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Study of The Morphological and Mechanical Behavior of Injected Polypropylene Composites Reinforced with Short Glass Fibers

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***Abstract*— We present in this paper an experimental study of the mechanical and morphological behavior of injected thermoplastic composites. We have shown the influence of the injection temperature and the mold temperature and the injection speed and the geometry of the mold cavity on the one hand on the fibers orientation and on the other hand on the tensile behavior and impact strength of polypropylene composites reinforced with 20 to 30% of short glass fibers. Particular attention has been paid to the contribution of the fiber / matrix interface in improving the mechanical performance of the material.**

***Keywords*— Injection molding- Fibers orientaion- Mechanical characterisation - Morphological characterisation Short glass fibers- Reinforced Polypropylene - Thermoplastics composites**

I. Introduction

Transportation manufacturers as well as many other plastics users are constantly researching new formulation for new applications and performances. Short fibers reinforced thermoplastics are often desired both for their rather large formulations/ processing great possibilities, and their low cost.

The mechanical performances of these materials depend not only on the structural parameters and of the intrinsec properties of the resin and the additives, or on the only fraction of each components matrix/ fibers, but they depend deeply on the processing parameters such as for injection processing: injection speed, injection temperature, injection pressure, mold temperature [18].

The thermoplastic composites injected present an anisotropic microstructure. The evolution of this anisotropy depends not only on the nature and the concentration of the components, but also of the thermodynamic conditions of injection molding. The thermomechanical behavior of these materials depends on the variation of their microstructure. We present an experimental study of the mechanical behavior and the morphological properties of the injected thermoplastic composites.

We highlighted, the influence of the temperature of injection, the temperature regulation of the mould, the speed of injection and the mould print geometry on the fiber's orientation and behavior on the tensile and crash test of the polypropylene composites reinforced with 20 and 30% short glass fibers. A detailed attention was given to the contribution of the interface fibers/matrix in the improvement of the mechanical performances of material.

It is thus more or less marked according to the molding conditions and the geometry of the mould cavity (injection's gate, mould's thickness, flow, pressures, temperatures…). The determination of the influence of each one of these parameters on the microstructure and the knowledge of the relations which connects it to the produced part mechanical characteristics is of the interest to envisage particular conditions of transformation according to the parts to manufacture.

Thermomechanical history of thermoplastic polymers during the injection process, produce a formation of an anisotropic microstructure (crystallinity, orientation, dimension of the dispersed particles). This anisotropy influences the mechanical behavior of the produced parts carried, according to the strain's direction and of the relative position of the injection gate.

Several works was devoted to the study of the morphological variations which occur within a injected part, according to its thermomechanical history, showed the existence of several structural fields which depend at the same time on the nature of polymer and the processing conditions [1, 2].

Cox and coll. [3] study the polyamide (Pa6-6) charged with glass fibers 33%. They notice a significant evolution of stress the rupture according to the time of filling of the mould's print. The rise in the temperature of injection of polymer leads mainly to a reduction in the skin of material and to a reduction in the impact strength of the injected part [4-5-19].

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In many cases of thermoplastic composites, the bad dispersion of fibers or a lack of cohesion between fibers and the matrix prevents obtaining the required properties. The dimension and the orientation of fibers affect the mechanical properties of the finished product: the rupture stress, elongation at fracture, impact strength [14-17].

II. EXPERIMENTAL

A. *Materials*

For this study we have selected polypropylene reinforced with short glass fibres.

The polypropylene matrix was APPRYL@ 3120 MN1; this is widely marketed as transparent non-coloured granules; the density is .905; the melts conditions units, index 12g/10min and the melting point 166°C.

Glass fibres were E type, diameter 10 µm and lengths (450 µm ≤L ≤700µm), supplied by Owens-Coming.

To study the behavior of the composites subjected to the various parameters of injection, we used an injection molding machine industrial. This machine presents a clamping strength of 130 tons, equipped with a standard screw 32 mms in diameter and with an instrumented mould. It is controlled by a microprocessor. This system allows in particular the automatic adjustment of the press (mould clamping strength, temperature of the heating collars of the barrel and the nozzle…) and the adjustment of the parameters of the cycle of injection (injection speed, cooling time, injection pressure, injection temperature …).

We employ a polypropylene reinforced with short glass fibers (10 to 30%) to study the fibers orientation and the mechanical behavior in a injected part. The geometry selected is a rectangular plate which the thickness and injection gate are variable (Figure I).

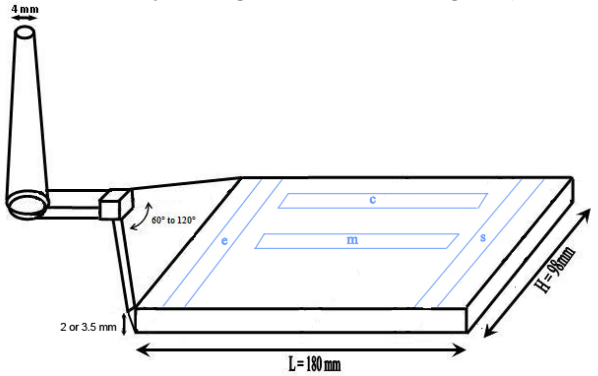

Figure I Geometry Of The Injected Plates

Influential parameters used are:

- Injection speed
- Mold temperature
- The part geometry (thickness and angle of opening of the injection threshold).

To establish the relationship between processing parameters and fiber orientation, we inject a series of samples. We polishing using standard metallographic techniques. We dissolve the glass fibers located on the surface polishing with hydrofluoric acid, then we fill the ink surface. We make the observation with the reflection optical microscope.

Qualitative analysis of fibers orientation allows a comprehensive approach to the fibers orientation at different positions of the plate (parallel and perpendicular to the flow direction).

The conditions of injection used are described in table I:

TABLE I
PARAMETERS OF INJECTION

Parameter	Value
Injection Temperature	240° C
Mold temperature regulation	60° C
Injection pressure	60Mpa
Holding pressure	55Mpa
Holding time	8 s
Screw speed rotation	156 tours/min
Vitesse d'injection	20–85 (mm/s)

III. RESULTS AND DISCUSSION

A. *morphological analysis*

Microstructural analysis and study of fibers orientation is needed to better control the variations of the different mechanical properties of composites based on molding conditions and the fibers rate.

A Level-1 Analysis of the fibers orientation

During the flow of the composite in the print of the mould, each fiber moves and is directed according to the stress imposed by its environment: the matrix, other fibers, walls of the print. After solidification, we observe a complex distribution of orientation which varies from one place to another of the part.

Thus the total observation of a sample cut out in the thickness of the plate at a distance of 90 mm of the injection gate, leads us to the following remarks (Figure II).

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Figure II Total observation of the fibers orientation in the thickness of the part [10]

- in skin: the fibers are directed randomly in the plan of the plate
- under the skin: the fibers are mainly directed in the direction of the flow
- in core: the fibers are directed perpendicular to the direction of the flow.

The observation of molded parts showed the influence of processing parameters and geometry of the flow on the fibers orientation, which partly explains the variation in mechanical properties according to the loading direction [11-12 - 13-14].

We have shown, the variation of the core layer thickness based on injection parameters (temperature, velocities) and its impact on the mechanical behavior (tensile and impact strength) of injected parts [10].

Furthermore, we observed the evolution of the fibers orientation in the length of the injected plates reinforced 30% glass fiber polypropylene. The plates are provided with an injection gate of 120 ° and a thickness of 3.5 mm. The position of the observed surfaces was z = 0.75 mm from the plate center. (Figure III)

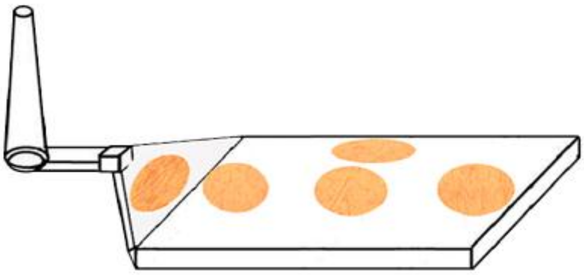

Figure III Observation of fibers orientation at different locations in the Part

It is noticed as previously that the fibers tend to be directed perpendicular to the entry of the part; on the other hand to the center of the part, they tend rather towards a distribution randomly.

To quantify the orientation of fibers, we calculated the function of orientation for each parameter in four points of the plate: entry, medium, exit and side [10].

A Level- 2Analysis of the crystal morphology

We conducted cross sections of 5 microns in the middle of the part and perpendicular to the injection direction. Observation under optical microscope between crossed polarizer and analyzer can highlight the crystalline entities in the thickness of the sample, different images are formed on a same section to obtain a «panoramic view «of the surface structure to the specimen center. We note a significant difference in morphology depending on the mold temperature during injection (Figure IV). The cooling kinetics plays an important role in the sample structure. A mold temperature of 100 ° C results in a decrease of the skin thickness of molding and an overall decrease in the number of spherulites accompanied by an increase of their volume. This change is performed by a reduction of the fraction of solidified polymer during the filling phase [7-8-16].

-A- -B-

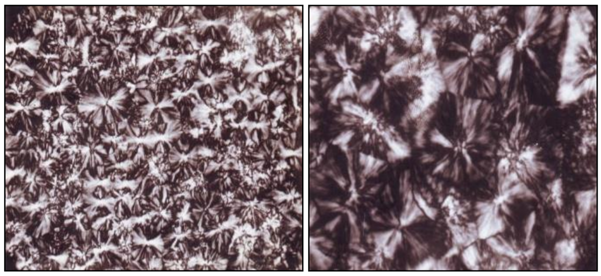

Figure IV Evolution of the polypropylene crystal morphology according to the mold temperature (Tm) ; (A : Tm = 30°C ; B : Tm = 100°C).

The analysis of cuts of samples injected with polypropylene reinforced with 20% sized glass fibers shows that the presence of fibers relatively plays a role of nucleating agent (Figure V).

We have demonstrated the formation in the vicinity of the fibers, depending on the rate of sizing, a lamellar structure different from that of the crystal lattice on the mass of the sample and oriented perpendicular to the fiber direction. It will therefore play an important role in the stress transfer between the fiber and the matrix.

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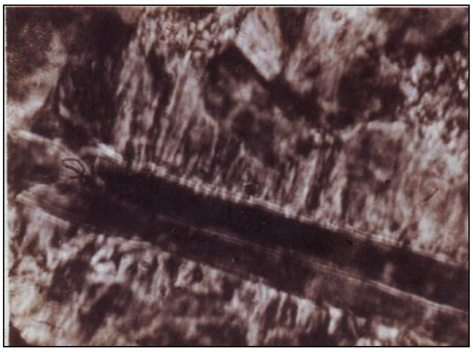

Figure V Evolution of the matrix crystalline structure in the vicinity of the sized glass fibers
Tm = 100°C

Interface fiber – matrix

-A- -B-

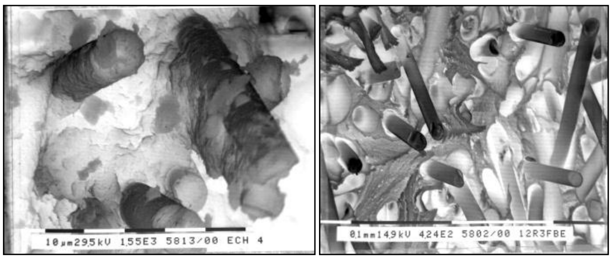

FIGURE VI MORPHOLOGICAL ANALYSIS BY SCANNING ELECTRON MICROSCOPY
A : TREATED FIBERS B : NO TREATED FIBERS

The analysis of the fracture surfaces of the injected specimens by scanning Electron Microscopy allows to highlight the evolution of the fiber / matrix interface in the presence or absence of the coupling agent (sizing).

B. Study of tensile behavior

The specimens in composites studied, cut in various positions of the rectangular plates (entered, medium, side, exit), show a very great anisotropy of the properties in traction.

The best characteristics (module and breaking stress) are obtained for samples cut in the positions (c) and (m) of the plates (fig 1). That is explained by: during the flow, the matrix imposes on the loads in suspension, under the effect of intense shearing in this zone, the fibers are directed more in the direction of the flow.

B Level-1Influence of the reinforcement rate

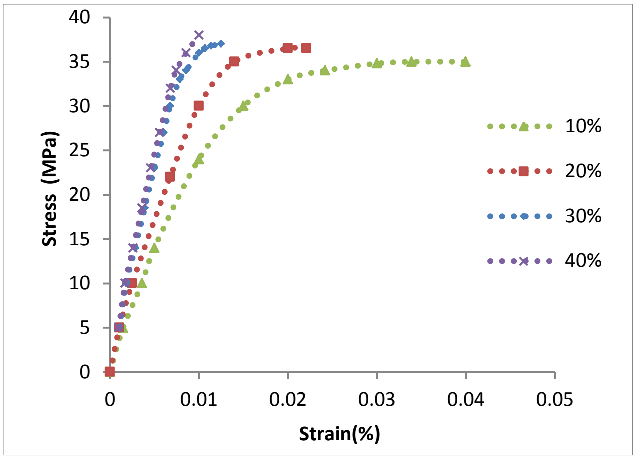

Figure VII Effect of the rate of reinforcement on the mechanical behavior

The observation of these curves shows that for the same deformation, the constraint increases with the rate of the reinforcement. The increase in the fiber rate improves the modulus of elasticity and the permissible maximum stress, on the other hand a reduction of the failure strain of material is noted.

These tests were carried out on specimens reinforced with 10% to 40% treated glass fiber, of which the goal is to highlight the effect of the fiber rate on tensile behavior of the polypropylene composite.

Influence of fibers treatment

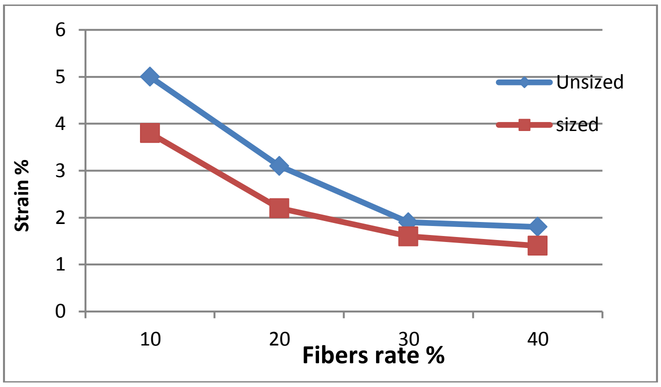

Figure VIII Untreated fibers Behavior in tensile test

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In the same way the increase in the rate of reinforcement and the fibers treatment involves an increase in resistance to the plastic flow and the breaking resistance.

B Level-2 Influence of mould temperature

Tensile tests were carried on injected sample at two mold temperatures: 30°C and 100°C. Figure 9 show the variations of mechanical characteristics depending on mold temperature.

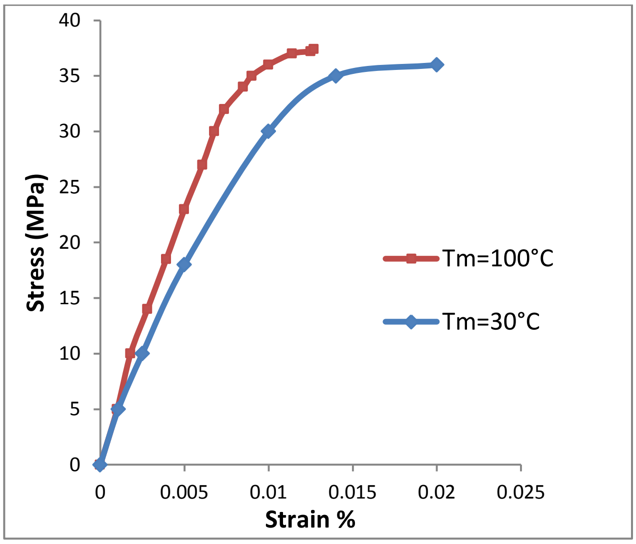

Figure IX The influence of the mold temperature on mechanical characteristics

The results show that the mold temperature seems to have a weak influence on the modulus elasticity of and breaking stress.

Indeed, when the mold temperature increase, the elasticity modulus and breaking increase, while failure elongation decreases.

We can explain this evolution by the reduction thickness of the layer solidified instantaneously during the filling of the print when the temperature of the mould increases, and it results a more developed microstructure of final part (more significant size of spherulites) and thus a better rigidity.The speed of injection presents the same effect as the mold temperature.

B Level-3 Influence of injection speed

We observe a downward trend of breaking stress when the injection speed increases, and that independently of the nature of the interface and the fibers rate used.

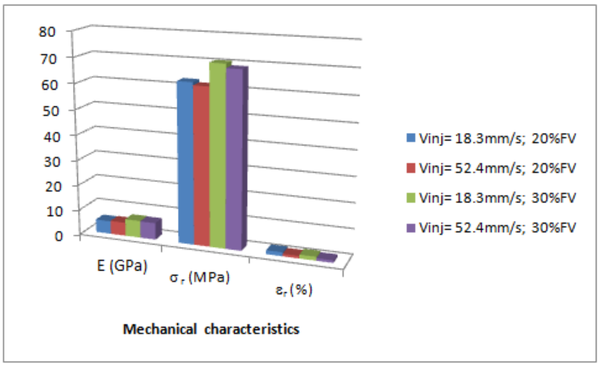

Figure X The influence of the injection speed on mechanical characteristics

The influence of the injection speed on the mechanical behavior is done via its influence on the fibers orientation of injected material. Moreover it is not the only parameter which conditions the fibers orientation. Indeed the thicknesses of the specimen and the reinforcement rate have in their turn a significant influence on this orientation. In the case of our samples, approximately 4,15 mm thickness, and the fibers rates used, 20% and 30%, the variation of the injection (18,3 mm/s and 53,4 mm/s) do not allow a significant evolution of the fibers orientation. That explains the weak influence on the mechanical properties of material.

C. Study of the impact resistance

The specimens tested were cut in rectangular plates injected (figure I; position: S).

Under these conditions, there is a dispersion of results within the same batch of samples. The values presented on table 6 are the average of the results obtained starting from ten specimens tested for each test.

Table II
Evolution of the impact resistance of polypropylene reinforced with 30% glass fibers according to the mould temperature and the injection speed

Test N°	T mould (°C)	Injection speed (mm/s)	E (Kj.m^{-2})
1	30	20	35± 1,7
2	30	52	31,2 ±2
3	60	20	29,1± 2,1
4	60	52	24,5 ± 2,3

We show that:

- when the temperature of the mould increases the impact resistance decreases
- the rise of the injection speed also weakened impact strength.

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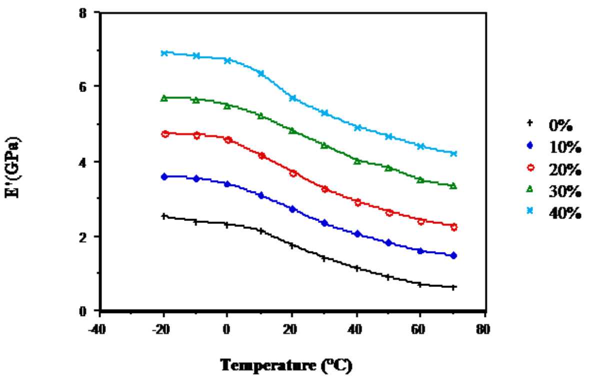
This evolution of the impact resistance according to the two temperatures is related on the reduction thickness of the materiel skin and to the crystalline structure developed during the phases of filling and cooling of the injection cycle.

D. Thermomechanical testing

Several studies have been devoted to the study of the rheological properties in the solid state materials other than metals. Moreover, experience shows that all thermoplastics exhibit a relaxation at very low temperatures.

This particular behavior depends on the injection conditions and fiber-matrix interface . The variation with time of the module is the result of molecular motions whose amplitude is a function of time and temperature.

It shows that the relaxation involves the effect of microstructure on the properties of molded parts The thermomechanical tests were performed with a dynamic mechanical measuring device (DMTA) . This measure gives the variation of the bending elasticity modulus E ' as a function of temperature at a given frequency, and the mechanical damping coefficient tg. The evolution of the dynamic elastic modulus of the polypropylene as a function of temperature and the rate of sized fibers is illustrated in Figure XI.



FigureXI Evolution of the dynamic elastic modulus as a function of temperature for different rates of sized fibers, Tmold = 30 ° C.

Figure XII shows the evolution of the angle tangent of mechanical loss versus temperature for different levels of fiber. The peak of the transition decreases with increasing rate of fiber. This results in a decrease of the damping of the material and therefore of its impact resistance.

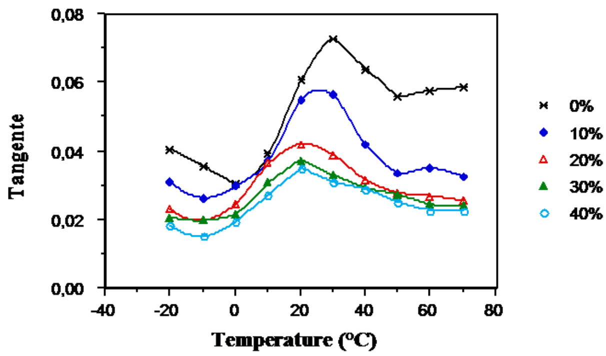


Figure XII Evolution of the angle Tangent of mechanical loss of the polypropylene according to the temperature and the rate of short glass fibers unsized Tmold = 30 ° C.

IV. CONCLUSION

The use of thermoplastics composites reinforced with short glass fibers in engineering applications is increasing and diversifying. There are used in the automotive, electric and aeronautical industry. The improvement of their mechanical performances passes by the control and the knowledge of the various mechanisms of their damage.

We led a thorough study of the mechanical behavior of polypropylene reinforced to various short glass fiber rates. We highlighted the incidence of the processing parameters on the mechanical properties of the injected part. The role of the interface fibers/matrix is also determining in the improvement of the mechanical performances in term of transfer of load between the components.

The microscopic observation of the injected part showed the influence of the parameters of transformation and the geometry of the flow on the orientation of fibers, which partly explains the variation of the mechanical properties with the direction of request.

REFERENCES

[1] Clegg D.W. 1994. Handbook of Polymer-Fiber Composites, Polymer Science and Technology Series. Longman Scientific & Technical. New York.

[2] A. Haddout, G. Villoutreix; Colloque Matériaux "Science et Industrie", Paris - Juin 1992.

[3] H. L. Cox, Mentzer; Polym. Eng. Sci., 7, 26(1986).

[4] G. Twigg, A. Poursartip, G. Fernlund, Tool-part interaction in composites processing. Part II: numerical modelling, Compos.: Part A 35 (2004) 135–141.

International Journal of Recent Development in Engineering and Technology
Website: www.ijrdet.com (ISSN 2347 - 6435 (Online) Volume 2, Issue 5, May 2014)

[5] A.M. Harte, J.F. Mc Namara, Use of micromechanical modelling in the material characterisation of overinjected thermoplastic composites, J. Mater. Process. Technol. 173

[6] M. Benhadou ; A. Haddout ; M. Mazouzi; G. Villoutreix; Injection de composites en polypropylène /fibres de verre courtes : étude de la distribution et de l'orientation des fibres ; AMCA 2008

[7] M. Bouarourou; L. Safrioui; A. Haddout; M. Benhadou; M. Mazouzi; G. Villoutreix; Etude du comportement en relaxation de composites injectes en polypropylène/fibres de verre courtes ; AMCA 2008

[8] M. Benhadou; A. Haddout; S. Ajana; M. Bouarourou; G. Villoutreix; Maitrise de la qualité des composites en polypropylène/fibres de verre courtes injectés ; "M2RI'08"

[9] M. Benhadou, A. Haddout, G. Villoutreix; Injection de composites en polypropylène/fibres de verre courtes : étude expérimentale de l'orientation des fibres dans le moule ; Xèmes Journées Maghrébines des Sciences des Matériaux JMSM'2006

[10] A. Haddout(1); M. Benhadou(1); F. Rhrich; L. Safraoui; G. Villoutreix(2) Experimental study of fibers orientation and distribution during the injection molding of thermoplastic composites; Journal of composite materials; submitted mauscript ID: JCM-14-0320

[11] A. Haddout, F. Rhrich, M. Benhadou, L. Safraoui; On Aircraft Materials, AMCA 2010, Marrakech 12 – 14 Mai 2010

[12] M. Benhadou, A. Haddout, S. Ajana, M. Bouarourou, G. Villoutreix Maitrise de la qualité des composites en polypropylène/fibres de verre courtes injectés ; "M2RI'08" Maîtrise et Management des Risques Industriels, Oujda 2008

[13] D. Dray, P. Gilormini, G. Regnier. Composites Science and Technology 67 (2007) 1601–1610.

[14] D.J Lee, M.W Kim, S.Y Kim, S.H Lee, J.R Youn. Korea-Australia Rheology Journal, Vol. 22, No. 2, June 2010 pp. 95-103.

[15] S.I.S Shaharuddin, M.S Salit, E.S Zainudin. Turkish J. Eng. Env. Sci. 30 (2006) , 23-34.

[16] Vélez-García GM , Wapperom P, Kunc V, Baird DG, Zink-Sharp A. J Microsc. 2012 Oct;248(1):23-33. doi: 10.1111/j.1365-2818.2012.03646.x

[17] E. Lafranche, P. Krawczak, J. P. Ciolczyk, J. Maugey. eXPRESS Polymer Letters Vol.1, No.7 (2007) 456–466. Available online at www.expresspolymlett.com. DOI: 10.3144/expresspolymlett.2007.64

[18] Ahmed N. Oumer and Othman Mamat. Asian Journal of Scientific Research. 2013, Volume: 6, Issue: 3, Page No.: 401-410. DOI: 10.3923/ajsr.2013.401.410

[19] D.McNally. Polymer-Plastics Technology and Engineering. 03/2008; 8(2):101-154. DOI:10.1080/03602557708545033