



Polymer composite strengthening by developed injection moulding technique

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ABSTRACT

Purpose: This work presents method of mechanical strengthening of nanoreinforced polymer and immiscible polymer blends of polypropylene (PP) and polycarbonate (PC) obtained by non-conventional injection moulding technique.

Design/methodology/approach: Melt manipulation of solidifying polymer inside mould cavity has been used for improving fracture toughness of polymer composites and nanocomposites.

Findings: Improvement of mechanical properties, especially for nanocomposites, has been obtained due to different processing conditions and ratios of particular components.

Research limitations/implications: Nanocomposites of polymer blends and montmorillonite were moulded by direct injection moulding according to melt temperature and stroke time-number combination included in design of experiments.

Practical implications: Comparison of analyzed composites brought satisfying results proving advantage of implicated method and composites ratio as well.

Originality/value: Non conventional injection moulding method and their influence on strengthening polymer composites and nanocomposites is promising technology with multi-applicable character.

Keywords: Engineering polymers; Composites; Injection moulding; Polymer processing

MATERIALS

1. Introduction

Thermomechanical treatment of melt polymer due to the processing setup during injection moulding process brings possibility to manipulate polymer structure (tailoring) and therefore determining its mechanical behavior [1,2]. Changing of material's composition, like e.g. grafting, which reduces the molecular mobility affecting the deformation capabilities of the material, and thermomechanical processing treatment allow resolutely tailoring the material morphology for improved mechanical performance [3,4].

Injection moulding of polypropylene as one of most common worldwide use technique in production of domestic and advanced engineering materials [5,6] has been use in this research. Manipulation of the melt polymer during the cooling phase can be achieved by non-conventional injection moulding techniques, such as SCORIM. The solidifying polymer is subjected to reciprocal flow movements inside the mould cavity by extracting and retracting external hydraulic pistons. SCORIM can improve structure giving possibility to create highly oriented multilayered structure with enhanced mechanical properties [6,7].

Application of reinforcing materials, especially nanosized particles (e.g., layered silicates like montmorillonite-MMT) is a

recent way for improving the mechanical behavior of polymer composites [8,9,10]. Polymer nanocomposites are a worldwide research field in the last years, namely those based on layered silicates. The amount of nanoparticles in a polymer nanocomposite can vary at different ratios. Generally, for the sake of high cost of nanofillers, filling is employed up to 5% of nanoclay [11]. However, ratios of nanoparticles up to 10% has been reported [12]. The challenging area is the production of good polymer nanocomposites by compounding the nanofiller with polymer matrix (e.g. PC or PP) by melt mixing, in situ polymerization and other processes. The main issue for guaranteeing good mechanical properties is the exfoliation and dispersion states [13]. Exfoliation of nanocomposites can be obtained by transporting the molecules of silicate layers from the bulk to the interlayer zone between the silicate sheets [14]. The interactions between polymers in a blend can be altered by the addition of nanosized particles. The study of polymer-polymer reinforced nanocomposites is therefore and attractive route to obtain high mechanical performance polymeric materials.

Abbreviations used in article

CIM	Conventional Injection Moulding
N-CIM	Non Conventional Injection Moulding
SCORIM	Shear Controlled Orientation in Injection Moulding
ST	Stroke time [s] – time of pistons' movements
SN	Stroke number – number of pistons' movements
Tm	Melt temperature [°C]
Run	Set of variable processing parameters (ST, SN, Tm)

2. Experimental procedure

The polymers used in this work are shown in Table 1. PP has been used as the matrix and PC as the disperse phase on the polymer-polymer composites. Ratios of particular materials, which composites are composed of, are listed in the Table 2. The materials were blended in a rotative drum at the speed of 60 rpm.

Table 1.

Materials used in the experiments

Polypropylene (PP)	Moplen HP 501M, Basell
Polycarbonate (PC)	Lexan 123R, GE Plastics
Montmorillonite (MMT)	Nanofil 5, Süd-Chemie
PP grafted with maleic anhydride (MAP)	Licomont AR 504, Clariant

Table 2.

Particular components (wt %)

	Abbr.	PP	PC	MAP	MMT
1	PP	100	-	-	-
2	PP/MAP	97	-	3	-
3	PP/MMT	97	-	-	3
4	PP/PC	70	30	-	-
5	PP/PC/MAP	67	30	3	-
6	PP/PC/MMT	67	30	-	3

Two types of injection moulding techniques have been used in the work: i) SCORIM technique (melt manipulation injection moulding; ii) conventional injection molding - for comparison reason. Injection moulding machine Ferromatik Milacron type K-85 was used in experiment. Machine was equipped with a special mounted mold with two externally controlled hydraulic pistons. Injection process was performed under stabilized condition and specimens were moulded into rectangular bars with dimensions of 130x13x8 mm.

The molding programme was defined according to a design of experiments (DOE) approach. Complete set-up bases on 2 levels (extreme settings during injection moulding process) and concerning 3 factors (main injection parameters) organized in 8-run table (Table 3). Three processing parameters were considered in the molding programme: the melt temperature (for CIM and N-CIM), stroke time and stroke number (just N-CIM). Other processing parameters were kept constant:

- injection velocity 10 mm/s
- hydraulic pressure 150 bar
- holding pressure 50 bar
- mold temperature 30 °C
- cooling time 30 s

Specimens were tested by 3-point bending test with crosshead speed 10 mm/min (according to the ASTM E399 standard) at controlled room temperature (23°C). Fractured specimens were previously notched in a Ceast notch cutter type 6816 with a notch depth of $a = 6.35$ mm and then sharpened with razor blade. For each processing run were tested at least five specimens in a universal testing machine Instron type 4505. Aim of mechanical characterization was comparison of synergetic effects of polymer-polymer composites and influence of processing conditions on the fracture behaviour. The fracture energy, used as comparable value between processing parameters, was calculated by a J-integral value, J_0 , directly derived from the integration of the load-displacement diagram basing on the total energy absorbed during test, U :

$$J_0 = \frac{\eta_e U}{B(W-a)} \quad (1)$$

3. Discussion of experimental results

Conventional Injection Moulding

Fracture behaviour of composites processed by CIM (Fig. 1) show analogous scheme depending of melt temperature. PP/PC and PP decreases while temperature rises. PP and PP/PC fracture energy decreases while temperature rises. PP/PC composites show much higher values than PP (for both temperatures). Inverse behaviour is reported for nano-composites, where higher results belongs to PP. The addition of the nanoclay to the pure PP improved the fracture toughness, mainly for the $T_m = 280$ °C. Pure matrix of PP favors well distribution of nanoparticles. For the PP/PC nanocomposite the fracture response is lower than for PP nanocomposite and is similar for both temperatures.

Table 3. Set-up of processing parameters

		RUN							
		1	2	3	4	5	6	7	8
Stroke time [s]	ST	1	1	1	4	3	3	3	3
Stroke number	SN	3	3	12	12	3	3	12	12
Melt temperature [°C]	Tm	240	280	280	240	280	240	240	280

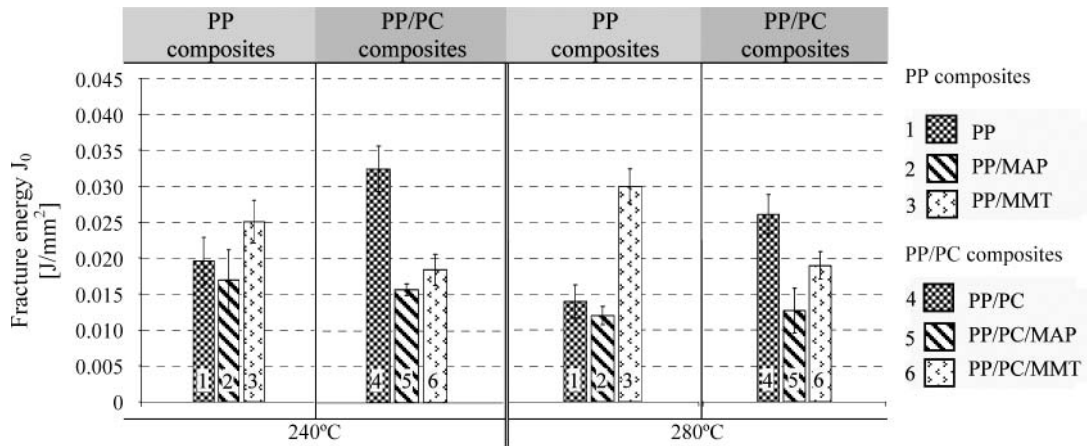


Fig. 1. Fracture toughness of the materials systems processed by CIM

PP/MAP blends show always the lowest J_0 values within each set of composites. The specimens molded with low temperature (240°C) of both PP and PP/MAP, have the highest J_0 values. These mouldings should present the low degree of crystallinity (low melt temperature), evidencing therefore superior fracture toughness. For the PP nanocomposites, the increment on the melt temperature leads to an increase of J_0 values. High melt temperatures should favor a better exfoliation and promote smaller nanoparticles clusters (nanoclay agglomeration). This still needs to be observed by carrying out a detailed morphology characterization. The PP/PC blend shows improved fracture behavior at both melt temperatures (a maximum value of 0.032 J/mm²).

Non-Conventional Injection Moulding

Results of N-CIM technique at low temperature are presented on diagram (Fig. 2). The x-axis illustrates number and time of piston movements (strokes) respectively, according to Table 3. The maximum fracture energy for pure PP derives from highest value of ST-SN (3-36) reaching 0.022 J/mm². The increment of ST and SN increases the fracture toughness of the moldings. Variations of the fracture toughness of pure PP and PP/PC blend with the processing conditions for N-CIM at high temperature is presented on Fig. 3.

The mechanical behaviour of the PP/PC blends presents higher values of J_0 compared with the pure PP. This is more evident for the specimens molded with low ST.

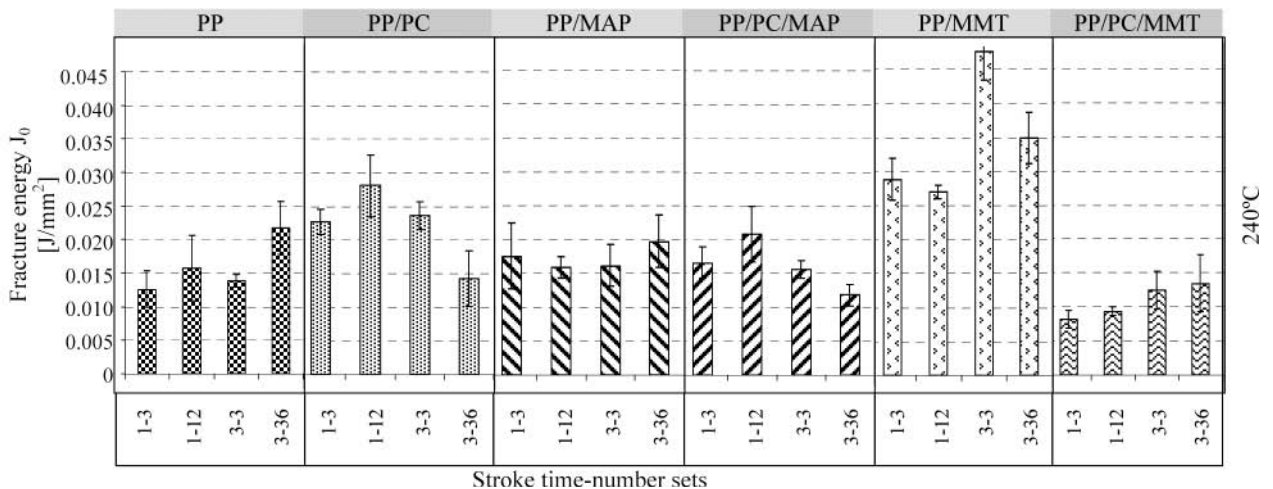


Fig. 2. Fracture energy of the materials systems processed by N-CIM

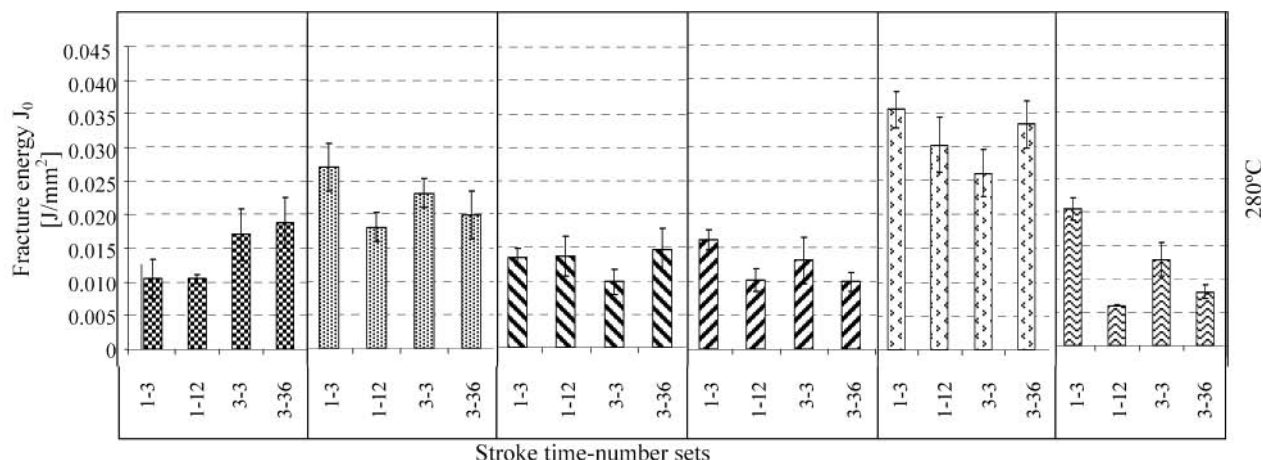


Fig. 3. Fracture energy of the materials systems processed by N-CIM

4. Summary

The melt manipulation during solidification alters significantly the fracture toughness of the mouldings.

The addition of MAP and MMT results in different fracture toughness values. The higher improvement on J_0 comes from PP/MMT, especially when N-CIM is used. Adding MAP to pure PP is weakening composite, mainly in CIM.

The PP/PC blend shows improved fracture behavior when compared to pure PP, principally when processed by CIM.

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References

- [1] M. Arroyo, R. V. Suárez, B. Herrero, M. A. López-Manchado, Optimization of nano-composites based on polypropylene/polyethylene blends and organo-bentonite, *Journal of Materials Chemistry* 13 (2003) 2915-2921.
- [2] S.S. Ray, M. Okamoto, Polymer/layered silicate nanocomposites: a review from preparation to processing, *Progress in Polymer Science* 28 (2003) 1539-1641.
- [3] J. C. Viana, N. Billon, A. M. Cunha, The thermomechanical environment and the mechanical properties of injection moldings, *Polymer Engineering and Science* 44 (2004) 1522-1533.
- [4] J. Konieczny, L.A. Dobrzański, R. Nowosielski, J.J. Wysocki, A. Przybył, Soft magnetic nanocomposite with powdered metallic ribbon based on cobalt and polymer matrix, *Proceedings of the 12th Scientific International Conference „Achievements in Mechanical and Materials Engineering” AMME’2005, Gliwice – Zakopane, 2005, 327-330.*
- [5] E. Bociąga, T. Jaruga, J. Koszul, Plastic flow investigation in multicavity injection mold, *Proceedings of the 12th Scientific*

- International Conference „Achievements in Mechanical and Materials Engineering” AMME’2003, Gliwice – Zakopane, 2003,107-110.*
- [6] D. Kwiatkowski, J. Nabialek, A. Gnatowski, The examination of the structure of PP composites with the glass fibre, *Archives of Materials Science and Engineering* 28/7 (2007) 405-408.
- [7] R.A. Sousa, R.L. Reis, A.M. Cunha, M.J. Bevis, Processing and properties of bone-analogue biodegradable and bioinert polymeric composites, *Composite, Science and Technology* 63 (2003) 389-402.
- [8] M. Alexandre, P. Dubois, Polymer-layered silicate nanocomposites: preparation, properties and uses of a new class of materials, *Materials Science and Engineering* 28 (2000) 1-63.
- [9] P. Uribe-Arocha, C. Mehler, J.E. Puskas, V. Alstädt, Effect of sample thickness on the mechanical properties of injection-molded polyamide-6 and polyamide-6 clay nanocomposites, *Polymer* 44 (2003) 2441-2446.
- [10] L.A. Dobrzański, *Fundamentals of Materials Science and Metallurgy. Engineering Materials with fundamentals of Materials Design*, WNT, Warsaw, 2002 (in Polish).
- [11] M. Bilewicz, J.C. Viana, A.M. Cunha, L.A. Dobrzański, Morphology diversity and mechanical response of injection moulded polymer nanocomposites and polymer-polymer composites, *Journal of Achievements in Materials and Manufacturing Engineering* 15 (2006) 159-165.
- [12] P. Uribe-Arocha, C. Mehler, J.E. Puskas, V. Alstädt, Effect of sample thickness on the mechanical properties of injection-molded polyamide-6 and polyamide-6 clay nanocomposites, *Polymer* 44 (2003) 2441-2446.
- [13] L.A. Utracki, *Polymer Blends Handbook*, Kluwer Academic Publishers, Boston-London, 2002.
- [14] S. Lee, Y.T. Ma, H.W. Rhee, J. Kim, Exfoliation of layered silicate facilitated by ring-opening reaction of cyclic oligomers in PET-clay nanocomposites, *Polymer* 44 (2005) 2201-2210.
- [15] J.C. Viana, Development of the skin layer in injection moulding: phenomenological model, *Polymer* 45 (2004) 993-1005.