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Polymer Testing 25 (2006) 668–676

POLYMER TESTING

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Material Properties

Properties of lignocellulosic material filled polypropylene bio-composites made with different manufacturing processes

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Received 2 February 2006; accepted 17 March 2006

Abstract

Using polypropylene as the matrix polymer and rice-husk flour and wood flour as the reinforcing filler, the tensile and Izod impact properties of lignocellulosic filler reinforced polypropylene bio-composites made with different extruding systems were examined by assessing their mechanical properties and the morphology of the fracture surfaces. The test samples were injection molded, in order to determine the mechanical and morphological properties of the bio-composites made with two different extruding systems and at different filler loadings. The tensile strength and modulus of the bio-composites fabricated using the twin-screw extruding system were improved as compared with those fabricated using the single-screw extruding system, due to the improved dispersion of the fillers in the composite. There was no difference in the Izod impact strength of the composites fabricated using the twin-screw and single-screw extruding systems, the similar impact strength of both samples with different extruding processes might be due to the fact that impact test is not discriminating enough to reveal the difference in dispersion status of the present composites. The SEM micrographs revealed well-dispersed fillers on the fracture surfaces of the test samples fabricated using a twin-screw extruding system.

Keywords: Extruding system; Mechanical properties; Morphology; Dispersion

1. Introduction

Lignocellulosic material filled thermoplastic polymer composites have been widely used since the 1980s, with the majority of these materials being wood-plastic composites (WPCs) in the initial stages. Since then, however, forestry resources have declined, and this has encouraged the development of substitutes for wood as the source of these lignocellulosic materials. These substitute materials include many plants similar to wood such as rice-husk, bagasse, rice straw, coconuts, etc. The chemical components and contents of these materials are similar to those of wood. Nowadays, these lignocellulosic materials are being used more and more in the form of fibers or particles [1,2]. These natural fillers have many positive advantages, such as their lightweight, low cost, reduced abrasion of machinery, non-toxicity, etc. With the growth of these WPC industries, much research has gone into

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0142-9418/$ - see front matter
doi:10.1016/j.polymertesting.2006.03.013

both the materials used, viz. the reinforcing fillers, matrix polymers and additives, and the manufacturing process itself. In the manufacturing process, there are many parameters which have an impact on the properties of the final products.

There are various methods of manufacturing lignocellulosic material-thermoplastic polymer composites, depending on the processing technique which is employed. These techniques can be roughly divided into two main processes, the non-woven web process and the melt-blending process, of which the most active research is currently focused on the manufacture of melt-blended composites. A single- or twin-screw type extruder is used in this melt-blending process to blend the thermoplastic polymer with natural fillers such as wood flour, rice-husk flour, various fibrous materials, etc. The single-screw extruder system was developed in the early days of the plastic industry and the screw does not have a regularly shaped mixing part. This system requires the pre-blending of the filler and matrix polymer, and this pre-blended mixture is not precisely mixed in the pre-blender and extruder, because the single-screw can only feed the melted mixture into the die. The twin-screw extruder system, on the other hand, has both a feeding zone (conveying zone) and a mixing zone [3], which means that the screw has a multiplex shape. In this system, the fillers are very well blended with the matrix polymer, because the mixture passes through a number of mixing blocks. In this system, the screw can feed and mix the melted mixture simultaneously [4], and the extruded strands and pellets have an excellent degree of dispersion of the fillers [3,5]. The degree of dispersion of the filler has a strong influence on the products, affecting their physical, mechanical and morphological properties.

In this study, we examined the relationship between the type of extruding system, single- or twin-screw, and the composite properties, focusing on the effects of the extruding system on the properties of lignocellulosic filler-polypropylene composites.

2. Experimental procedure

2.1. Materials

2.1.1. Matrix polymer

The thermoplastic polymer polypropylene (PP) was supplied by Hanwha L&C Corp., South Korea, in the form of homopolymer pellets with a density of 0.91 g/cm^3 and a melt flow index of 12 g/10 min (230 °C/2160 g).

2.1.2. Reinforcing filler

The lignocellulosic materials used as the reinforcing filler in the composite in order to obtain the comparative data, were rice-husk flour (RHF) and wood flour (WF), with particle sizes of 80–100 mesh. The RHF and WF were both supplied by Saron Filler Co., South Korea. The chemical constituents of the fillers are shown in Table 1.

2.1.3. Compatibilizing agent

The compatibilizing agent, MAPP (maleated polypropylene), was obtained from Eastman Chemical Products, Inc., in the form of Epolene G-3003^{TM} which has an acid number of 8 and a molecular weight of 103,500.

2.2. Sample preparation

The RHF and WF were oven dried at 100 °C for 24 h and then stored over a desiccant in sealed containers. The polypropylene was pre-blended with the lignocellulosic fillers and compatibilizing agent in a two-roll rheomixer. Mixing was continued at 200 °C for 15 min at a rotor speed of 20 rpm. A laboratory-scale single-screw extruder was employed to compound the pre-blended mixture. The extruded strand was quenched in a water bath and then pelletized and stored in sealed packs containing a desiccant. The process using the single-screw extruding system is shown in Fig. 1.

A laboratory-scale twin-screw extruder was employed to compound the RHF and WF with the polypropylene, the latter being used as a matrix polymer. The process using the twin-screw extruding system and the typical twin-screw configuration are shown in Fig. 2. The PP was blended with the lignocellulosic fillers and the compatibilizing agent. The extruded strand was quenched in a water bath and then pelletized and stored in sealed packs

Table 1
Chemical constituents of the lignocellulosic fillers (rice-husk flour and wood flour)

	Holocellulose	Lignin	Ash	Others
RHF[a]	59.9	20.6	13.2	6.5
WF[a]	62.5	26.2	0.4	10.9

Values are percentage by weight.
[a]Spec. from Saron Filler Co.

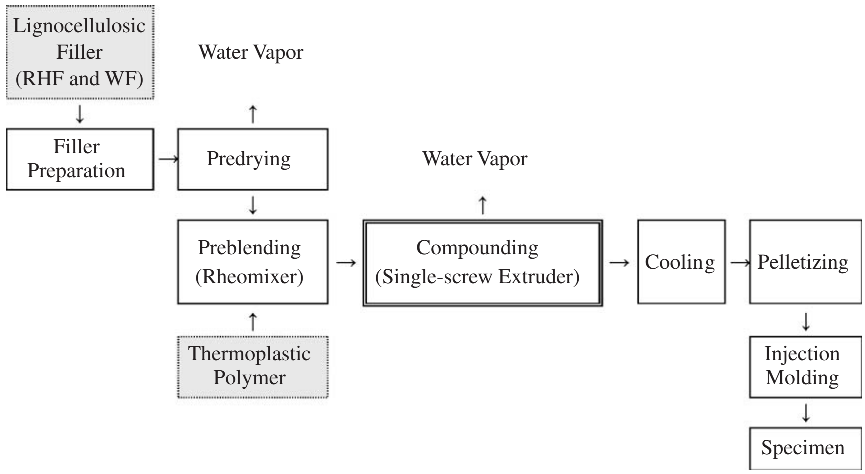


Fig. 1. Compounding process using single-screw extruding system.

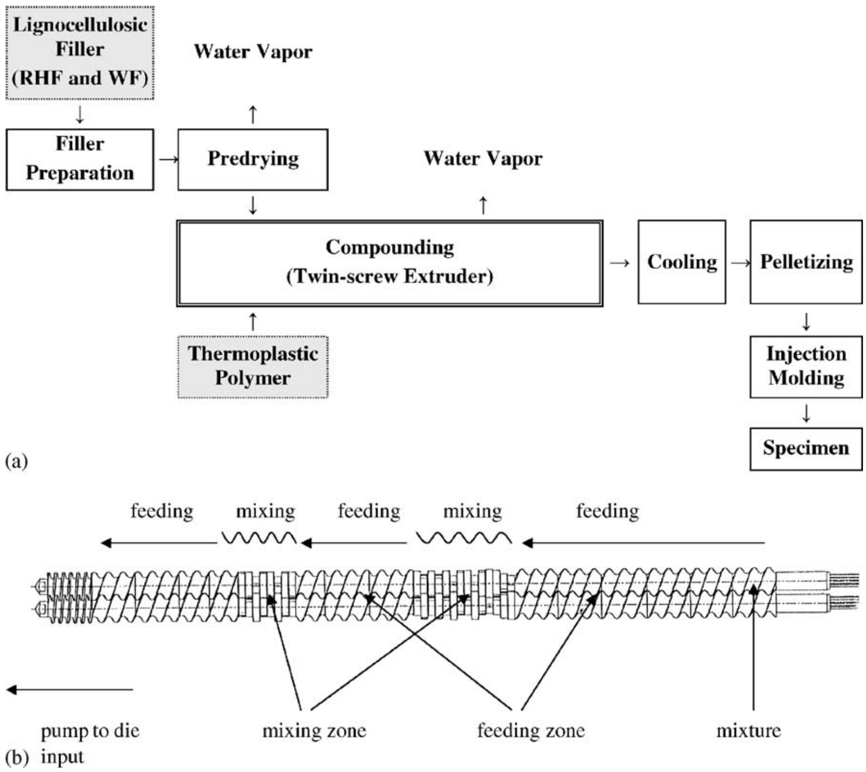


Fig. 2. Compounding process using twin-screw extruding system and typical twin-screw configuration.

containing a desiccant. Four levels of filler loading (10, 20, 30 and 40 wt%) and a compatibilizing agent content of 3 wt% were used in the sample preparation.

The tensile and Izod Impact test specimens were injection molded at 200 °C, an injection pressure of 1200 psi and a mold pressure of 1500 psi. After being molded, the test specimens were conditioned before testing at 23 ± 2 °C and 50 ± 5% RH for at least 40 h according to ISO 291-2005 [6].

2.3. Tensile test

The tensile tests were conducted according to ISO 527-2005 [7] with a Universal Testing Machine

(Instron Co.). The tests were performed at a crosshead speed of 100 mm/min and at room temperature. Each value obtained represented the average of five samples.

2.4. Izod impact test

The notched and unnotched Izod impact strength tests were conducted according to ISO 180-2000 [8] at room temperature. Each value obtained represented the average of five samples.

2.5. Morphology

Studies on the morphology of the tensile and Izod impact fracture surfaces of the composites were carried out using a JSM-5410 LV (JEOL Co. Ltd.) scanning electron microscope.

3. Results and discussion

3.1. Tensile properties with different extruding systems

The tensile strengths of the composites made of lignocellulosic filler-polypropylene are shown in Fig. 3 as a function of the filler loading and the type of extruding system. The tensile strengths of the composites slightly decreased with increasing filler loading, due to the poor interfacial bonding and the presence of agglomerate fillers, which is the same tendency as that reported in a previous study [2]. The weak bonding between the hydrophilic filler and the hydrophobic matrix polymer obstructs the stress propagation, and causes the tensile strength to decrease as the filler loading increases [2]. In addition, poor dispersion causes agglomeration of the fillers, as well decreasing the tensile properties. The tensile strength of the composites made with the twin-screw extruding system was improved compared with those made with the single-screw extruding system, due to the better dispersion of the filler in the matrix polymer. To improve the interfacial bonding strength between the filler and the matrix polymer, a compatibilizing agent, maleated polypropylene (MAPP), was used. The tensile strengths of the 30 wt% lignocellulosic filler filled polypropylene composites at a compatibilizing agent content of 3 wt% show the same tendency according to the type of extruding system as those made without the compatibilizing agent. The force-elongation curves of the rice-husk flour filled

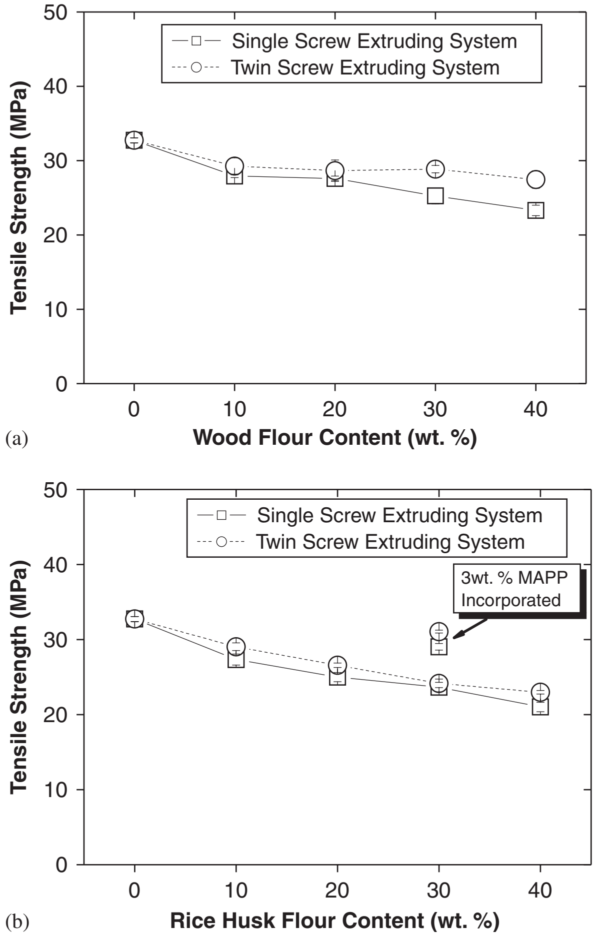
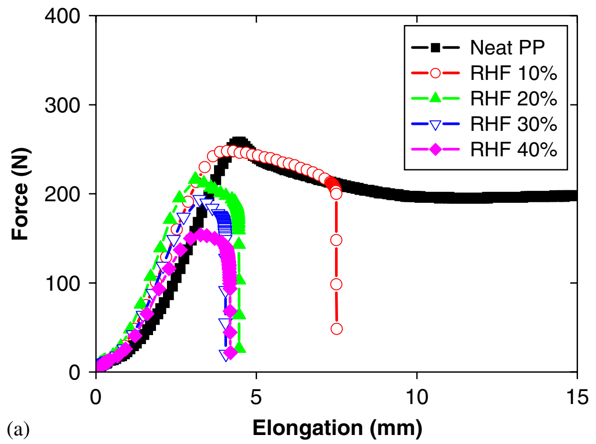


Fig. 3. Tensile strengths of the lignocellulosic filler-polypropylene composites made with the two extruding systems at various filler loadings: (a) WF-PP composite, and (b) RHF-PP composite.

polypropylene composites according to the filler loading and type of extruding system are shown in Fig. 4. The samples made with the twin-screw extruding system present not only improved tensile strength, but also improved tensile modulus, due to the better stress propagation. Poorly dispersed fillers can easily agglomerate, and this obstructs stress propagation. On the other hand, well-dispersed fillers prevent the obstruction of stress propagation, thus allowing the stress to be well propagated [9,10].

The force-elongation curves of the 30 wt% rice-husk flour filled PP composites (3 wt% of compatibilizing agent incorporated) are shown in Fig. 5 for each type of extruding system. The compatibilizing agent has a positive effect on the tensile properties,



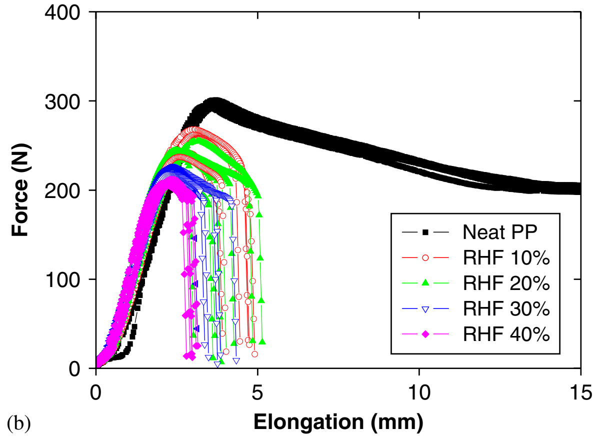


Fig. 4. Force-elongation curves of the RHF-PP composites made with the two extruding systems at various filler loadings: (a) single-screw system and (b) twin-screw system.

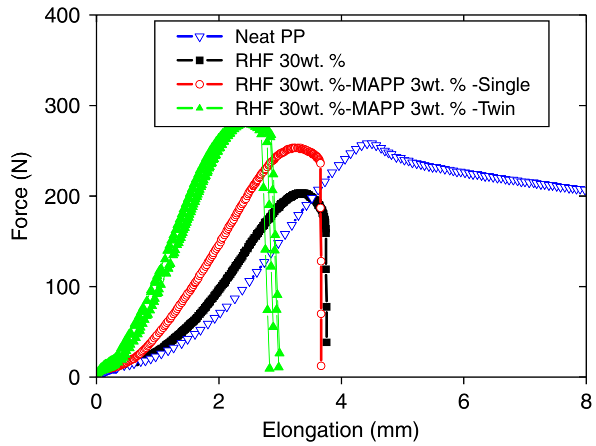
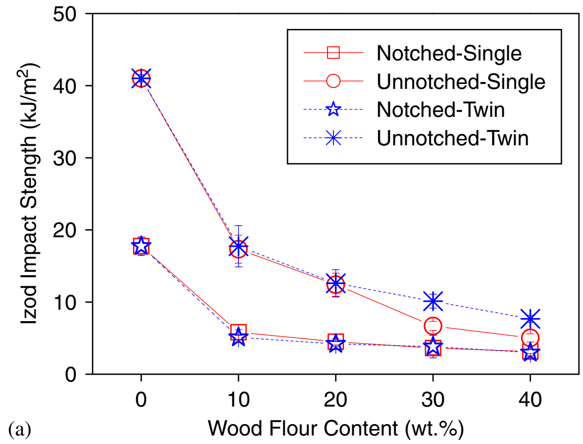


Fig. 5. Force-elongation curves of the 3 wt% MAPP incorporated 30 wt% RHF-PP composites made with the two different extruding systems.

due to the strong interfacial bonding between the filler and the matrix polymer [1,2], and the tensile strength and modulus of the MAPP incorporated composite made with the twin-screw extruding system were significantly improved as compared with those of the composite made with the single-screw extruding system, as shown in Fig. 5.

3.2. Impact properties with different extruding systems

The Izod impact tests were conducted at room temperature. The notched and unnotched specimens were tested and Fig. 6 shows the Izod impact strengths of the lignocellulosic filler-PP composites made with the different extruding systems. The Izod impact strengths of the composites made with the two different extruding systems are almost the



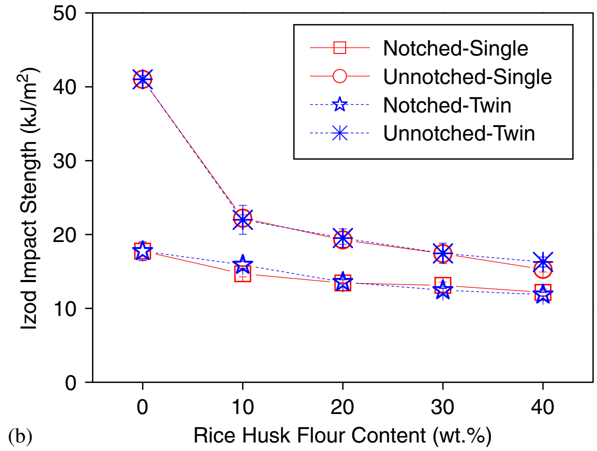


Fig. 6. Izod impact strengths of the lignocellulosic filler filled PP composites made with the two different extruding systems: (a) WF-PP composite and (b) RHF-PP composite.

same. The matrix polymer's resistance to crack propagation has an influence upon notched impact strength, while unnotched impact strength will be affected by the energy consumed due to plastic flexural deformation prior to crack initiation. The degree of dispersion of the fillers might influence the notched impact performance, but the similar impact strength of both samples with different extruding processes might be due to the fact that impact test is not discriminating enough to reveal the difference in dispersion status of the present composites. The notched specimens exhibited lower impact strengths than the unnotched specimens, which is the same tendency as that reported in a previous study [2], the impact properties are affected by the crack initiation and crack propagation mechanism between the filler and the matrix polymer. The Izod impact strengths of the MAPP incorporated RHF-PP composites made with the different extruding systems show the same tendency as those made without any compatibilizing agent, as shown in Fig. 7. The MAPP incorporated sample exhibited almost the same impact strength as the sample made without any compatibilizing agent, which is the same tendency as that reported in a previous study [2].

3.3. Morphology

The tensile fracture surfaces of the RHF-PP composites made with the two different extruding systems at filler loadings of 10 and 30 wt% and a compatibilizing agent (MAPP) content of 3 wt% are shown in Fig. 8. In the case of the composite made

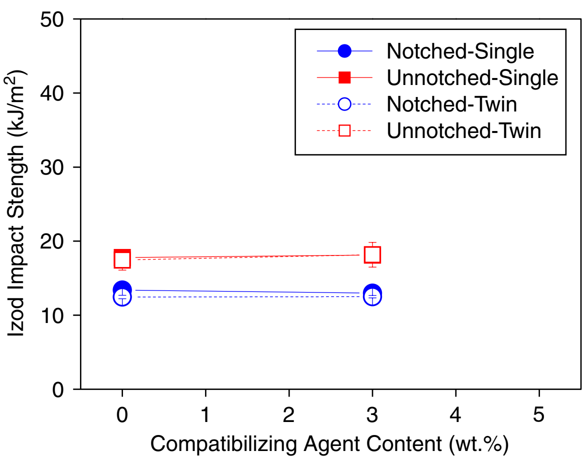


Fig. 7. Izod impact strengths of the RHF-PP composites made with the two different extruding systems at a compatibilizing agent content of 3 wt%.

with the twin-screw extruding system without any compatibilizing agent and at a filler loading of 10 wt%, the filler particles are well dispersed in the matrix polymer, as compared with the composite made with the single-screw system, but some cavities are to be seen where the filler has been pulled-out. The presence of these cavities means that the interfacial bonding between the filler and the matrix polymer is poor and weak [2]. In the composite made without any compatibilizing agent the fillers are not fractured, whereas in the composite made with compatibilizing agent, the interfacial bonding between the filler and the matrix polymer is strong, and the fracture occurred in the filler itself. This means that the stress is well propagated between the filler and the matrix polymer in the composite incorporating the compatibilizing agent, causing it to have a higher tensile strength and modulus. A few traces of filler particles, either pulled out or fractured filler particles, are to be seen in the micrographs of the composites containing compatibilizing agent. All of the micrographs of the samples made with the twin-screw extruding system show well dispersed fillers, as compared with those made with the single-screw extruding system [5]. The unnotched Izod impact fracture surfaces at the notched tip of the composites made with the two different extruding systems at a filler loading of 30 wt% and a MAPP content of 3 wt% are shown in Fig. 9. Pulled-out traces of filler particles are to be seen in the samples made without any compatibilizing agent, and this is due to the weak bonding between the filler and the matrix polymer. Fractured filler particles are to be seen in the micrographs of the MAPP incorporated composites, demonstrating the increased brittleness of the composite, which caused it to have a slightly decreased impact strength. All the micrographs of the samples made with the twin-screw extruding system show well dispersed fillers, as compared with those made with the single-screw extruding system.

4. Conclusion

The tensile properties of the composites made with the twin-screw extruding system were better than those of the composites made with the single-screw extruding system, due to the improved dispersion of the filler. The tensile strength and modulus of the lignocellulosic filler-PP composite made with the twin-screw extruding system

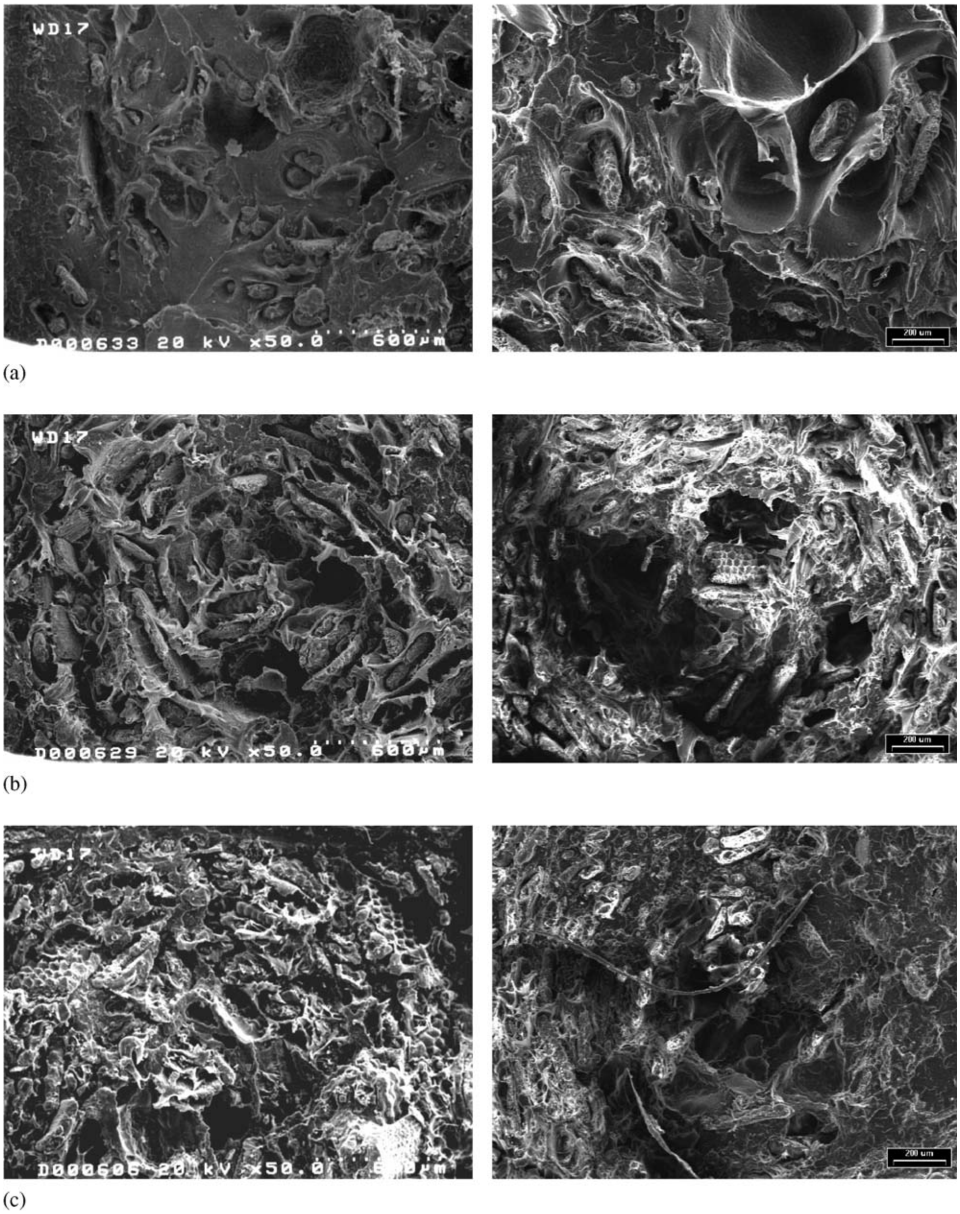


Fig. 8. Tensile fracture surfaces of the RHF-PP composites made with the two different extruding systems: (a) 10 wt% of RHF-PP (left-single screw system, courtesy of Ref. [2], right-twin). (b) 30 wt% of RHF-PP (left-single screw system, courtesy of Ref. [2], right-twin). (c) 30 wt% RHF-PP with 3 wt% MAPP (left-single screw system, courtesy of Ref. [11], right-twin).

were improved in the case of the composite made without any compatibilizing agent and significantly improved in the case of the composite made with the compatibilizing agent, as compared with those made with the single-screw extruding system. The Izod impact strengths of the composites made with the two different extruding systems were almost the same, the degree of dispersion of the fillers might influence the notched impact performance, but the similar impact strength of both samples with different extruding processes might be due to the fact that impact test is not discriminating enough to reveal the difference in dispersion status of the present composites. All of the SEM micrographs of the fracture surfaces of the composites made with the twin-screw extruding system show well dispersed

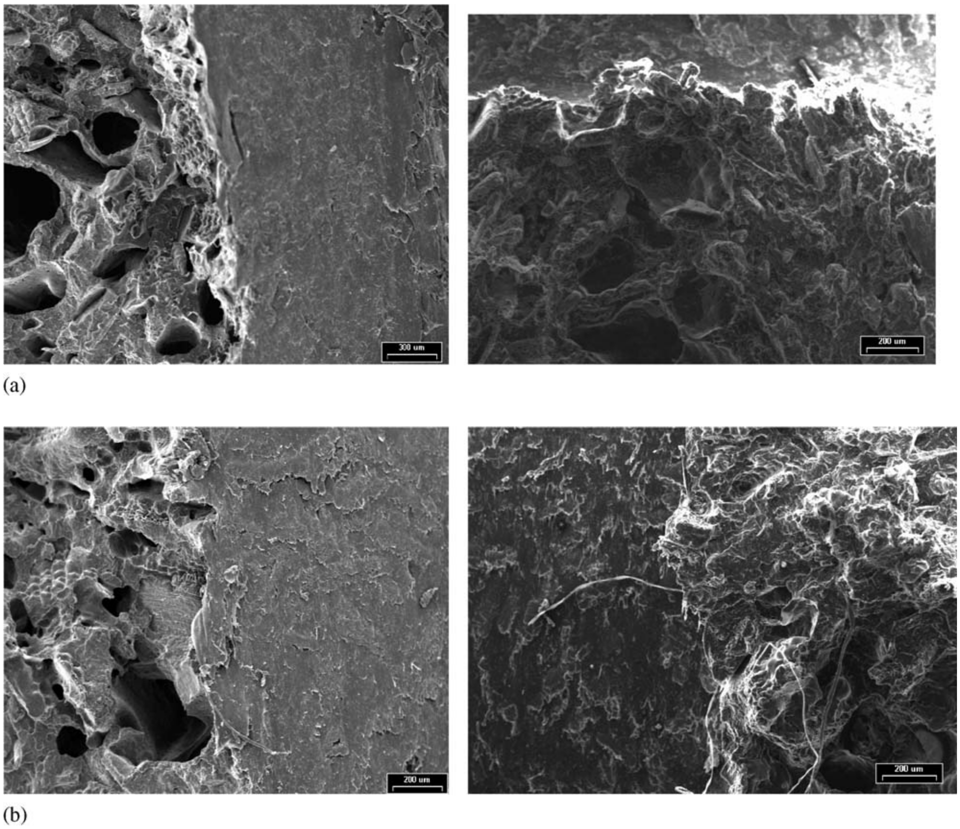


Fig. 9. Unnotched Izod impact fracture surfaces of the RHF-PP composites made with the two different extruding systems at the notched tip: (a) 30 wt% of RHF-PP (left-single screw system, courtesy of Ref. [2], right-twin). (b) 30 wt% RHF-PP with MAPP (left-single screw system, courtesy of Ref. [11], right-twin).

fillers, as compared with those made with the single-screw extruding system.

Acknowledgement

This work was supported by a Korea Research Foundation Grant funded by the Korean Government (MOEHRD, Basic Research Promotion Fund) (M01-2004-000-20097-0) and Hee-Soo Kim and Sumin Kim are grateful for the graduate fellowship provided by the Ministry of Education through the Brain Korea 21 Project.

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