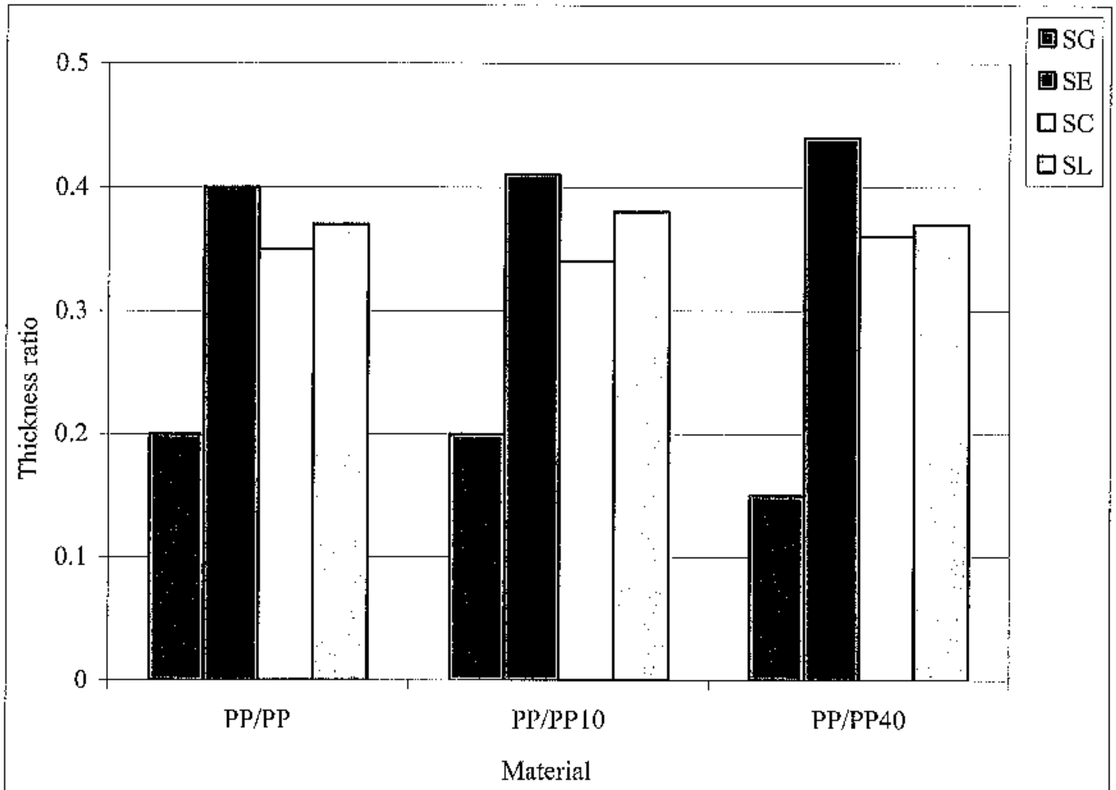


Fig. 8. Skin to part thickness ratio (PP as skin)

Ait-Messaoud, Fig. 8

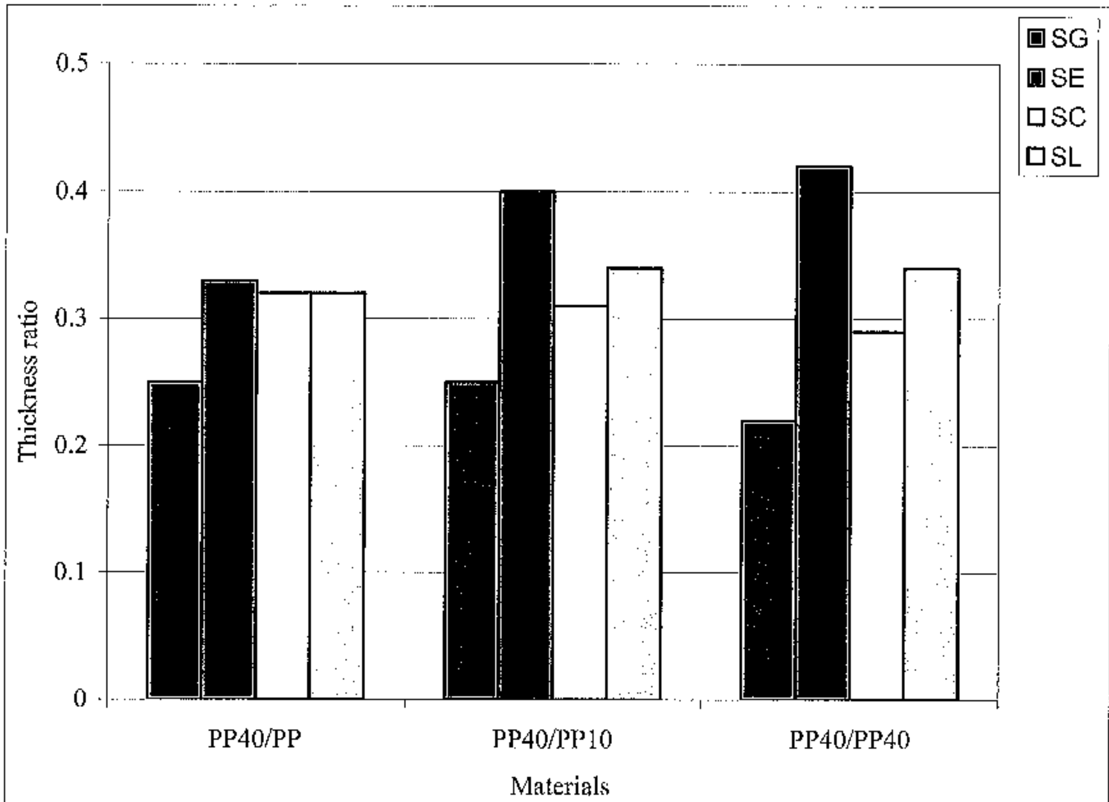


Fig. 9. Skin to part thickness (PP40 as skin)

Ait-Messaoud, Fig. 9

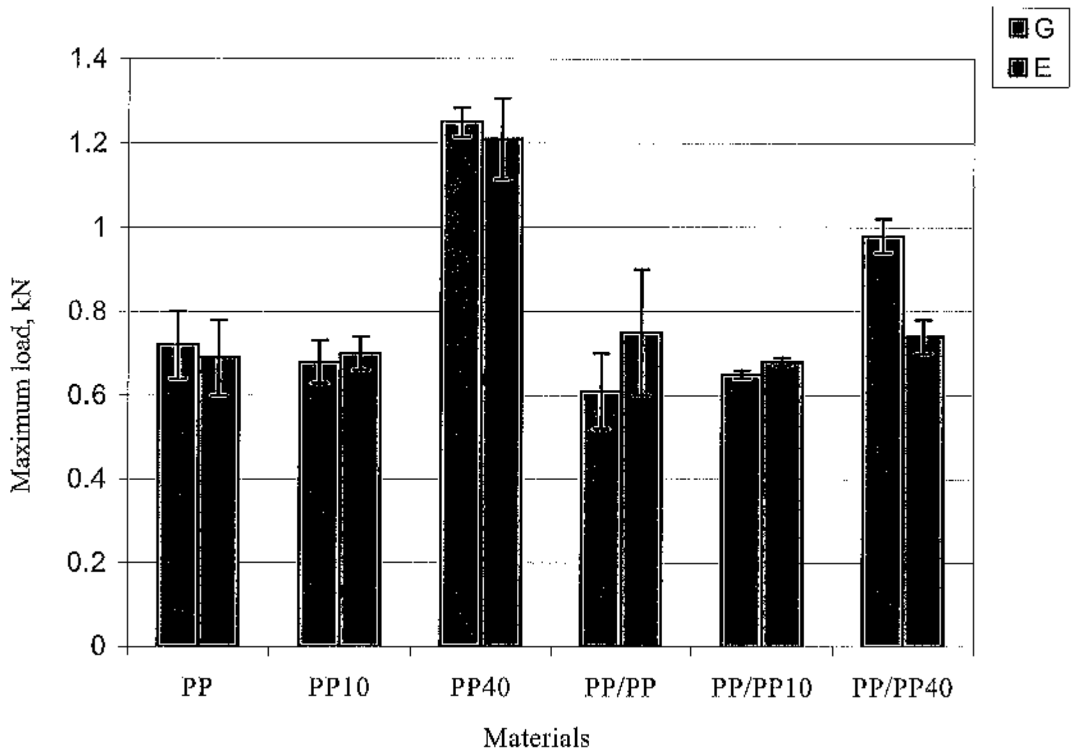


Fig. 10. Maximum load versus materials (PP as skin)

Ait-Messaoud, Fig. 10

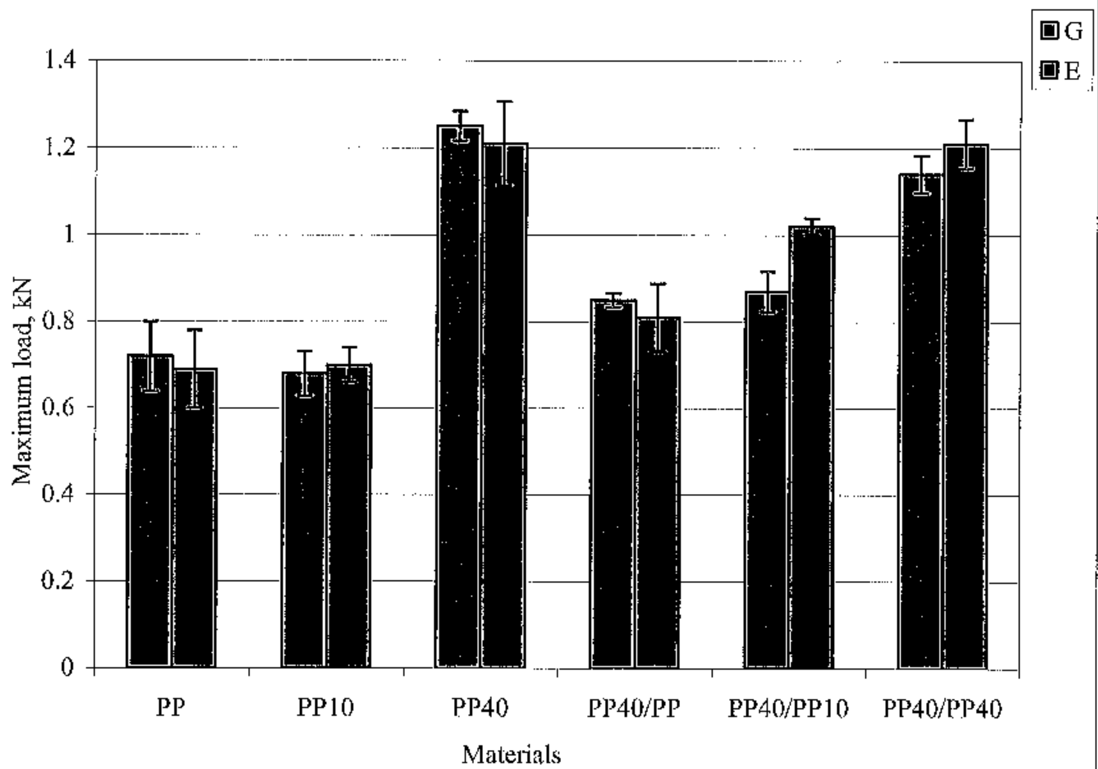


Fig. 11. Maximum load versus materials (PP40 as skin)

Ait-Messaoud, Fig. 11

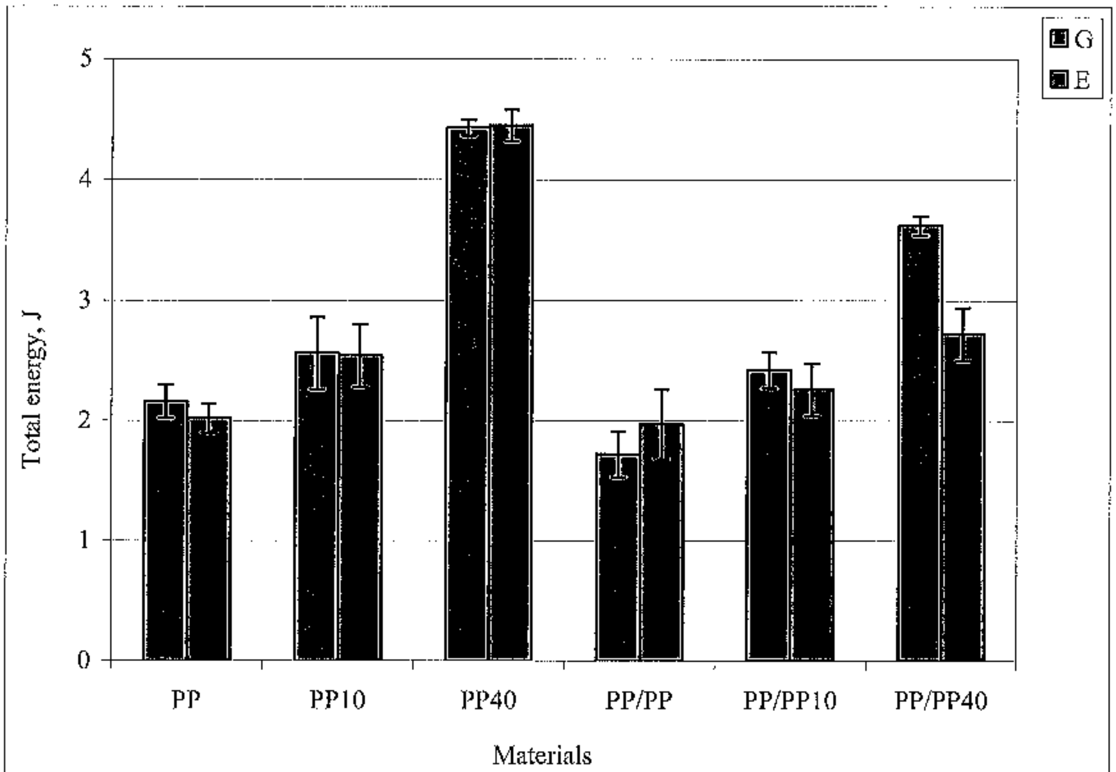


Fig. 12. Total energy versus material (PP as skin)

Ait-Messaoud, Fig. 12

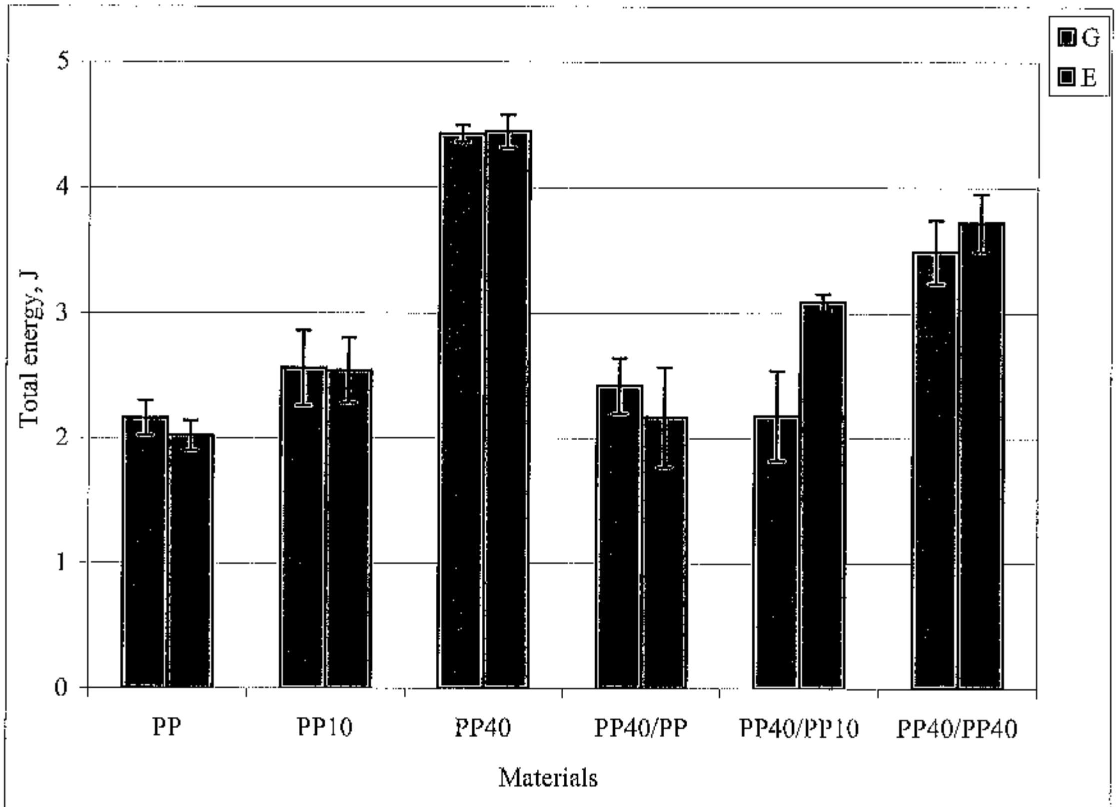
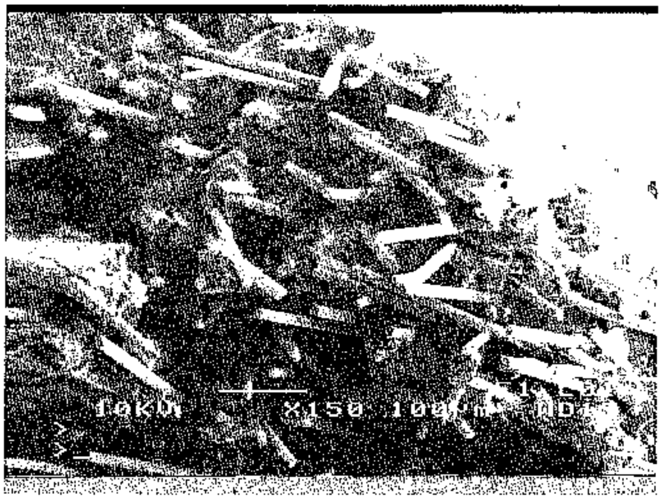
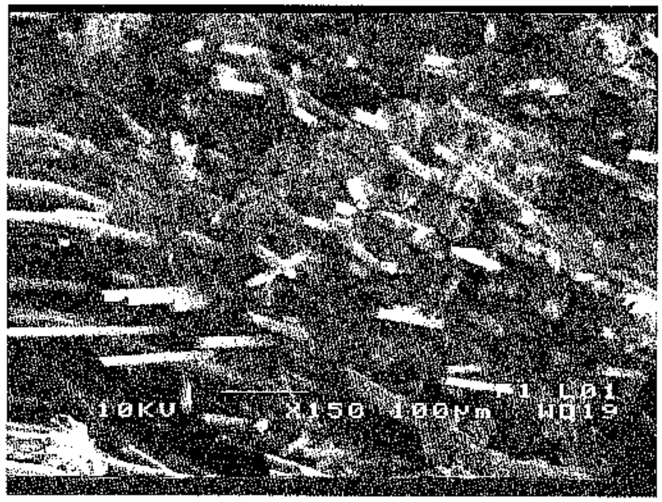


Fig. 13. Total energy versus materials (PP40 as skin)

Ait-Messaoud, Fig. 13



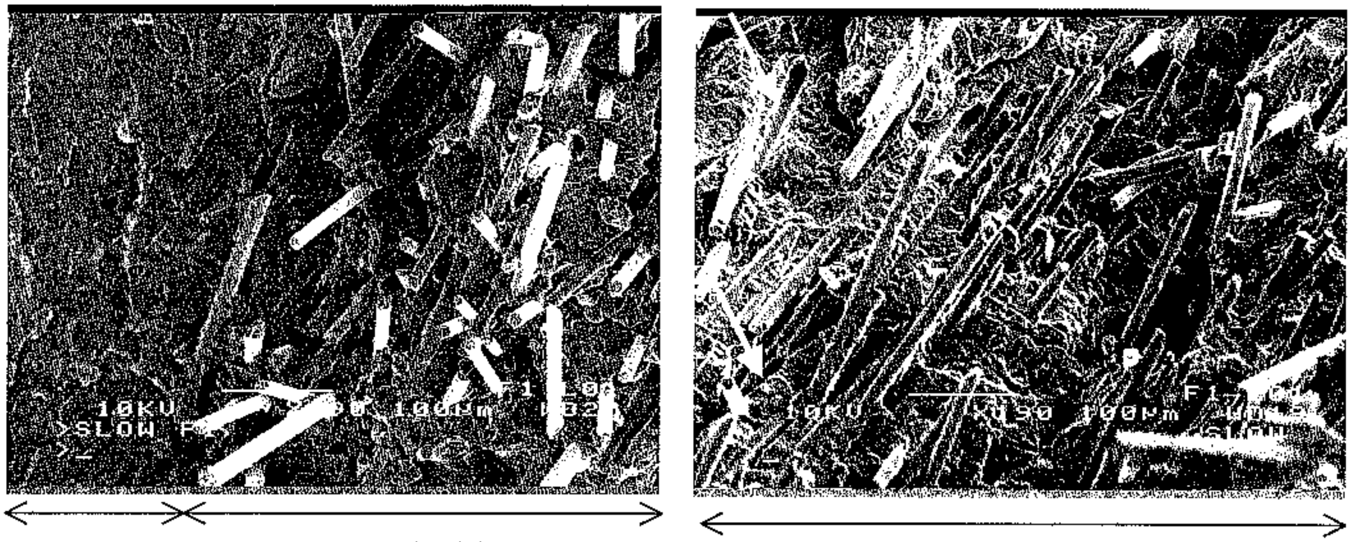
a)



b)

Fig.14: SEM pictures of specimen PP40 taken from sample LC. a) Skin region, b) Core region

Ait-Messaoud, Fig. 14



Skin (PP) Core (PP40)

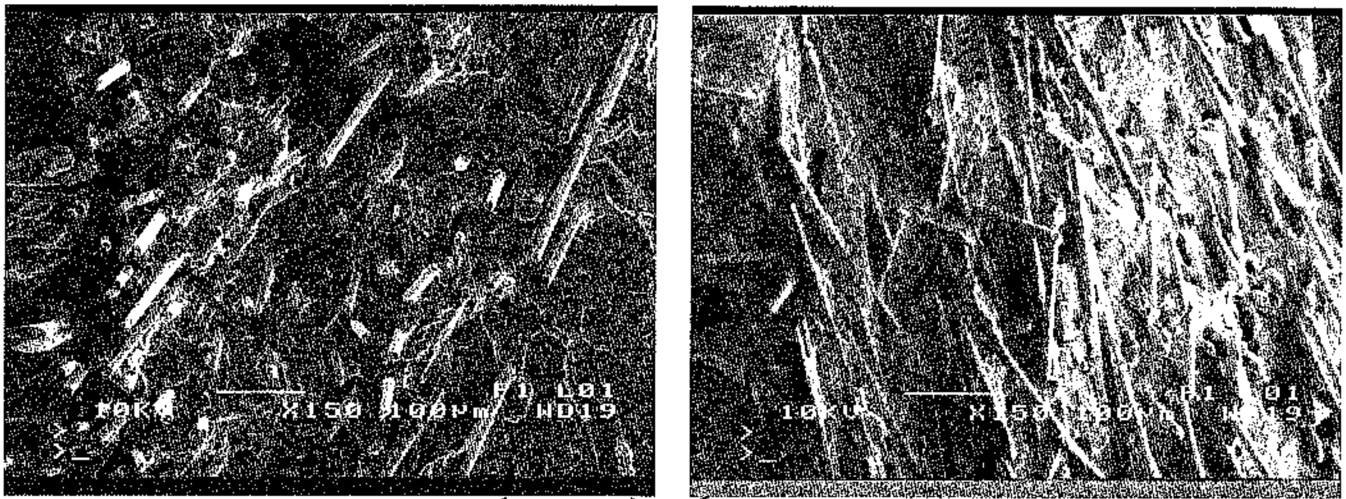
a)

Central region of PP/PP40

b)

Fig.15: SEM pictures of specimen PP/PP40 taken from sample TG broken at the center,
a) Interface skin/core, b) Central region of the core

Ait-Messaoud, Fig. 15



Skin (PP)   Central region of the core (PP40)

a)   b)

*Fig.16: SEM pictures taken from sample LC broken at 18 mm from the end of the part PP/PP40.
a) Interface skin/core, b) Central region of the core.*

Ait-Messaoud, Fig. 16