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# Rice-husk flour filled polypropylene composites; mechanical and morphological study

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**Abstract**

To determine the possibility of using lignocellulosic materials as reinforcing fillers in the thermoplastic polymer composite, polypropylene as the matrix and rice-husk flour as the reinforcing filler were used to prepare a particle-reinforced composite in order to determine testing data for the physical, mechanical and morphological properties of the composite according to the filler loading in respect to thermoplastic polymer. In the sample preparation, four levels of filler loading (10, 20, 30 and 40 wt.%) were designed. In the tensile test, six levels of test temperature (−30, 0, 20, 50, 80 and 110 °C) and five levels of crosshead speed (2, 10, 100, 500 and 1500 mm/min) were designed. Tensile strengths of the composites slightly decreased as the filler loading increased. Tensile modulus improved with increasing filler loading. Notched and unnotched Izod impact strengths were lowered by the addition of rice-husk flour. The composite became brittle at higher crosshead speed, and showed plastic deformation with increasing test temperature.


*Keywords:* Lignocellulosic material; Thermoplastic polymer; Particle-reinforced composite; Mechanical properties; Plastic deformation

## 1. Introduction

These days various synthetic polymers are being prepared combined with various reinforcing fillers in order to improve the mechanical properties and obtain the characteristics demanded in actual application. Studies are ongoing to find ways to use lignocellulosic fibers in place of synthetic fibers as reinforcing fillers. These natural fillers are especially being sought since the production of composites using natural substances as reinforcing fillers is not only inexpensive but also able to minimize the environmental pollution caused by the characteristic biodegradability [5], enabling these composites to play an important role in resolving future environmental problems. The need for materials that are non-toxic to the human body and have appropriate characteristics for specific purposes is ever increasing due to the lack of resources and increasing levels of environmental pollution. Thus, research is proceeding to develop composites using various recycled wastes [12,13], especially in developing composites using most environmentally friendly agro-wastes (lignocellulosic materials) as reinforcing fillers and thermoplastic polymers as matrixes. The convenience of these composites lies in the fact that the ingredients are obtained easily from natural wastes and hence the composites can be made relatively easily. They can be used to resolve environmental problems and to produce products with various physical properties and effective functions. Lignocellulosic materials as reinforcing fillers in plastics, in place of the previously used inorganic substances and synthetic fibers, offer a major benefit in terms of environmental protection. The benefits offered by lignocellulosic materials include making the final product light [9], decreasing the wear of the machinery used, low cost, biodegradability [5], and absence of residues or toxic byproducts.

In the present study we used a thermoplastic polymer (polypropylene) as the matrix and a lignocellulosic material (rice-husk flour) as the reinforcing filler to prepare a particle-reinforced composite to examine the possibility

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of using lignocellulosic materials as reinforcing fillers and to determine testing data for the physical, mechanical and morphological properties of the composite according to the reinforcing filler content in respect to thermoplastic polymer.

## 2. Experimental

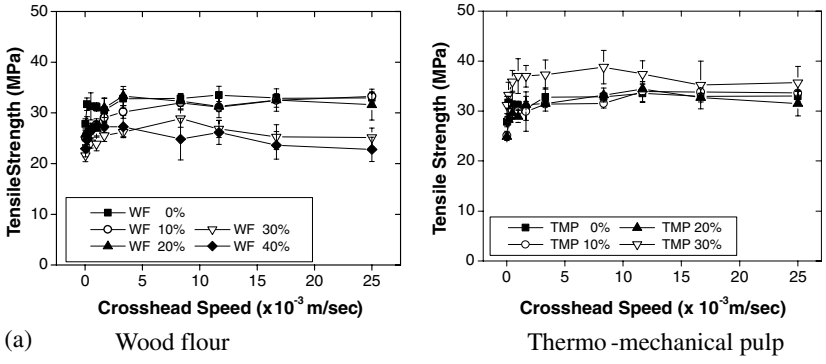
### 2.1. Materials

The thermoplastic polymer polypropylene 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). The reinforcing filler in the composites was rice-husk flour (RHF), and to obtain the comparative tensile strength data wood flour (WF) and thermo-mechanical pulp (TMP) were also used as the filler; the particle sizes were 80–100 mesh. RHF and WF were supplied by SARON FILLER Co., South Korea. The chemical constituents of the fillers are shown in Table 1.

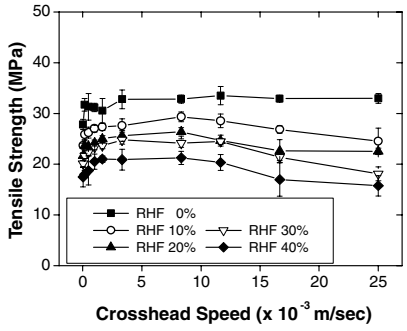
Table 1
Chemical constituents of the fillers (rice-husk flour, wood flour and rice-husk powder)

| | Holocellulose | Lignin | Ash | Others |
|---|---|---|---|---|
| RHF[a] | 59.9 | 20.6 | 13.2 | 6.5 |
| WF[a] | 62.5 | 26.2 | 0.4 | 10.9 |
| RHP[b] | 60 | 20 | 17 | 3 |

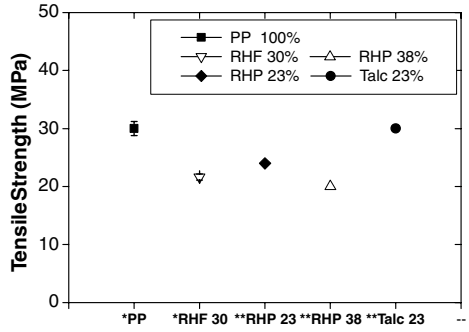
Values are percentage by weight.
[a] Specified from SARON FILLER Co.
[b] Rice-husk powder from Ref. [5].




(a) Wood flour Thermo -mechanical pulp




Rice-husk flour




(b)

* : Our results, ** : Results of reference [5] (rice-husk powder and talc)

Fig. 1. (a) Tensile strengths of the composites made of lignocellulosic filler (RHF, WF and TMP) and polypropylene matrix at different filler loadings and crosshead speeds. (at room temp.); (b) tensile strengths of the composites made of lignocellulosic and inorganic filler.

### 2.2. Sample preparation

RHF was dried at 100 °C for 24 h to adjust it to a moisture content of 1–2% and then stored over desiccant in sealed containers. The polypropylene was blended with RHF in a two-roll rheomixer. Mixing was continued at 200 °C for 15 min at a rotor speed of 20 rpm. A laboratory-size, twin-screw extruder was employed to compound the RHF with the polypropylene as a matrix polymer. The extruded strand was pelletized and stored in sealed packs containing desiccant. Four levels of filler loading (10, 20, 30 and 40 wt.%) were designed in the sample preparation. Tensile and Izod impact test specimens were prepared using an injection molding machine at 200 °C, an injection pressure of 1200 psi, and a device pressure of 1500 psi. After molding, test specimens were conditioned before testing at $23 \pm 2$ °C, $50 \pm 5\%$ RH for at least 40 h according to ASTM D 618-99 [2].

### 2.3. Mechanical testing

Tensile tests were conducted according to ASTM D 638-99 [2] with a Universal Testing Machine (Zwick Co., NICEM at Seoul National University). The tests were performed at crosshead speeds of 2, 10, 100, 500 and 1500 mm/min, and temperatures of −30, 0, 20, 50, 80 and 110 °C. Notched and unnotched Izod impact strength tests were conducted according to ASTM D 256-97 [1] at room temperature. Each value obtained represented the average of five samples.

### 2.4. Morphological study

Studies on the morphology of the tensile and Izod impact fracture surfaces of the composites were carried out using a JEOL-5410 LV scanning electron microscope (NICEM at Seoul National University). SEM micrograph magnifications were ×50, ×75 and ×200.

## 3. Results and discussion

### 3.1. Tensile property

The tensile strengths of the composites made of lignocellulosic filler (RHF, WF and TMP) and polypropylene (PP) matrix at different filler loadings and crosshead speeds are shown in Fig. 1(a). The tests were conducted at room temperature. TMP–PP composites showed higher strength than WF–PP and RHF–PP composites. Generally, fiber-reinforced composites have higher tensile strength than particle-reinforced composites [4]. Tensile strengths of RHF–PP composites slightly decreased with increasing filler loading [6–8] and presented the peak value at a crosshead speed of 500 mm/min. As the filler loading increased, thereby increasing the interfacial area, the worsening interfacial bonding between filler (hydrophilic) and matrix polymer (hydrophobic) decreased the tensile strength, which nevertheless remained within acceptable levels [5] according to Fig. 1(b). Tensile modulus improved with increasing filler loading [7,8]. For irregularly shape fillers, the strength of the composites decreases due to the inability of the filler to support stresses transferred from the polymer matrix [7] while poor interfacial bonding causes partially separated micro-spaces between filler and matrix polymer, which obstructs stress propagation when tensile stress is loaded and induce increased brittleness.

The stress–strain curves of composites at different filler loadings (crosshead speed: 10 mm/min, at room temperature) and crosshead speeds (filler loading: 20 wt.%, at room temperature) are shown in Fig. 2. As the filler loading and crosshead speed increased, the composite became more brittle [5]. As the filler loading

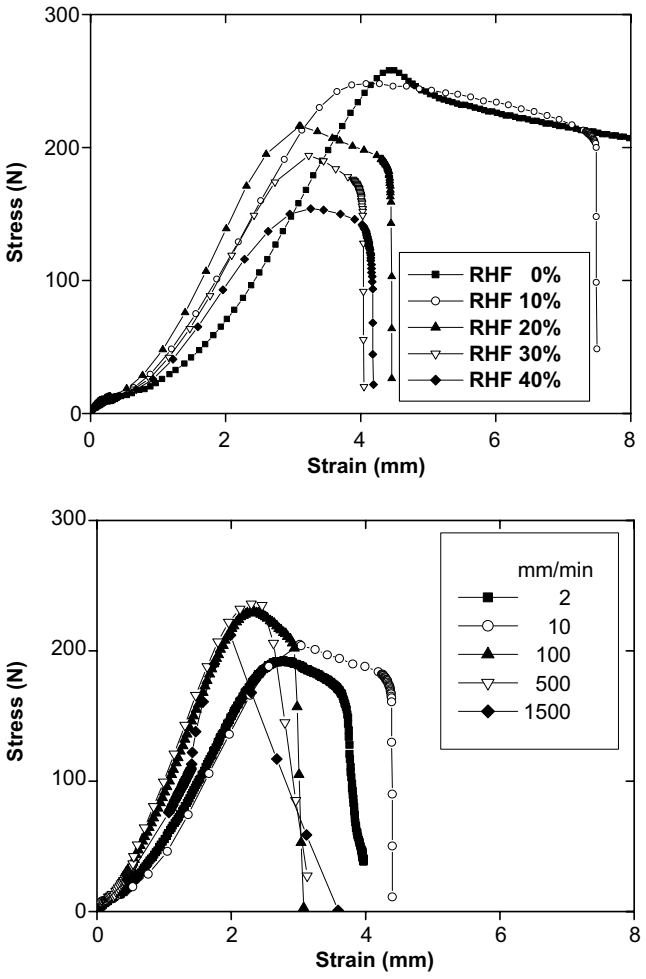



Fig. 2. Stress–strain curves of the composites at different filler loadings (crosshead speed: 10 mm/min, at room temp.) and crosshead speeds (filler loading: 20 wt.%, at room temp.).

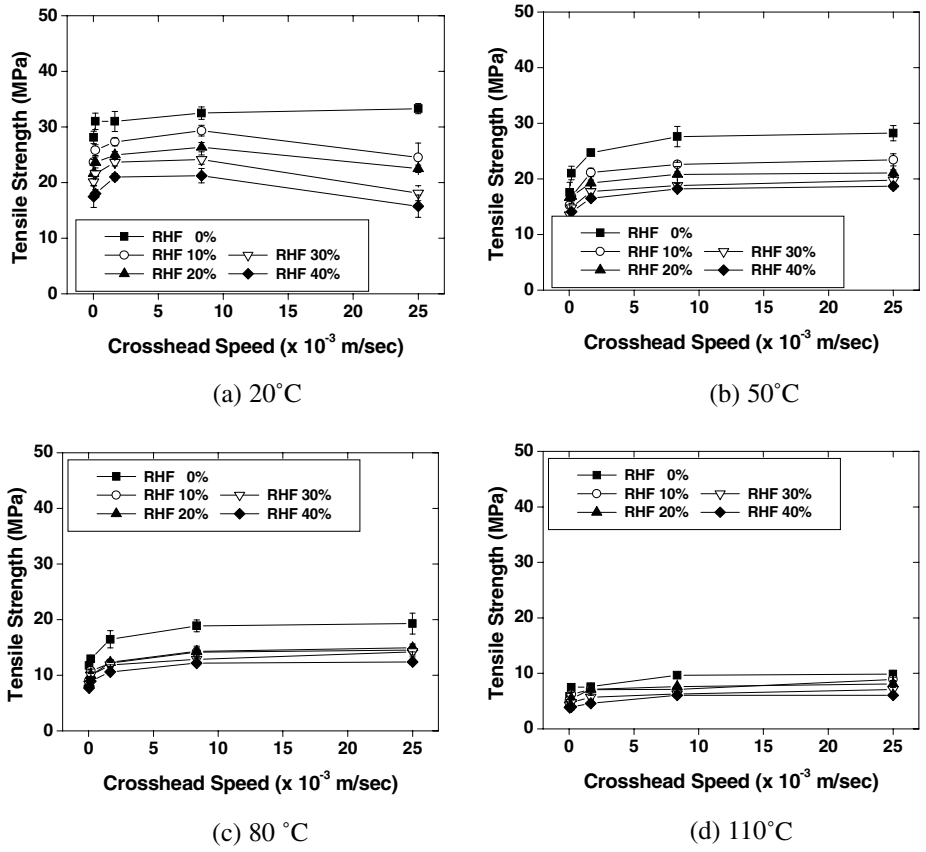



Fig. 3. Tensile strengths of the composites at different filler loadings, crosshead speeds and test temperatures.

increased, the fraction of thermoplastic polymer decreased and interfacial area increased, which increased the brittleness as described above. As the crosshead speed increased, the frictional resistance between filler and matrix polymer decreased, and this increased the brittleness. The nonlinearity in the curves is mainly due to the plastic matrix deformation.

The tensile strengths of the composites at different filler loadings, crosshead speeds and test temperatures are shown in Fig. 3, and at different test temperatures and filler loadings are shown in Fig. 4 (crosshead speed: 10 mm/min). The tensile strengths according to the filler loadings presented the same tendency at each test temperature. At the lower test temperatures (−30 and 0 °C), the composites exhibited strong and brittle properties like glass, but the tensile strength drastically decreased as the test temperature was increased from 0 to 20 °C due to the glass transition of the matrix polymer (polypropylene) [10]. The matrix polymer became more ductile and softened as the test temperature increased. Stress–strain curves of the composites at different test temperatures (crosshead speed: 10 mm/min, filler loading: 20 wt.%) are shown in Fig. 5. The tensile strength and tensile modulus of the composites decreased as the test temperature increased because the thermoplastic

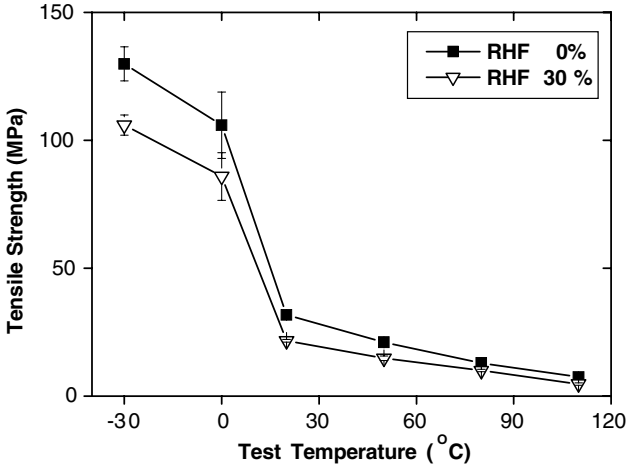



Fig. 4. Tensile strengths of the composites at different test temperatures and filler loadings (crosshead speed: 10 mm/min).

polymer was softened at these increasing temperatures and the composite showed more ductility.

### 3.2. Izod impact strength

The notched and unnotched Izod impact strengths of the composites at different filler contents are shown in

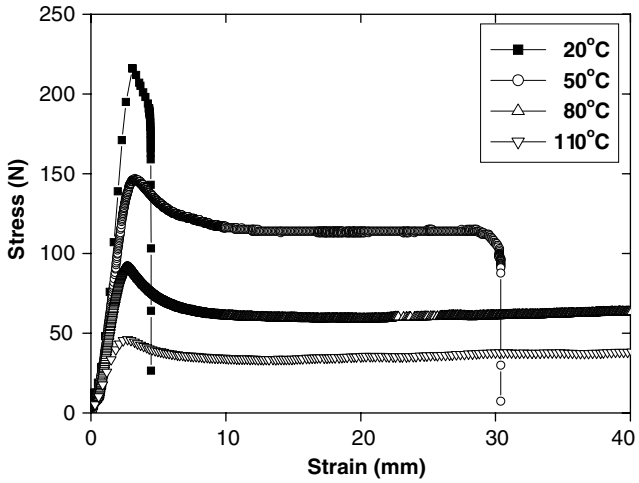



Fig. 5. Stress–strain curves of the composites at different test temperatures (crosshead speed: 10 mm/min, filler loading: 20 wt.%).

Fig. 6. The tests were performed at room temperature. The notched and unnotched Izod impact strength of the composites slightly decreased as the filler content increased [5]. In the case of notched samples, the impact strength decreased with increasing amounts of RHF added until a plateau was reached at a filler content of about 20 wt.%. Poor interfacial bonding induces microspaces between the filler and matrix polymer, and these cause numerous micro-cracks when impact occurs, which induce crack propagation easily and decrease the impact strength of the composites. Especially, the unnotched Izod impact strength of the composite made of 100 wt.% matrix polymer (polypropylene) showed significantly high impact strength, which was drastically decreased at a filler loading of 10 wt.%. The notched tip, which is the stress concentrating point, causes relatively low impact strength in the notched sample, thereby allowing the crack to propagate easily, and the same effect

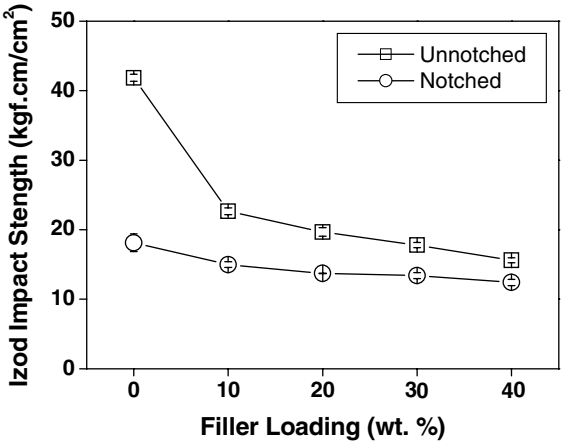



Fig. 6. Notched and unnotched Izod impact strengths of the composites at different filler loadings.

appeared at the filler-matrix interfacial area in the filler contained unnotched sample.

As can be seen in Fig. 6, unnotched Izod impact energies were considerably larger than notched Izod impact energies. This is due to the different fracture process for notched and unnotched samples. The unnotched impact behavior is controlled to a considerable extent by fracture initiation processes that, in turn, are controlled by stress concentrations at defects in the system. Notched impact behavior, meanwhile, is controlled to a greater extent by factors affecting the propagation of fracture initiated at the predominating stress concentration at the notched tip. In other words, unnotched Izod impact energies are not only a measure of crack propagation but also of crack initiation [3,11].

### 3.3. Morphological study

Fig. 7 shows the tensile fracture surfaces of the composites at different filler loadings. At a filler loading of 10 wt.% (Fig. 7(a)), a few filler particles are seen, the composite mainly representing plastic deformation. At 20 wt.% filler loading (Fig. 7(b)), slightly increased numbers of holes where filler particles have pulled out traces are seen, and at 30 wt.% filler loading (Fig. 7(c)), more traces are seen. At 40 wt.% filler loading (Fig. 7(d)), more filler particles are seen rather than polymer matrix, and the large amount of poor bonded interfacial area between filler and matrix polymer causes brittle deformation of the composite. Fig. 8 shows the notched and unnotched Izod impact fracture surfaces of the composites at 30 wt.% filler loading. SEM micrograph of the notched sample shows the clearly fractured surface at the notched tip as the stress concentrating point, which causes easy propagation of the crack when impact occurs. The unnotched sample shows an irregularly fractured surface, which is the interfacial area between filler and matrix polymer. The pulled-out traces of fillers can be seen in the SEM micrographs due to the poor interfacial bonding between filler and matrix polymer.

## 4. Conclusions

Tensile strengths of the composites slightly decreased while the tensile modulus improved as the filler loading and crosshead speed increased, but the composites had an acceptable strength up to a filler loading of 40 wt.%. RHF could be utilized as a biodegradable filler at end-of-use in polymeric materials to minimize environmental pollution rather than produce strong reinforcing filler. As the filler loading and crosshead speed increased, the tensile property became more brittle, and the poor interfacial bonding between filler and matrix polymer caused decreased tensile and Izod impact strength of the composites. Nevertheless, this problem could be reduced

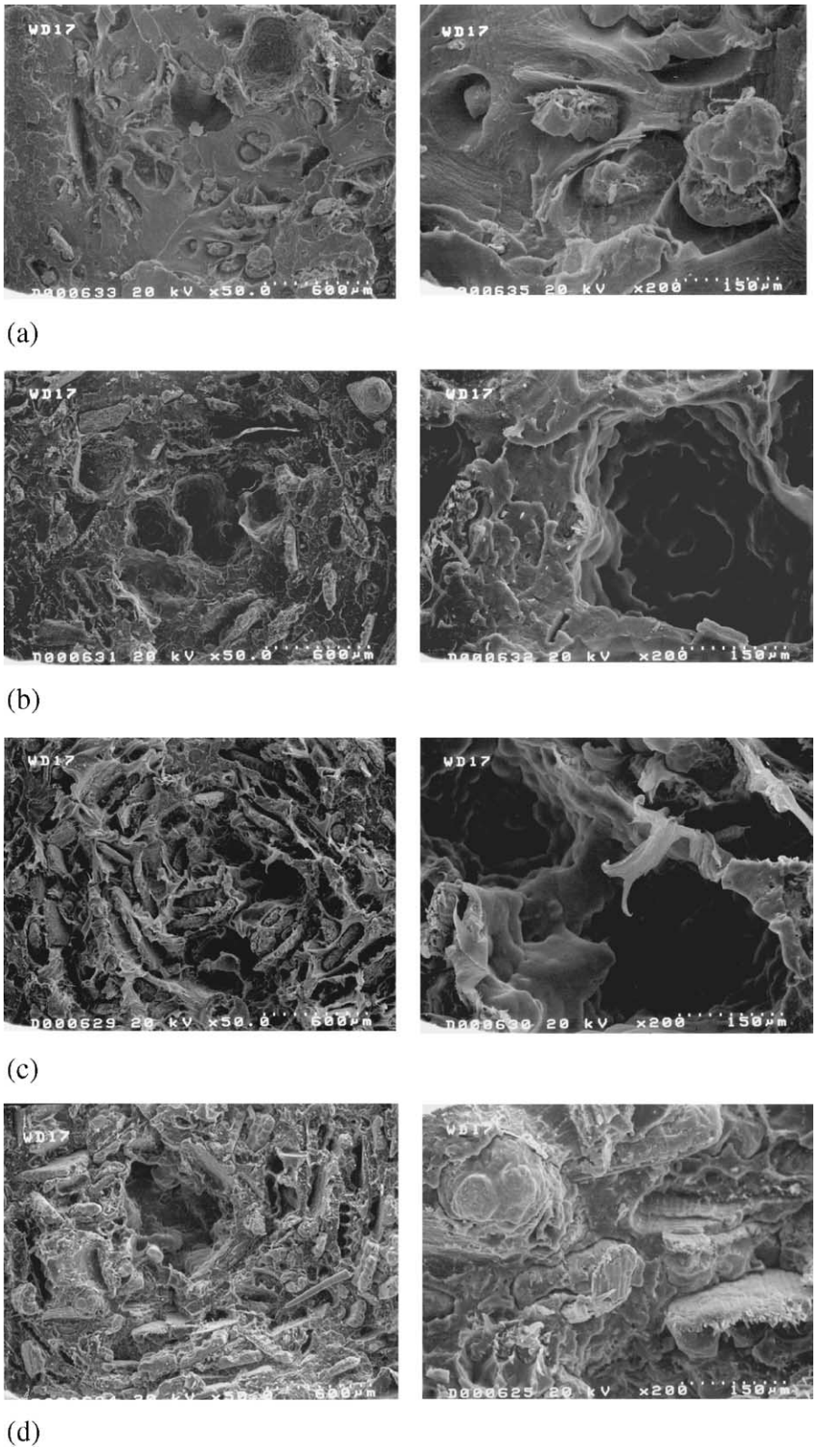
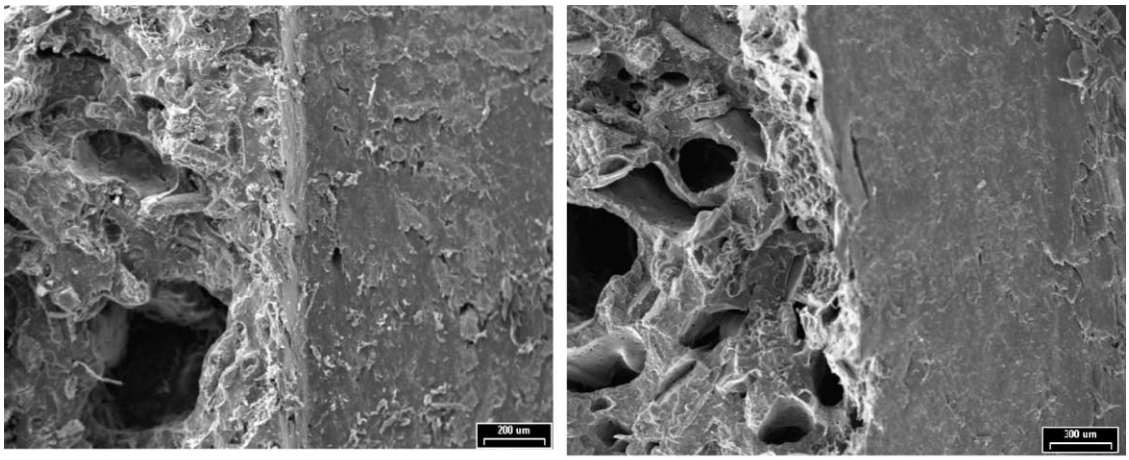



Fig. 7. SEM micrographs of the tensile fracture surfaces of the RHF–PP composites with different filler loadings. (a) 10 wt.% filler loading, (b) 20 wt.% filler loading, (c) 30 wt.% filler loading, (d) 40 wt.% filler loading.

by using a compatibilizing or coupling agent, and further extended research is suggested. As the test temperature increased, the thermoplastic polymer was softened and the composite showed plastic matrix de-




(a) Notched sample (b) Unnotched sample

Fig. 8. SEM micrographs of the notched and unnotched Izod impact fracture surfaces of the RHF–PP composites at 30 wt.% filler loading.

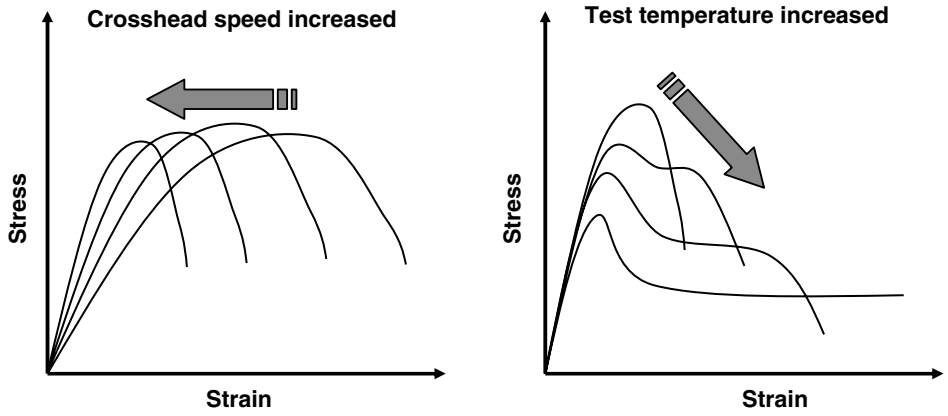



Fig. 9. Typical stress–strain curves of the composites according to various crosshead speeds and test temperatures in tensile tests.

formation, which decreased the tensile strength and modulus. Fig. 9 presents the typical stress–strain curves of the composites according to various crosshead speeds and test temperatures in the tensile tests. As the filler loading increased, morphological study revealed more filler particles and increased numbers of holes where filler particles have pulled out traces.

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