



## HYBRID INJECTION MOULDING: OVERMOULDING OF METAL INSERTS WITH PP

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### KEYWORDS

Hybrid injection moulding; metal overmoulding; adhesion; microstructure; degree of crystallinity.

### ABSTRACT

Hybrid injection moulding (HIM) is an advanced injection moulding technology that uses dissimilar materials to produce complex parts. In this work, pre-treated metal inserts were overmoulded with semicrystalline polypropylene. The effect of the injection moulding conditions and inserts temperature and roughness on the adhesion of both materials was studied. The effect of a simplified flame treatment on the metal surface modification and wettability was characterized using microscopy analysis and contact angle measurements. The crystalline structure of the overmoulded polymer was also evaluated by microscopy analysis. For the adhesion assessments, single lap shear tests were conducted at room temperature to measure the shear stress at the interfaces. It is shown that the temperature of the insert is one of the main variables affecting adhesion, even more than insert roughness and flame treatment.

### INTRODUCTION

Hybrid injection moulding technology allow the manufacturing by injection moulding of a unified product with components made of plastic and other material(s) (such as, metals, ceramics or even other plastic). This enables the production of rather complex highly load-resistant and low-cost multi-component parts. In these multi-component parts, the properties of different materials can be combined in a synergetic way: metals give high modulus of elasticity and ductile failure; glass provide transparency and rigidity; polymers allow for low weight, in-mould colour and good surface finishing (visible surface), complex design and high integration capabilities.

HIM uses in-mould insert technology to produce complex parts, where one component (metal, glass or polymer) is placed in the moulding tool and plastic is overmoulded on it. Hybrid mouldings feature increased

design efficiency, improved mechanical performance at low weight, high functional integration, reducing assembly steps, all leading to cost reduction.

HIM is a recent technique with a high potential, especially for automotive components. However, the use of such dissimilar materials poses several technology challenges. The imposition of geometric constrains during polymer solidification leads to differential shrinkage and warpage problems as well as significant residual stresses at the interface. The mechanical behaviour of the multi-component parts is dependent upon the properties of both materials. A synergetic combination between physico-chemical interactions and mechanical locking can lead to improve mechanical performance. Furthermore, the manipulation of interface/interphase morphology during processing is also important (e.g., improved adhesion, reduced residual stresses, enhanced mechanical behaviour).

Various factors are of decisive importance for bonding two materials, the type of surfaces (structure of the materials to be bonded), the wetting characteristics of the surfaces, the surface treatment (e.g., roughness) and the design of the joint (e.g. stress distribution)

Most polymers are hydrophobic with a low surface energy. They are therefore difficult to bond to other materials. Adhesion is promoted by surface modification that reduces the interfacial tension and increases interfacial area. Adhesion is also favoured for (mechanically or chemically) roughened surfaces due to the enlargement of the effective contact surface and the increase in the number of active centres. Processing conditions play also a relevant role in adhesion as they constrain the development of the interphase morphology. For example, it has been reported that crystallinity of a semicrystalline polymer has an effect on its adhesive properties (Boucher et al. 1996) and also on the contact angle (Pu and Severtson 2009).

In the present work, we study the adhesion development when a metal insert is overmoulded with a random semicrystalline polypropylene. Both the mould and melt temperatures were varied as well as the roughness and initial temperature of the insert. The insert was previously subjected to a simplified flame treatment. The surface of the metal inserts was investigated using

microscopy techniques and contact angle measurements. Simple mechanical tests were conducted at room temperature to evaluate the shear stress at materials interface, thus allowing inferring about the adhesion. The thermoplastic metal interfaces were analyzed by microscopy techniques for microstructure evaluation.

## EXPERIMENTAL

### Samples Preparation

Since there was no significant adhesion observable in samples with any surface treatment, the metal inserts were subjected to a simplified flame treatment. This is a method for chemical changing of the surface molecular structure of a substrate in a controlled manner increasing its surface energy and wettability. In this case, a simplified method was employed as there was no control of variables such as air to gas ratio, air and gas flow rates or distance between the tip of the flame and the object to be treated. The only controlled variable was the treatment time for reproducible results.

Metal inserts were used in three different variants. In the first case, the metal insert was preheated on a hot plate at 350°C for 30min and instantly inserted in the mould. In the second case, inserts were passed under a burner fed with propane gas, for 60sec and also instantly inserted into the mould. The last treatment followed the same procedure as the second, but the metal inserts were placed inside the mould cavity at ambient temperature.

The single lap shear specimens were produced by overmoulding the metal inserts on a standard cavity for tensile test specimens (according to EN ISO 527) using a 50Ton Engel injection moulding machine. In Figure 1 is depicted an image of a specimen, with the polymer overmoulded on the metal insert. A metal insert (90x10mm) was placed inside the mould cavity sealing the passage of melt material beyond the contact area (20x10mm). The polymer layer thickness in that zone is 2mm. The materials used to produce the specimens were standard steel and homopolymer polypropylene. The injection pressure used was 50bar and the holding pressure 40bar. Experimental conditions for all samples prepared are summarized in Table 1.

Table 1 - Experimental conditions.

Melt Temperature (°C)	Mould Temperature
220	40
220	60
260	60

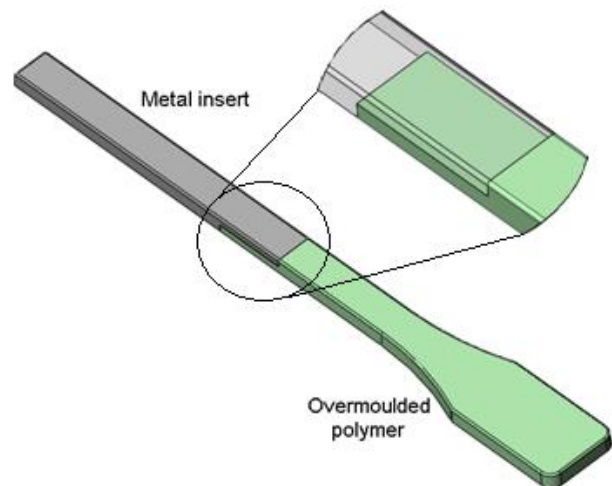


Figure 1: Single Lap Shear Specimen.

### Microstructural Analysis

Scanning electron microscopy (SEM) was performed by a scanning electron microscope (Nano SEM – FEI Nova 200), operating at 10kv - 15kv, to analyze the surface of the inorganic pre-treated inserts. Two magnifications were used (170x and 500x) for observing the overall topography of the samples and higher resolution for more detail, respectively.

An energy dispersive X-Ray analysis (EDX) was also performed by a X-Ray microanalysis integrated system to the SEM (Pegasus X4M) in order to assess the elements presented at the surface subsequent to the flame treatment.

In order to assess the morphology development and crystalline structure of the investigated overmoulded specimens, optical microscopy (OM) was performed using a Polarized Light microscope Olympus BH-2 UMA with a digital Leica camera. The 10µm specimens were sliced from the central part of the contact area of the polypropylene specimens, longitudinally to the flow direction, with a microtome (Anglia Scientific). Afterwards specimens were immersed in Canada balsam resin (with the same reflection index of the glass) between two microscopic glasses slides.

### Contact Angle Measurements

Contact angles were measured with deionised water on a Dataphysics OCA15 plus instrument by pendant drop method at room temperature to analyze the wettability of the metal samples prior and subsequent to the flame treatment.

A drop of the deionised water with a volume of 3µL was delivered to the insert surface by a microsyringe pump. The drop volume was selected so that there is no interference from gravity. The procedure was repeated



seven times for each sample giving a standard error for the mean value of approximately 5°. Preliminary surface cleaning was performed using isopropyl alcohol at room temperature. Subsequently, specimens were rinsed with distilled water.

### Morphological Characterization

The density of the overmoulded polypropylene samples was measured using a precision analytical balance equipped with SCALTEC equipment, in order to determine the crystallinity degree of the samples moulded with different conditions. The test specimens were cut from the contact area of the specimens.

The SCALTEC equipment consists of a support and a suspender properly adjust for the needs of measuring density, higher and lower, then a reference liquid. The reference liquid used was propanol (0.7881 g/cm<sup>3</sup>).

The test followed the ASTM D 792 standard using method A. The degree of crystallinity was calculated by

$$X_c = \frac{1/\rho_s - 1/\rho_a}{1/\rho_c - 1/\rho_a} \times 100 \quad (1)$$

where  $\rho_s$ ,  $\rho_c$  and  $\rho_a$  are, respectively, the densities of the sample, a 100% crystalline material and a 100% amorphous material.  $\rho_c$  and  $\rho_a$  are assumed 0.938g/cm<sup>3</sup> and 0.8534g/cm<sup>3</sup> (Alexander 1969).

### Mechanical Tests

After removal of the test specimens from the mould, the parts were allowed to cool to room temperature and were left to stable during 3 days. The overmoulded specimens were then tested on a Zwick Roell Z05 universal testing machine in a controlled environment (at 23°C and 55% relative humidity) and at a constant crosshead speed of 3mm/min. The test continued until the specimens failed. The shear force–strain data were collected and stored as data files. Every value of shear strength was averaged of the specimens prepared with the same conditions and the standard error was calculated.

## RESULTS AND DISCUSSION

### Scanning Electron Microscopy with Energy Dispersive X-Ray Analysis

The surface of the treated inorganic inserts was observed by SEM and is shown in Figures 2 and 3. A comparison between the surface prior and after the

flame treatment is shown in Figure 2. The topography of the sample surface does not change significantly. However, it appears that the surface of the sample subjected to the flame treatment is covered by an oxide layer. This is better observed in the atomic contrast image (see Figure 3) and is confirmed by the EDX analysis (Figure 4).

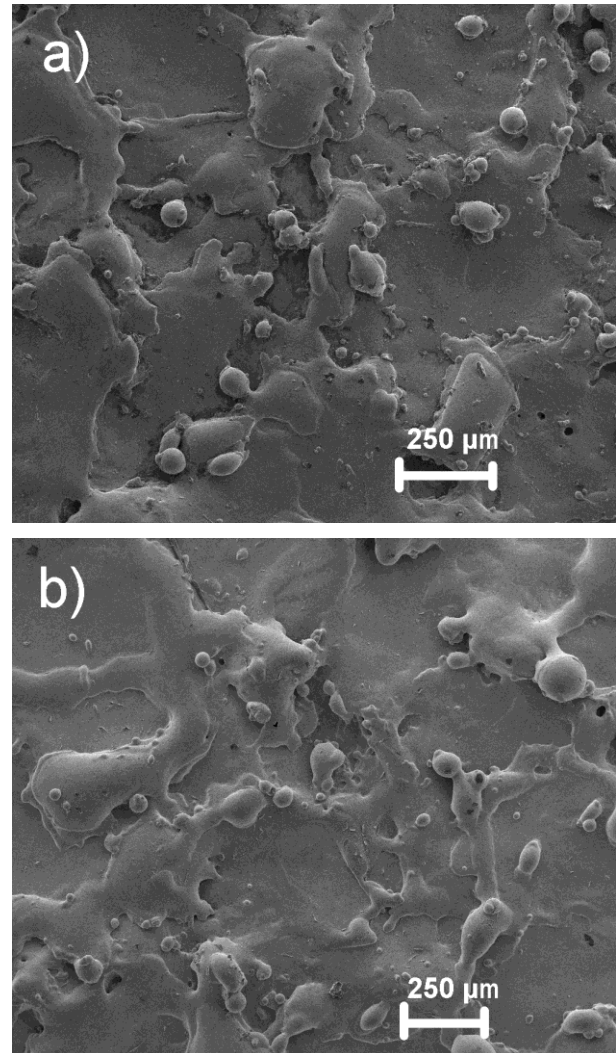


Figure 2: SEM micrographs of the metal samples. a) without treatment; b) with flame treatment.

In Figure 3a) (sample without treatment) the lighter areas correspond to Iron (Fe). The darker areas correspond to elements with lower atomic number (C, O, etc.). In the samples with flame treatment (Figure 3b), almost the entire area is covered by an oxide layer. This can also be observed in the EDX plot in Figure 4, where an additional oxygen element is presented at the surface of the insert.

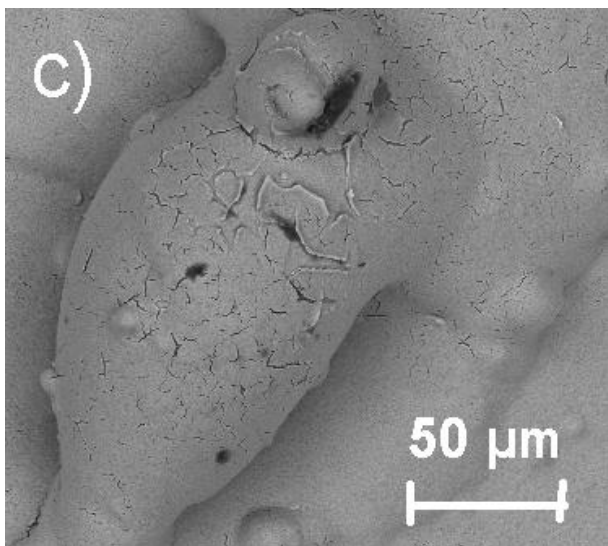
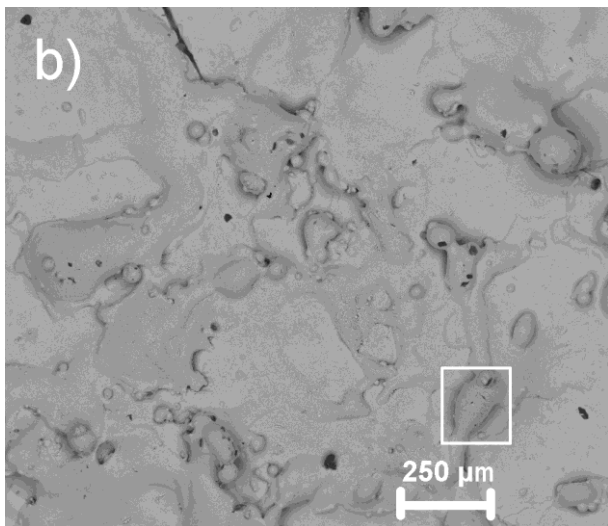
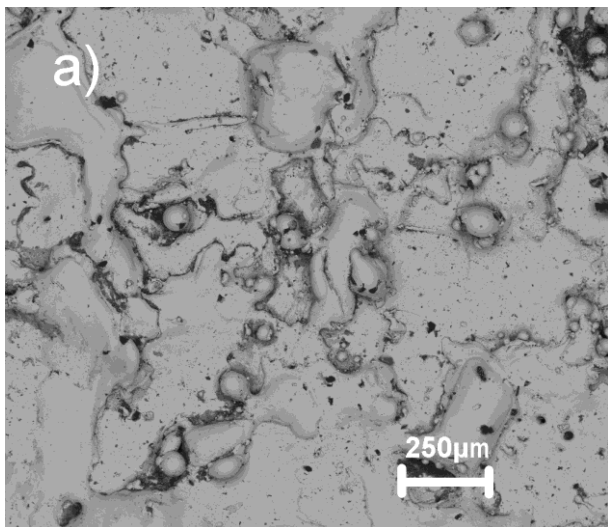


Figure 3: SEM micrographs of the metal samples showing the atomic contrast. a) Without treatment (magnification 170x, 15kV); b) with flame treatment (magnification 170x, 15kV); c) larger resolution of the detail marked in b) (magnification 500x, 15kV).

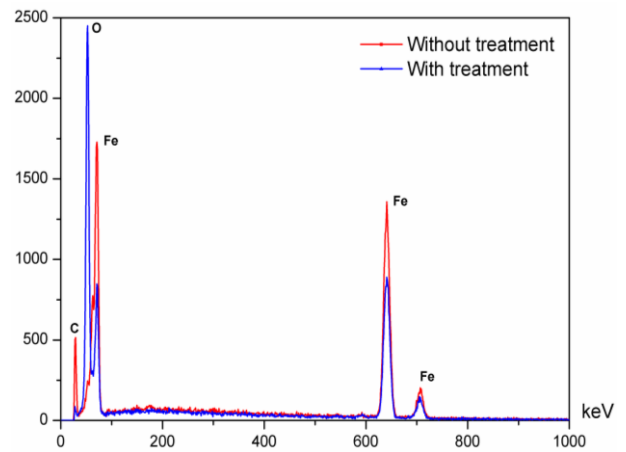


Figure 4: Overlay of both EDX spectrum (with and without flame treatment), for comparison purposes.

### Contact Angle Measurements

The wetting properties of the metal samples were evaluated by contact angle measurements. Figure 5 shows the variation of the water contact angle for samples subjected to the flame treatment in contrast to the ones with no treatment. This graph also shows the influence of the surface roughness in the measurements. The samples subjected to the flame treatment present a much lower contact angle. This reduction in contact angle demonstrated an increase in the surface wettability and surface energy following the flame treatment due to the additional polar functional groups, as was expected (Song et al. 2007).

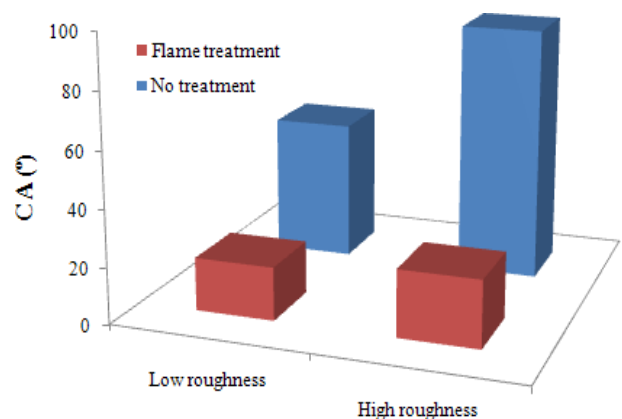


Figure 5: Evaluation of roughness influence in water contact angle measured in samples before and after flame treatment.

The sample with low roughness presents a reduction of, approximately, 63% of the contact angle. However, this reduction is even more prone in the ones with higher roughness. In this case the reduction is almost 74%.

### Microstructure Analysis

The microstructure of an injection moulded semicrystalline material always shows a heterogeneous/hierarchical structure due to the thermomechanical environment of the injection moulding process (Viana et al. 2002; Viana et al. 2004; Viana 2005; Todorov and Viana 2007; Ghosh et al. 2007). A typical microstructure of an injection moulded semicrystalline polymer, as observed by polarized light microscopy, is depicted in Figure 6. Near the mould walls, a skin layer of oriented chains in the flow direction is formed, due to high strain-rates and high cooling rates. The core region, where there is almost no orientation and the cooling is much slower, is composed of spherulite-like structures.

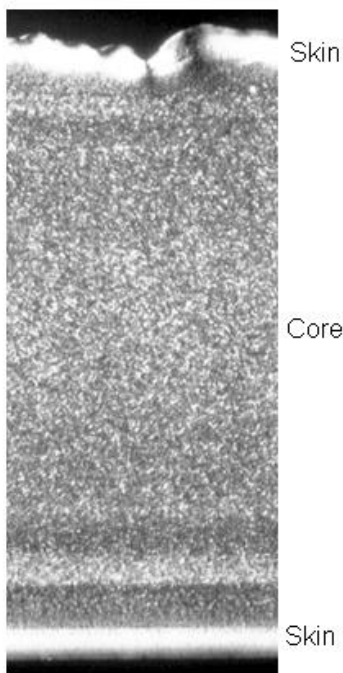


Figure 6 – Microstructure of an injection moulded semicrystalline polymer.

Figure 7 shows a cross section of the polypropylene overmoulded at 260°C and mould temperature of 60°C onto flame treated metal samples with cold and hot sample. The upper side of the sample is the one in contact with the insert and the lower is the one in contact with the mould. Figure 7a) shows a typical laminated skin-core microstructure. The regions of the sample in contact with the cold mould walls and insert (upper and lower sides of the sample) present a skin layer of highly oriented molecular chains in the flow direction, due to the high strain rates and rapid cooling. The core region is composed by spherulite-like structures, due to the extremely low shear deformation and slow cooling. In Figure 7b) the skin layer is only presented in the mould side of the specimen. The

spherulite structure of the core region is observed at the insert side of the specimen as well. This fact is due to the much slower cooling of the material in contact with the hot insert surface.

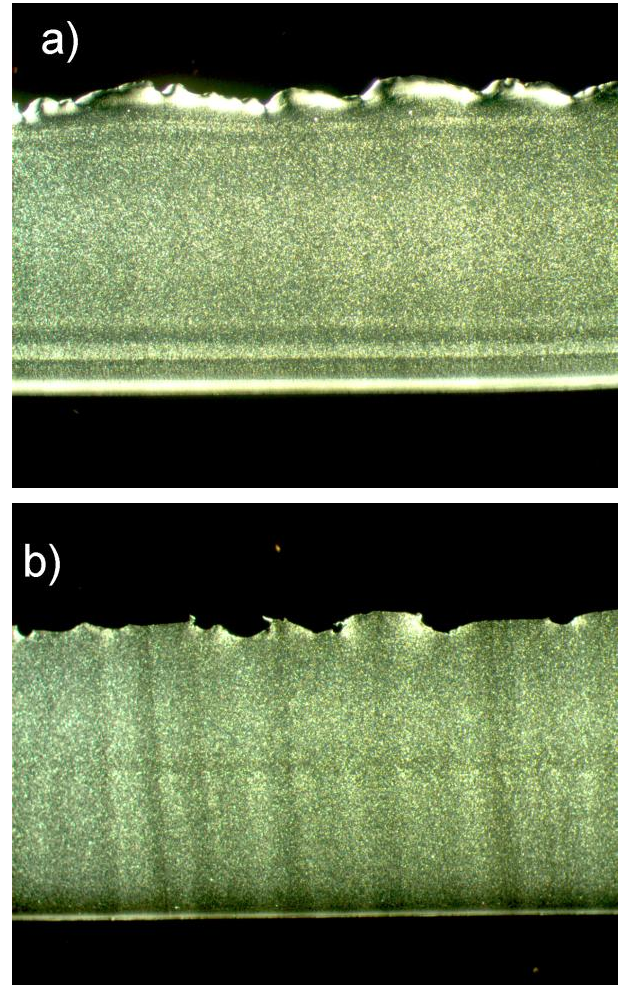


Figure 7: Optical microscopy of the polypropylene overmoulded at 260°C and mould temperature of 60°C, onto flame treated metal sample: a) cold insert and b) hot insert.

### Mechanical Test

Figures 8, 9 and 10 show plots of shear force against strain curves, depicting the mould and melt temperature influence on shear force. From these plots one can conclude that the flame treatment contributed to an increase in the adhesion. Furthermore, the hot treated samples exhibit higher shear force values than the cold samples, as would be expected.

The pre-heated samples were the ones presenting higher shear force values, even higher than flame treated samples.

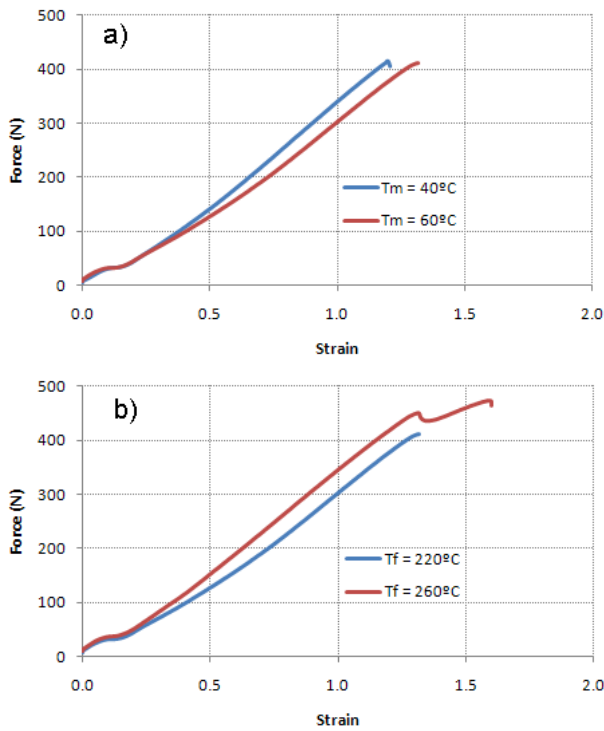


Figure 8: Plot of shear force against strain for pre-heated samples. a) Mould temperature influence and b) melt temperature influence.

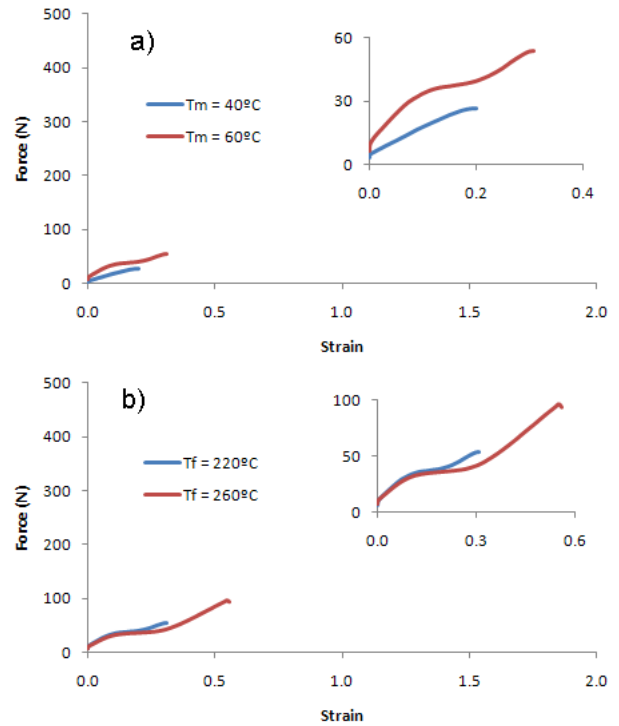


Figure 10: Plot of shear force against strain for flame treated cold samples. a) Mould temperature influence and b) melt temperature influence.

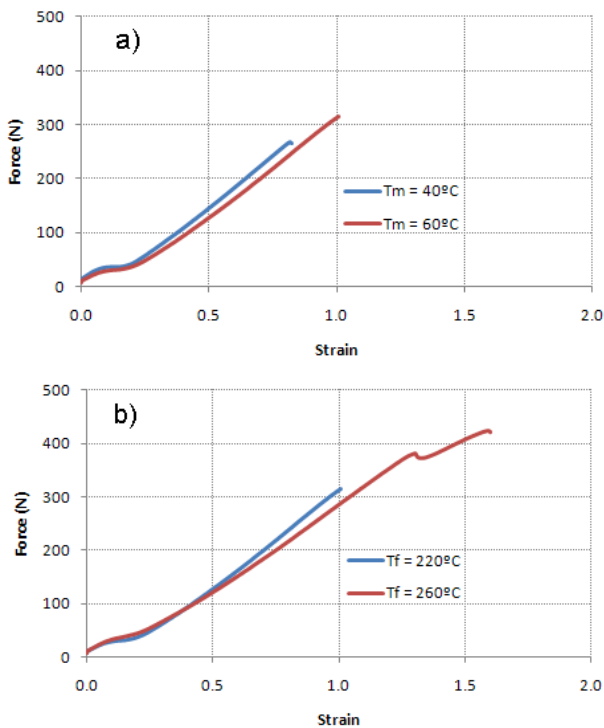


Figure 9: Plot of shear force against strain for flame treated hot samples. a) Mould temperature influence and b) melt temperature influence.

In Figure 11 is shown the metal insert roughness influence on the shear force. As would be expected, the insert with higher roughness presents a much higher bonding to the overmoulded polymer. Moreover, the samples moulded with higher melt and mould temperatures also present higher shear force (see Figure 12). This has been observed before (Ramani and Moriarty 1998) and is due to the subsequent lower viscosity of the overmoulded polymer, which enables a better wetting of the insert and, hence, a better replication of the rough surface.

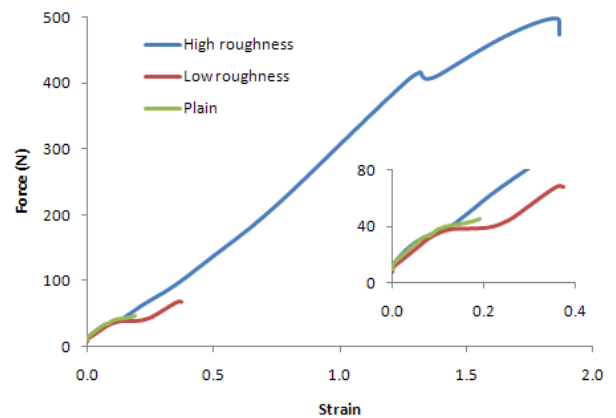


Figure 11: Plot of shear force against strain for samples presenting best shear results (pre-heated samples; mould temperature  $60^\circ\text{C}$  and melt temperature  $260^\circ\text{C}$ ).

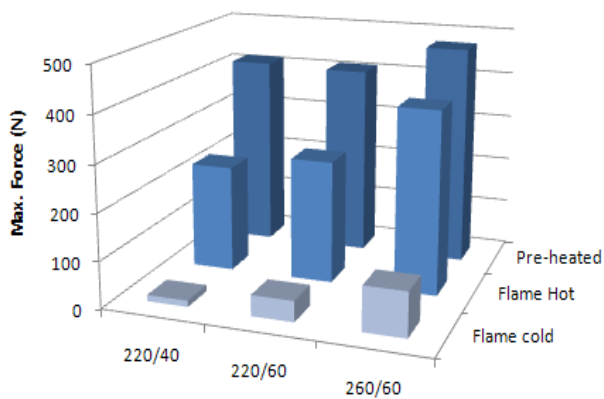


Figure 12: Processing conditions and pre-treatment influence on the maximum shear force.

### Morphological Characterization

Figure 13 shows the variation of the degree of crystallinity for the overmoulded PP material on the pre-treated metal surfaces with the mould temperature. As would be expected, the degree of crystallinity is higher for the hot plate samples because the metal insert surface is at a much higher temperature than the mould surface. Hence, the cooling rate is much lower. For the same reason, the hot sample (flame treatment) presents a higher degree of crystallinity than the cold sample. The specimens obtained by overmoulding the pre-heated metal inserts, besides being the ones presenting higher degree of crystallinity, are also the ones with better bonding results. Crystallinity seems this way to have some influence on adhesion.

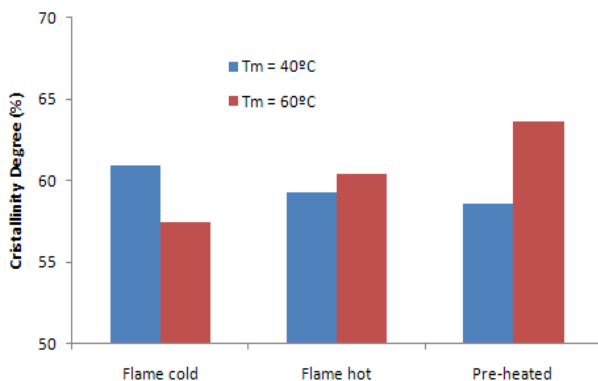


Figure 13: Effect of mould temperature and pre-treatment on the overmoulded material degree of crystallinity.

### CONCLUSIONS

This experimental work has demonstrated that raising the metal insert initial temperature above the melt temperature of the overmoulded polymer produced a

spherulite-like microstructure at the region of the sample in contact with the insert. Furthermore, the adhesion in this case has been increased significantly. This is due to the increase in the degree of crystallinity, but also to the increase in the ability of the material to wet the insert surface due to its slower cooling. This slower cooling allows the material to better replicate the insert surface roughness, increasing the mechanical interlocking. The flame treatment also increased the adhesion of the overmoulded samples, but the process alone did not produce the desired difference when compared to the samples with pre-heating.

### REFERENCES

- Alexander, L.E. 1969. *X-Ray Diffraction Methods in Polymer Science*, Willey-Interscience, New York.
- Boucher, E.; J.P. Folkers, H. Hervet; L. Leger; and C. Creton. 1996. "Effects of the Formation of Copolymer on the Interfacial Adhesion between Semicrystalline Polymers" *Macromolecules* 29 No.2, 774-782
- Ghosh, S.; J.C. Viana; R.L. Reis; and J.F. Mano. 2007. "Effect of Processing Conditions on Morphology and Mechanical Properties of Injection-molded Poly(L-lactic acid)" *Polymer Engineering and Science* 47 No.7, 1141-1147
- Pu, G. and S.J. Severtson. 2009. "Dependency of Contact Angle Hysteresis on Crystallinity for n-Alkane Substrates" *The Journal of Physical Chemistry C* 113 No.16, 6673-6680
- Ramani, K.; and B. Moriarty. 1998. "Thermoplastic Bonding to Metals via Injection Molding for Macro-composite Manufacture" *Polymer Engineering and Science* 38 No.5, 870-877
- Song, J.; U. Gunst; H.F. Arlinghaus; and G.J. Vancso. 2007. "Flame Treatment of Low-density Polyethylene: Surface Chemistry Across the Length Scales" *Applied Surface Science* 253, 9489-9499
- Todorov, L.V.; and J.C. Viana. 2007. "Characterization of PET Nanocomposites Produced by Different Melt-based Production Methods" *Journal of Applied Polymer Science* 106 No.3, 1659-1669
- Viana, J.C.; A.M. Cunha; and N. Billon. 2002. "The Thermomechanical Environment and the Microstructure of an Injection Moulded Polypropylene Copolymer" *Polymer*, 43 No.15, 4185-4196
- Viana, J.C.; N. Billon; and A.M. Cunha. 2004. "The Thermomechanical Environment and the Mechanical Properties of Injection Moldings" *Polymer Engineering and Science* 44 No.8, 1522-1533
- Viana, J.C. 2005. "Structural Interpretation of the Strain-rate, Temperature and Morphology Dependence of the Yield Stress of Injection Molded Semicrystalline Polymers" *Polymer* 46 No.25, 11773-11785



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