Influences of impurities in recycled plastics on properties of PIM sandwich panels

Kun Qi, Jim Song and Karnik Tarverdi*

Wolfson Centre for Materials Processing
Brunel University, Uxbridge, Middlesex, UB8 3PH, UK
*corresponding author: jim.song@brunel.ac.uk

Abstract

Powder impression moulding (PIM) is a novel technology for manufacturing lightweight sandwich panels from plastics in powder form. The process is featured by its high tolerant to impurities or contaminants in the feedstock and thus requires much less materials segregation and cleaning operations when use recycled plastics. This paper investigate the influences of polymer impurities and soil contamination on structure and properties of PIM sandwich panels using compositions that simulate a PE-rich recycled plastic feedstock. It is demonstrated that the PIM process can accommodate considerable impurities (rPET residues or soil contamination) in a core dominated by LDPE/HDPE blendes. The variation of flexural properties can be predicted or controlled through monitoring of impurities. There exist significant scopes for reduction of the degree of sorting and cleaning in recycling systems by using lower grades of recyclates and for reduction of the associated costs and energy consumption.

Key words: PIM, recycling, mixed polymers, impurities, contamination, sandwich panels.

1. Introduction

Recycling rate of plastics is significantly lower than other materials because of the difficulties and high costs in separation and cleaning of waste plastics[1]. This has stimulated efforts in the developments of plastic recognition and separation techniques over the years. Techniques based on flotation/sedimentation[2-3], on magnetic/optical properties[4-5] or on colour and chemical composition of the materials [6-7] are often used in combination in plastic recovery facility (PRF). High purity PS (http://www.axionconsulting.co.uk/) and food grade PE and PET are now commercially available (http://www.closedlooprecycling.co.uk).With each step improvement in purity, however, more complex and expensive systems are required which results in higher investment and running costs and more material rejection. It is thus highly desirable that the down-stream processing technologies can produce high quality products utilising the relatively low-purity recycled plastics and in this regards, Powder Impression Moulding (PIM) is a technology with great potentials. Using low grade plastics in powder form, PIM process is capable of moulding lightweight panels with solid skins sandwiching a foamed core. The skin powder materials are laid on two halves of a heated flat –bed mould and sintered to solid shins and on to one of the moulds, powder with blow agent for the core material is spread. The moulds are then closed and heated to a temperature at which a foamed core is produced and bond to the skins. As minimum material flow is required and the non-foaming particles (e.g. impurities or contaminants) can be encapsulated by the dominant composition in the material, PIM is much more tolerant to mixed plastic or impurities in the feed stock than conventional extrusion and moulding techniques for recycling. This enables PIM to produce high performance sandwich panels that have found many applications in construction (e.g. hauling boards, bathroom wet floor systems and concrete moulds). It is also possible to manufacture hybrid structures with embedded pipes or reinforcements (http://ert4c.com).

This paper presents an investigation in the influences of impurities and contamination on the PIM process and quality of the sandwich panels. The compositions studied simulate a system of contaminated PE-rich mixed plastics with a HDPE/LDPE blend at different ratios representing the dominating composition, up to 15 wt % recycled PET representing additional plastic impurity and up to 15 wt% sand representing contamination from soil.

2. Experimental details

2.1 Materials

The materials used are listed in Table 1. The virgin HDPE and LDPE were supplied in pulverized powder form with particle size of 100-400μm. The powder of rPET was from recycled bottles with particle size of

50-400 µm. The HDPE was used to form the skins of the sandwich panel. The LDPE/HDPE blends, rFET and sand (mean particle size ~250µm) were used in the formulation of the cores as shown in Table 2 using an Orthoplan design tool (SPSS15, SPSS Inc.). The LDPE/HDPE blends as the dominating PE composition of the cores were prepared at 6 different mass ratios. The rPET and sand were added up to15 wt% base on total mass of the feedstock and the blow agent OBSH was added at 1.5 wt% based on mass of the core. The PE blend/rPET/sand/OBSH compounds were mixed using a high speed mixer for 15 minus prior to moulding as described below.

Table 1: Raw material information

Materials	Supplier	Grade	Descriptions	T_m°C
HDPE	Exxon, UK	HMA014	for HDPE/LDPE blends	135
LDPE	Exxon, UK	LD362	for HDPE/LDPE blends	115
rPET	Severnside Recycling UK	>95 purity	as Polymer impurity	248
Sand	Wickes, UK	Block paving	as "Soil contaminant"	-
OBSH*	Celogen, UK	Industrial grade	Blow agent (powder)	-

* OBSH-Oxybis Benzene Sulfonyl Hydrazide with recommended working temperature of 158-160°C

2.2 Sample preparation

Table 2: Compositions for the cores (wt%)

code	PE %	ratio of LDPE/HDPE	Sand %	rPET %
M01	80%	**90/10**	5%	15%
M02	95%	**90/10**	0%	5%
M03	85%	**50/50**	0%	15%
M04	95%	**75/25**	5%	0%
M05	90%	**75/25**	0%	10%
M06	75%	**100/0**	10%	15%
M07	100%	**100/0**	0%	0%
M08	80%	**100/0**	15%	5%
M09	70%	**75/25**	15%	15%
M10	85%	**100/0**	5%	10%
M11	90%	**50/50**	5%	5%
M12	75%	**50/50**	15%	10%
M13	85%	**90/10**	15%	0%
M14	85%	**75/25**	10%	5%
M15	90%	**50/50**	10%	0%
M16	80%	**90/10**	10%	10%

For each composition in Table 2, solid planks (200x200x3 mm) were compression moulded with a hot press at 190°C and 5 MPa. For elongational viscosity measurements of the LDPE/HDPE blends, samples of 20x10x1mm were compression moulded at 190 °C and 10 MPa. Sandwich panels were moulded with a purpose–built laboratorial PIM machine. The mould (internal dimensions: 450 x 300 x20mm) was fitted with sensors for monitoring temperature and clamping force during moulding. The open moulds are preheated to 190 °C using an oil heater. 250g of the HDPE skin material was applied on each side of the mould and allowed to sinter. 710g of the core material was then applied to the lower half of the mould. The mould was then closed and clamped for 15 min to allow the thermal decomposition of the blow agent and foaming of the core. It was then brought in contact with a cooling station connected to a chiller to bring the temperature to room temperature.

2.3 Characterisations and tests

The melt flow rate (MFR) of the LDPE/HDPE blends was measured (at 190°C and under a 2.16 kg weight) with a melt rheometer (MeltFlow ST, Haake) while the uniaxial elongational viscosity of the LDPE/HDPE blends was measured at 150 °C in nitrogen atmosphere and constant strain rates of 0.01, 0.1, and 1.0/s with an elongational rheometer (ARES, TA Instrument, UK) equipped with an Extensional Viscosity Fixture (EVF) as detailed elsewhere [8-9].

Specimen were cut with a band saw from the moulded sandwich panels to 300 x 35 x20 mm and allowed a 3-week conditioning at 50±5% RH and 23 ±1 °C for relax before density measurements and mechanical testing. Density of the panels was obtained from their mass and dimensions. 3-point flexural tests were conducted at crosshead speed of 10 mm/min for flexural modulus and strength of the panels using an Instron mechanical tester and a minimum of five samples were used.

Foamed cores were sectioned with a sharp blade to assess foam cell structures at various positions of the moulded panels with a stereo microscope (SZX16, Olympus).

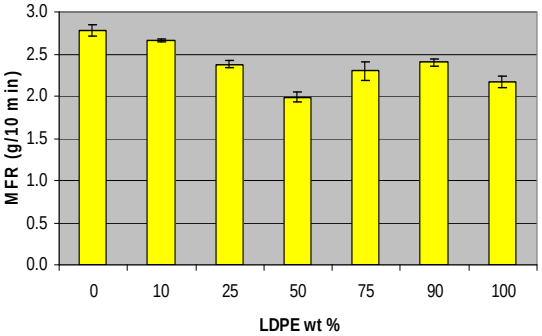

Fig.1: Variation of MFR with wt% of LDPE in the LDPE/HDPE blends.

3 Results and Discussion

3.1. Rheology of the LDPE/HDPE blends

MFR controls processing pressure and mould filling in extrusion or moulding process. Influence of composition of the LDPE/HDPE blends on MFR is shown in Fig. 1. For the chosen grades, the difference in the pure HDPE and LDPE is marginal (2.8 and 2.2 g/10min) and blending gave rise to intermediate MFRs, reaching a minimum at 50/50. This, together with the fact that in PIM, the melt only needs to flow locally driven by pressure created from the foaming agent, indicated that blending of the PEs will not have significant influence on their melt flow behaviour.

Elongational viscosity is critical to processing of polyethylene[10-11] and in particular, strain hardening has been found desirable in foaming of PEs as it stabilizes the cell structure by avoiding cell wall rupture as bubbles grow[12-14]. Fig. 2 shows the effect of LDPE content on strain hardening (Note that the curves have been spaced out vertically for clarity and thus do not represent true viscosity values). The pure LDPE shows considerable strain hardening at high elongation strain as a result of the high content of long chain branches while the pure HDPE (with low content of long chain branches) exhibit very little strain hardening. Increasing LDPE content in a LDPE/ HDPE blend therefore should benefit expansion and foam stability in PE blends.

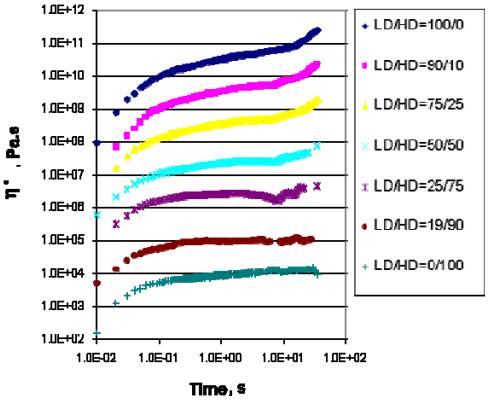

Fig. 2: Elongational viscosity showing change in strain hardening in the LDPE/HDPE blends (strain rate 0.1 s^{-1}).

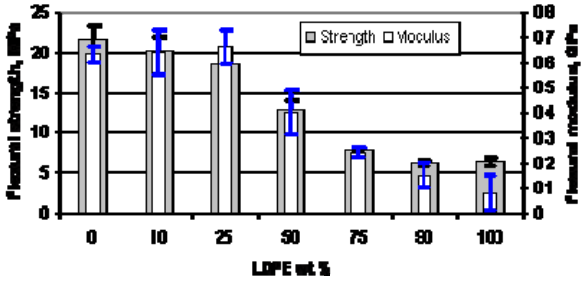

Fig. 3: Flexural properties of LDPE/HDPE blends.

3.2 Properties of the LDPE/HDPE solids

Flexural properties of the LDPE, HDPE and their blends are shown in Fig. 3. The HDPE shows significantly higher flexure modulus and flexure strength than the LDPE and as expected, blending of the LDPE and HDPE powders resulted in reduction of the flexural properties as LDPE content increases. With inclusion of the impurities, rPET and sand, the statistical significance of various factors (LPDE/HDPE ratio and contents of the sand and rPET) on flexural properties, was analysed using Orthoplan (SPSS15, SPSS Inc.) for a level of confidence of 95%. As shown in Fig.4, LDPE/HDPE ratio has significant effect on flexural modulus and strength of the solid compounds whereas sand and rPET impurities up to 15 wt% have negligible effect.

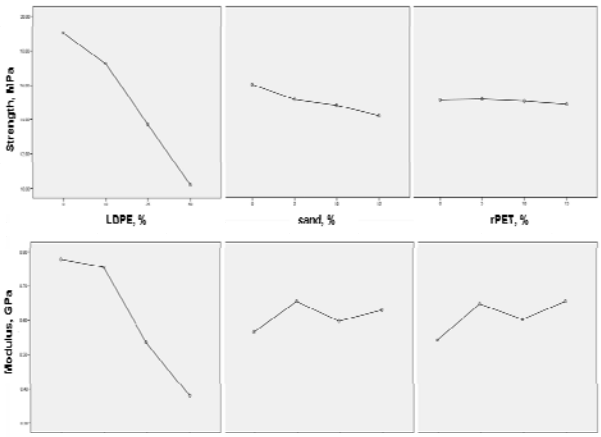

Fig.4: Effect of LPDE/HDPE ratio, sand and rPET contents on estimated marginal means of flexural strength (top) and modulus (bottom) of the solids.

3.3 Structure and properties of PIM panels

With exceptions of M06 and M10, densities of most sandwich panels (Fig. 5) are slightly lower than the theoretical density of 448 kgm^{-3} calculated from the constant mass of the materials and mould cavity. This is in part attributable to leakage of molten materials as flash during moulding - an indication of good expansion of the core. All compositions were foamed satisfactorily judged by good mould filling and surface finish and no advert effect was observed for compositions with high sand and rPET contents.

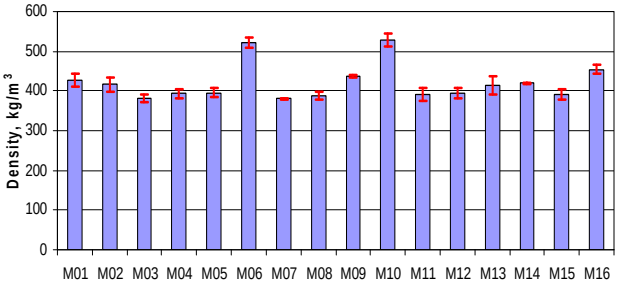

Fig. 5: Density of the sandwich panels with their core formulations shown in Table 2.

The higher density M06 and M10 PIM samples have thickness of 17.5 and 18.5mm, respectively,

considerably less than that of the mould height of 20mm. The relatively higher density may be attributable to processing i.e. they were demoulded too soon when the core were still at high temperature.

Fig. 6 shows the optical micrographs of a typical cross section of PIM samples. The microstructure is characterised by a closed-cell porous core well integrated with the solid skins. Assessment of the cross sections of all sandwich samples showed that the cores were uniformly foamed without formation of large voids. Close examination of the pores (e.g. Fig.7) showed that they are quite uniform in size ranging from 50 to 300 um in diameter. The increase of sand resulted in some refinement of the pore size which may be attributable to enhanced nucleation at the sand/polymer interfaces.

Figure 6: Optical micrographs of cross section of the PIM samples M16 with 80 wt% PE (LDPE/HDPE=90/10) and 10 wt% sand and 10 wt% rPET (from left to right: the top skin, the core, and the bottom skin).

Fig.7 Optical micrographs of the PIM sample core (from left to right) M04, M15, and M13, with 5, 10 and 15 wt % sand showing its effect on pore sizes.

Flexural failure of sandwich panels is more complex to predict than solid beams and flexural strength depend on the mode of failure including face yielding, face wrinkling, core failure and debonding[15]. During the three-point bending tests, there were no visible signs of premature failure of the core or interfacial debonding indicating good material integrity and absence of large defects. The sandwich panels fail under tensile stress at the bottom surface staring from ductile failure of the HDPE skins and propagating through the core.

Flexural stiffness of sandwich panels reflects their resistance to bending deformation and varies with modulus of the core, the skins and geometric factors[15]. For a symmetric sandwich panel with rectangular section such as those in this work, the bending stiffness of the sandwich beam, D, can be expressed as[15]:

$$\frac{D}{b} = \frac{E_s t^3}{6} + \frac{E_s t d^2}{2} + \frac{E_c c^3}{12}$$

where E_s and E_c are the elastic modulus of the skin and core materials, respectively; b is width of the panel; d is distance between centroids of the skins, t and c are thicknesses of the skin and the core, respectively. The first and third terms describe the

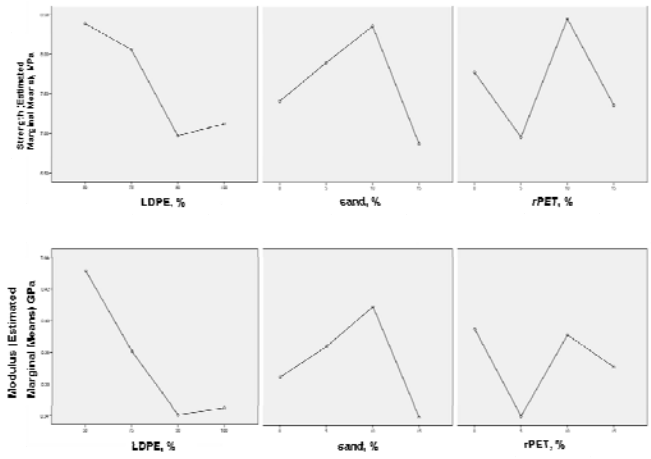

Fig. 8: Effect of control factors, LPDE/HDPE ratio, sand, and rPET on estimated marginal means of flexural strength (top) and modulus (bottom) of the PIM samples.

bending stiffness of the skins and of the core about their centroids. Together, they give the stiffness of the panel if there was no bonding between the skins and the core. The mid term describes the bending stiffness of the skins about the centroid of the panel itself.

Statistic significance of influences of the three core composition factors (the LDPE/HDPE ratio and wt% of sand and rPET) on flexural strength and modulus are shown in Fig. 8. Both flexural strength and modulus decrease with the increase of LDPE/HDPE ratio which is consistent with the results for solids shown in Fig. 3. Up to ~10 wt%, increase of sand is beneficial for both strength and stiffness. This may be attributable to the refinement of the pore structure shown in Fig. 7 which leads to increase in the foam properties. The trend for the down-turn of properties at high sand content is not yet fully understood but it was observed that at high sand percentage, pores in the vicinity of sand particles coalesced and formed bigger voids which could contribute to the weakening of the core properties. It may be attributable to enbrittlement effect of the sand particles. Addition of the rPET particles does not seem to have a consistent trend of influence. However most of the prediction points give reasonable strength and modulus. Further work will be necessary by look into the effect of rPET content alone by fixing values of the rest parameters.

4. Conclusions

Impurities in PE-rich feedstock (with minimum 70 wt% mixed PE) from recycled plastics was simulated with 3 variables in the compositions for the core: the LDPE/HDPE ratio simulating mix of LDPE (e.g. films) and HDPE (e.g. from milk bottles); sand (up to 15 wt%) simulating contamination from soil and rPET (up to 15 wt %) simulating PET residues in the PE-rich stream. A system of formulations was designed following an orthogonal design method to study the influence of them on flexural properties of the PIM sandwich panels.

All formulations could be moulded to satisfaction and the sand and rPET impurities did not hinder foaming of the core during the PIM process. The increase of LDPE/HDPE ratio tends to produce softer and lower strength panels. The PE-rich system can accommodate ~10 wt % sand which was found beneficial for refining the pore structure and enhancing the flexural properties. At higher levels, the properties tend to decrease due probably to formation of relatively larger voids near sand particles. No clear trend was observed for formulations containing up to 15 wt% rPET but no advert effect on foaming was observed and reasonable flexural properties were achieved. Further work is necessary to identify the actual limits of inclusion for sand and PET powder for a chosen LDPE/HDPE ratio.

This work demonstrated that the PIM process can accommodate considerable impurities in PE-rich core in forms of mixing of HDPE and LDPE, rPET residues or soil contamination. The variation of flexural properties can be predicted or controlled through monitoring of impurities. There exist significant scopes for reduction of the degree of sorting and cleaning in recycling systems by using lower grades of recyclates and for reduction of the associated costs and energy consumption.

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