

# Rotational molding of plasma treated polyethylene/short glass fiber composites

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*Abstract. Rotational molding is a manufacturing technique for producing 3D hollow parts by adding plastic powder to a shell-like mold and rotating the mold while heating it with the powder. In contrary to the injection molding all the process operates at atmospheric pressure which makes reinforcement of the rotomolded product difficult. In this paper plasma treated PE and short glass fibers were dry mixed and used to produce composites by rotational molding process and characterized in terms of morphology and mechanical properties.*

## Introduction

Rotational molding is a manufacturing technique developed in 1940 for producing 3D hollow parts by adding plastic powder to a shell-like mold and rotating the mold while heating it with the powder. Rotational molding process is shown schematically in Figure 1.

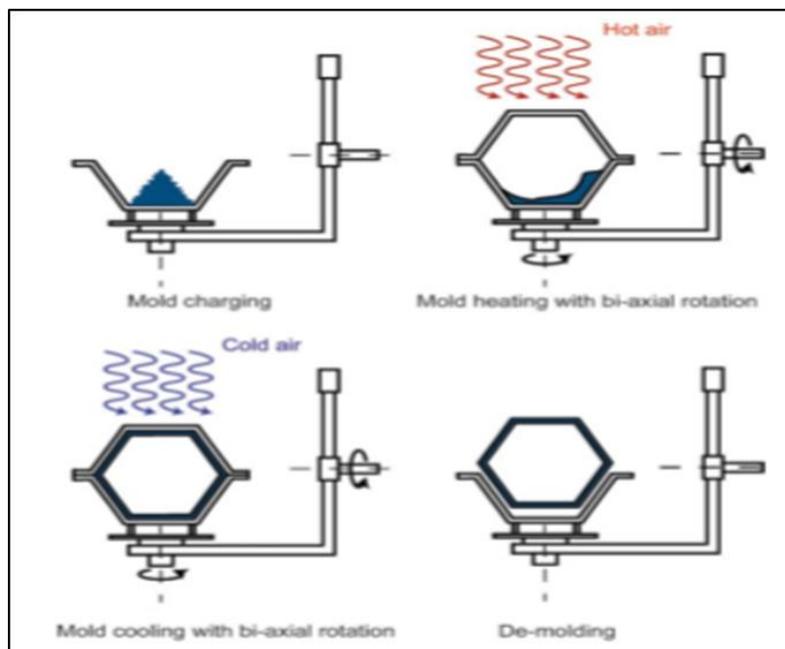


Figure 1. Rotational molding process

In the first process step, a hollow mold is filled with powdery or liquid plastics. The filled and closed mold starts rotating about two axes. While maintaining the rotation, the mold is heated up in the second process step, often accomplished by convection within an oven. While heating the polymer, it successively forms a homogenous porous layer of polymer melt at the inner mold surface. This porous layer densifies while the polymer is held in the melt state. In the third step, the still rotating mold is cooled down by compressed air or water which is applied to the outer mold surface, causing the polymer to solidify within the mold and to form a solid hollow body. After reaching the demolding temperature, the mold is opened, and the part is demolded in the last process step [1,2,3]. Large volume, hollow parts such as Kayaks, traffic barriers and storage tanks for various purposes with a volume up to 100,000 l are typical applications for rotational molding. In general, seamless hollow bodies with a high degree of design freedom and with very low residual stresses can be produced with this production technique. However, it has its limitations such as the costs of complex geometry form and long processing time. Additionally, the process is limited to selected thermoplastic polymers, predominately to polyethylene and polypropylene. Mechanical properties of these polymers are relatively poor and rotomolded products find applications in the fields where structural requirements are not particularly critical. Therefore, different attempts have been made to overcome the limitations of rotational molding products in terms of mechanical properties. Some of the developed approaches include the incorporation of different types of reinforcement [3,4].

In contrary to the injection molding all rotational molding process operates at atmospheric pressure which makes reinforcement of the rotomolded product difficult. The main problems that arise when using reinforcements in rotational molding relate to the fact that the process is low shear and the mixing and the adhesion of the filler inside the polymeric matrix is not superior which leads to the segregation and the agglomeration of the reinforcement [8].

Glass fibers is one of the most used fibers in reinforcement of polymeric matrix. Only few studies have reported the use of glass fibers in rotational molding. Poor distribution of long glass fibers in the rotomolded parts have been reported because of the high aspect ratio of fibrous reinforcement [5], in contrast the short fibers showed a tendency to improve the mechanical properties of the rotational molding product by modifying the rotomolding process, for example using of inner air pressure, the double shot mold charging process, the chemical modification of the fiber and/or powder or by the melt compounded method [6,7,9].

Cold plasma surface modification has been established as an effective and low-cost technology for surface hydrophilization of polymer materials due to grafting of wide range of reactive species (hydroxyl, carbonyl, carboxyl, ether, amine, peroxides etc.) on the polymer surfaces [9], which improve compatibility between the polymer matrix and natural and/or glass fibers. In our previous studies we investigated the effect of the plasma treatment of polyethylene on the adhesion to the glass fibers and how this adhesion improved mechanical properties of the composite samples prepared by the pressure-free molding process. An improvement up to 73% in the tensile strength was obtained using the plasma treated PE [10].

In this article plasma treated PE and short glass fibers were dry mixed and used to produce composites by rotational molding process and characterized in terms of morphology and mechanical properties.

## 1. Experimentation

### 1.1. Materials

The matrix polymer used is linear low-density Polyethylene “DOWLEXTM 2629UE”, with density 0.935 g/cm<sup>3</sup> and melt flow index (MFI) 190°C/2.16 kg = 7.0 g/10 from Dow Chemical Company, US; Plasma modification of PE powder was processed by Surface Treat, a.s. (Czech Republic). Short milled glass fibres with an average length of 0.19 mm and average diameter of 14µm by LanXESS Company were used as reinforcement.

### 1.2. Sample preparation and mechanical Characterization:

A laboratory scale “rock-and-roll” rotational moulding machine with electrical heating were used to prepare the samples, the machine is designed to undergo rocking action about one axis (rock) and full 360° rotation along perpendicular axis (roll) and. The mould is prism-shaped with dimensions 260x95x95mm made from Aluminum. The samples were prepared by first dry-blending the glass fibers with the powder for 10 minutes to get a homogeneous blend, then the mixture were filled in the mould and the mould were closed and loaded to the cold oven, the oven were heated to 250, the air cooling started when the internal air temperature (PIAT) reached 200, when the demoulding temperature were achieved the finished part were removed from the mould. During heating and cooling mould rotation speed was maintained at 10 rpm, while the oven was rocking by 45 degree. Samples with different fibers content were prepared, and the samples were coded as “UTPE” for the composites produced using untreated Polyethylene and “TPE” for the composites produced using plasma treated polyethylene, composites with 3 deferent content of glass fibers (5wt%,10wt% and 20wt%).



Figure 2. Mould used for rotational moulding and the produced PE sample box

Tensile properties were measured according to ASTM D638 using universal testing machine TINUS OLSEN H50KT at a gauge length of 60 mm and speed of 50 mm per minute. The tensile strength was calculated by dividing the measured tensile force (N) by the cross section of the original sample before deforming (in mm<sup>2</sup>). The impact strength was measured by Charpy Impact Tester and the impact strength was calculated by dividing the absorbed impact energy (J) by sample area at the notch. The specimens prepared according to ASTM D6110. Each value was calculated as an average value of five tested samples produced in the same conditions. Composite samples were cryogenically fractured (liquid nitrogen) and the exposed surfaces were coated with chrome under vacuum. micrographs were taken on a scanning electron microscope.

## 2. Results and discussion

### 2.1. Tensile properties

Tensile strength of the composites is presented in Figure 3, the tensile strength for all the composite produced using untreated polyethylene was lower than unfilled LLDPE, This behavior is related to poor fiber-matrix adhesion limiting interfacial stress transfer, the presence of high amount of the air bubbles in the samples Figure (4-a), It is known that Surface porosity and bubbles within the part result in unfavorable aesthetics, while leading to deterioration in mechanical properties of the plastic moldings. On the other hand, the composites produced using plasma treated polyethylene showed a slight increase up to 10% in the tensile strength at 5%wt and 10%wt of glass fiber content comparing with LLDPE. This improvement can be associated to the formation of adhesive bonds between the OH groups grafted on the PE powder surface and the glass fibers a result of plasma treatment, additionally no bubbles can be noticed in TPE composite Figure (4-b) this could be related to the fact that Plasma treatment may improve the flow ability of the matrix. At 20wt% of glass fiber content the tensile strength dropped by 6.5 % comparing to unfilled material and this is mostly associated with the presence of fiber clumps at higher content of glass fibers.

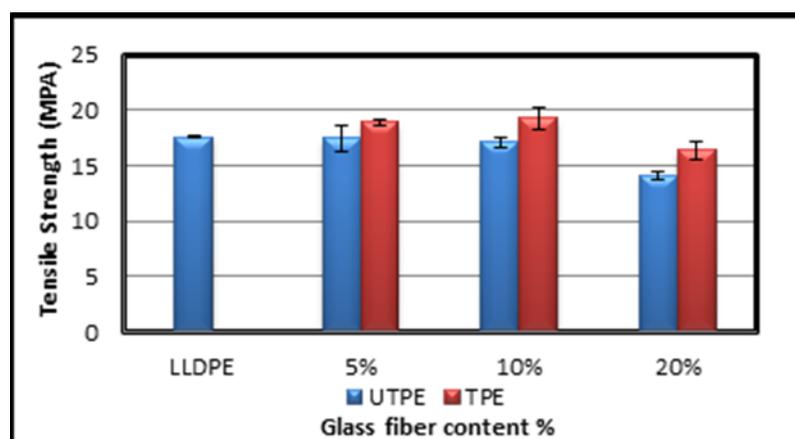


Figure 3. Tensile strength of UTPE and TPE composites.

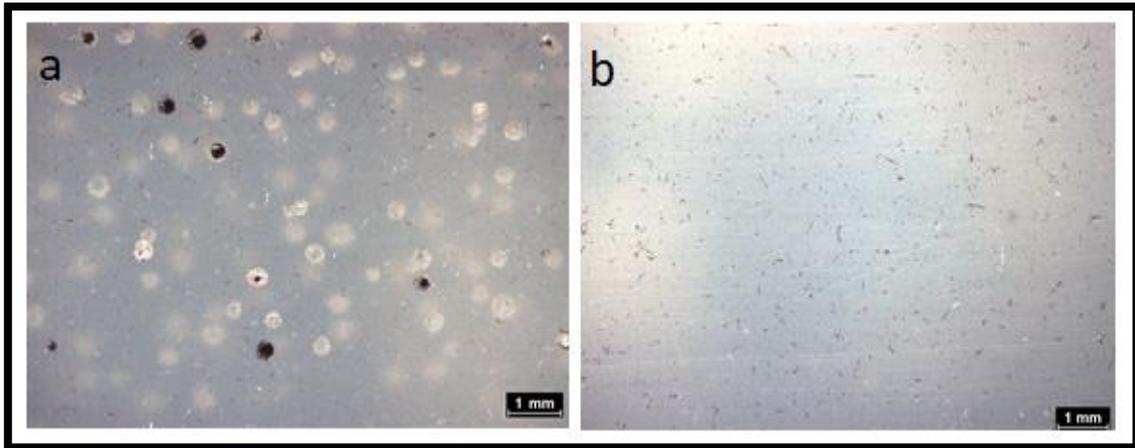


Figure 4. Optical Microscopy images of the outer surface of: a-UTPE composites b-TPE composites.

Figure 5 shows SEM images of the rotomolded composites, poor adhesion between the untreated matrix and the glass fibers is clear in Figure 5-a, on the other hand the improvement of the adhesion between treated matrix and the fibers can be indicated in figure (5-b).

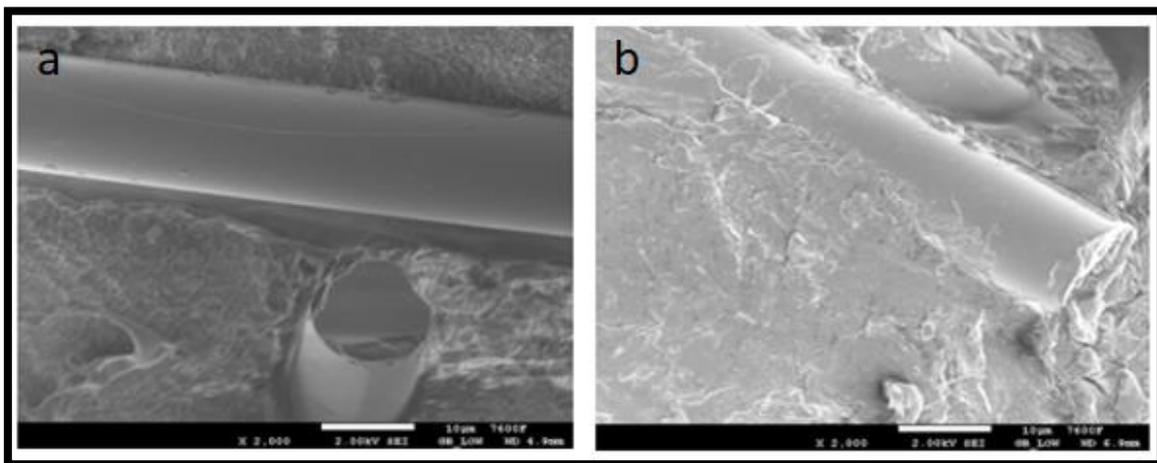


Figure 5. SEM images of rotomolded composites with (a) untreated PE, (b) treated PE.

As expected tensile modulus (Figure 6), this improvement is due to the incorporation of a rigid phase (fibers) in the matrix. Similar trends were obtained using different type of natural fibers [11,12].

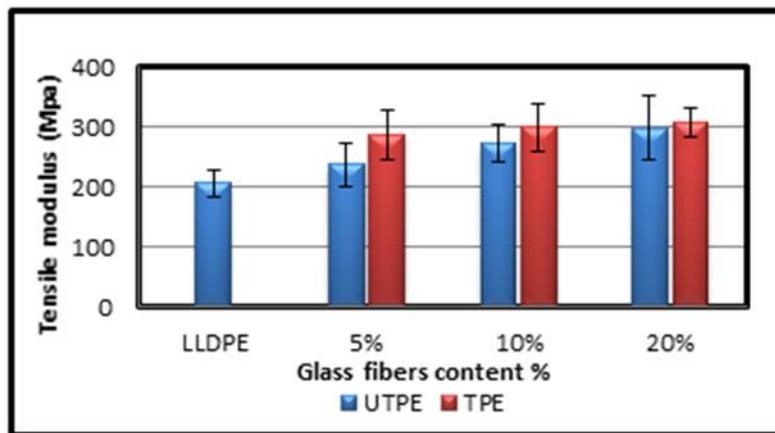


Figure 6. Tensile modulus of UTPE and TPE composites.

## 2.2. Impact strength:

Figure 6 presents the result of impact test, and it shows that as the fiber content increase impact strength decrease, independent on the type of matrix as the addition of rigid particles leads to more fibre-fibre interaction and fibre ends promoting defects and facilitating crack initiation and propagation (13-15), at 20%wt the impact strength decrease by almost 10% for both TPE and UTPE composites. Since the interface is controlling the mechanical behaviour of composites, this is why the TPE composites perform slightly better than UTPE composites, since a better interface was produced between glass fibres and LLDPE; i.e. lower amount of voids and defects.

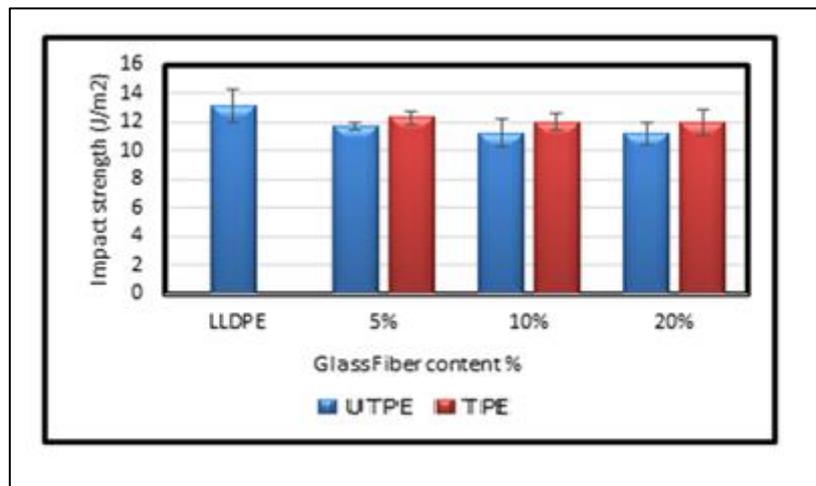


Figure 7. Impact strength of UTPE and TPE composites.

## Conclusions

Glass fibers composites based on unmodified and plasma modified polyethylene matrixes were successfully manufactured via rotational moulding. plasma treatment of PE powder improves the mechanical properties of the composites produced using treated powder comparing to the composites produced using untreated powder. Tensile strength increased by 10% as the fiber content increased

up to 10 wt.%, Tensile modulus, increased as the fibers contents increased for all composites, composite prepared with treated powder showed even higher modulus.

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