JOURNAL OF MATERIALS SCIENCE 39 (2004) 2465– 2472

Enhanced sandwich structure of powder-based composites

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The enhanced sandwich structure of powder-based composite with relatively high volume fraction of powders composed of fine densified skins and periodically localized densified core-material for improving their mechanical properties and formability is presented. The structurized composites are fabricated by rolling process with the aid of a novel dynamic three-dimensional rolling technique. This methodology with various actuating phases exhibits different characteristics of forming schemes to fabricate desired structure of composites and provides assorted effects on the forming properties. Consequently, the enhanced sandwich structures of the composite sheet enable the tensile strength and the failure strain to be controllable and improved significantly. In addition, the bendability and the flowability of the composite sheets meaningfully related to their formability are greatly enhanced. © *2004 Kluwer Academic Publishers*

1. Introduction

Biodegradable and compostable products, especially those made of renewable materials from agrowastes, are an essential innovation for the applications of environmental materials. The demands of the fabrications of various disposable containers or packaging with high biodegradability and compostability are urgently needed. Numerous attempts are now being made to produce fully biodegradable plastics by means of the use of various agrowastes, plant carbohydrates, plant protein, and vegetable oils. Meanwhile, all of those materials can be dissolved in storm sewers and oceans without affecting marine life or wildlife [1, 2].

The cost of today's biodegradable polymers is substantially higher than that of synthetic polymers. Moreover, their long-range performance has not been fully assessed because the development of biodegradable plastics is relatively new. Consequently, a mixture of agricultural waste including hulls from corn, wheat, rice, and soy (major component) and biodegradable polymers (minor component) is an attractively alternative choice [3, 4]. A biomass (starch/PVA) made of thermoplastic starch and polyvinyl alcohol (PVA) has been proposed by George [5]. The results showed that the mechanical strength, forming capability, and biodegradability of the starch-PVA material have been enhanced. Meanwhile, the biodegradable rate is almost the same as that for only the starch.

The biodegradable rate of rice husk (RH) can be highly enhanced while it was grinded into powders [6]. The biodegradability tests of the RH/PVA mixture (RHP) including the growth of microbes and the amount of CO_2 released show that all of microbes tested can use the RHP as sole carbon resource and energy resource, and they can release CO_2 rapidly during the growth process. Meanwhile, the colonies appeared after 2–3 days of incubation [7]. Accordingly, the mixture of the renewable RH powders and the biodegradable PVA can be used as an excellent environmental material for making disposable composites or products.

The forming properties of powder-based composite sheet with relatively high volume fraction of powders are quite poor at room temperature because of their poor plasticity and flowability. As blending the mixture of powders and binder, some additives or much amount of cemented medium are needed for increasing flowability and plasticity. However, as forming this mixture into a part with high depth and/or thin wall, the actuating pressure must be greatly high for either the compression molding or the injection molding. And the products with thin thickness of wall will have insufficient strength. In essence, a very complicated device providing the compression molding with multiaxial actuating forces are considered necessary for obtaining high and uniform density of products [8].

Experimental result showed that the RH/PVA powders (RPP) with PVA content of 20–25% in weight could be directly formed into various shapes of containers by means of compression molding technique while the specimen were heated up to above 80°C [9]. However, thicker wall of specimen and greatly high working pressure are needed due to poor flowability of the RPP. For making containers with thin-wall, one of the best ways is to calender RPP into thin sheet and to be fabricated into various shapes of products. However, good quality of disposable containers made from RH/PVA composite (RPC) sheets should have fast biodegradation rate, high degree of biodegradation, and low material cost. Therefore, the fabricated RPC sheet with very low content of PVA or quite high volume fraction of RH powder is essentially needed. It was found that the minimal mixing ratio of PVA/RH, from which the mixture can be rolled into qualified sheet, is 1/10 in weight for RH powders with randomly distributed around 250–400 μm.

An alternative forming process is to deform the powder-based composite sheets into various shapes of green parts or products at room temperature. During the mold forming of the sheet, sophisticated forming behaviors involving bending, shrinking, buckling, crack initiating, extruding, compressing, and finishing shaping were observed, which is thoroughly different from the forming of metals [9]. Therefore, it is intrinsically needed that the composite sheets have good tensile strength, ductility, bendability, and flowability for the cold forming. Usually, the powder-based composite sheets, such as biodegradable rice-husk/polyvinyl-alcohol composite (RPC), can be fabricated by means of conventional rolling process. However, it was found that as enhancing the tensile strength of the RPC sheet to a great extent by rolling, its failure strain or ductility become poor. Meanwhile, the bendability and the flowability of the RPC sheet meaningfully related to forming properties were also reduced [9–11]. Theoretically, the relationship among engineering strain e, bend radius R and thickness T for a sheet subjected to a bending moment is expressed as [2]

$$e = 1/[(2R/T) + 1] \tag{1}$$

This relationship indicates that the engineering strain e decreases with the increased bend radius R or the decreased thickness T. Obviously, the forming depth of a sheet will reduce with the increasing of the bend radius R during the deep drawing. Consequently, multiple stages of forming process are necessary for producing high depth or complex shape of products. Resourcefully, as for an enhanced sandwich structure of RPC sheet with finer densified skins, its effective thickness T is intrinsically reduced due to thinner thickness of skin. Therefore, the induced engineering strain e in the skin becomes smaller, and/or the allowable bend radius of the RPC sheet is reduced. Essentially, as forming and shaping the RPC into a deep container, it was found that the smaller the minimum bend radius of the specimen is, the deeper the container can be produced [9–11].

It has been found that the drawing depth of an axial symmetric container with cone angle of 40° and wall thickness of 1 mm made from RPC sheet was confined to below 27 mm [9]. While further drawing the container to that with depth of 48 mm, another three stages of forming process were needed. The main reason leads to the poor formability of the composite sheets could be attributed to their stringer porous-structure, insufficient bonding strength, and poor surface texture. This is because there is an inverse relationship between bendability and the tensile reduction of area of the material [2]. The minimum bend radius is, approximately,

$$R_{\min} = T[50/(r - 1)] \tag{2}$$

where r is the tensile reduction of area of the sheet metal. Therefore, bendability depends on the form of inclusion also, which is related to the tensile reduction of area. Because of their pointed shape, inclusions in the form of stringers are more detrimental than globular-shaped inclusion. It was found that globular-shaped pores embedded in the composite are more advantageous to enhance the bendability than the stringer pores [12]. According to Equations 1 and 2, the enhanced structure of RPC sheet structurized by highly densified thinner skins and reasonable dense/loss core-material can decrease effective thickness T and increase tensile reduction of area r to enhance bendability.

It is found in this work that the rolling schemes significantly influence the fabricated structures of the RPC sheets, and the configurations of the structures are meaningfully related to the forming properties. Essentially, the stripy porous structure and the poor skin texture of the RPC sheets were observed. Therefore, an enhanced structure of composite sheet providing for both better mechanical strength and forming properties is worth pursuing.

A new biodegradable rice-husk/polyvinyl-alcohol mixture with relatively high volume fraction of powders is used as the raw material for the proposed processing and fabricated into desired sandwich structures of thin sheets. The prime objective in this work is to give the characterization of the enhanced sandwich structures of composite sheets with fine densified skins and partially densified core-material in the mechanical and forming properties. Herein, the outer skins have to be highly densified and the densified core-material must be cross-linked to the outer skins. Consequently, the tensile strength, the failure strain, the bendability, and the flowability of RPC sheet are examined and compared to the conventional stringer porous structure.

1.1. Experimental procedure

The mixture of rice-husk (RH) powders and polyvinyl-alcohol (PVA) solution with the weight ratio of 1:4:10 for PVA:water:rice-husk powder is blended into RH/PVA granules and then rolled into sheets. Polyvinyl alcohol pellet has the following properties: solution viscosity = 24–33 cps, water-solubility = 86–89%, degree of polymerization = 2000–2100, molecular weight = 99000–104000, glass-transition temperature $T_g = 60$–80°C, pH value = 5–7. The diametral sizes of rice-husk powders are randomly distributed around 250–400 μm. The RPC sheet with the mixture ratio of RH:water: PVA = 2:7:10 (RH:PVA = 1:5) can be easily form into a deep container through the use of straight vacuum forming at room temperature. However, for increasing the biodegradation rate of product and reduce the cost of raw material RPC, the water-soluble PVA used as the bonding medium of rice-husk powders is confined to a minimum value. The RH/PVA composites with compositions other than 1:4:10 to arrive the minimum value have been developed [9, 13]. This work showed that the minimum value of PVA content depends upon the average diameter size of RH powders because the surface area per unit volume of the RH powders increases with the decreasing of the powder size. For example, the minimum value of PVA content of RPC sheet fabricated from RH powder with diameter distribution ranged from 125 to 150 μm is confined to PVA: water: RH powder = 1:4:8 [9].

Mechanical properties of PRC sheets fabricated into that with different porous structure were examined by using simple tensile test. The composite panel was cut as the specimen with dimension of 80 × 20 mm (length × width) according to CNS standard (1354, P3004) for paper tensile test. CNS is the Chinese National Standard related to the method of test for tensile breaking strength and elongation of paper and paperboard. The aluminum/RPC endtabs were attached to two ends of specimens to protest specimens and transfer the load from INSTRON testing machine to specimens. The testing of specimen was operated at constant speed of 0.6 mm/min.

A multiple stepped bar with various diameters (6–36 mm) was conducted to the bending tests of the RPC sheets with different rolling conditions to find the allowable minimum bend radius which is closely related to the formability of the composite sheet. The RPC panel was cut as the specimen with dimension of 80 × 15 mm (length × width). The minimum bend-radius and the allowable minimum bend-radius of RPC sheet are defined as the bend radius that can be formed on a strip without the generation of noticeable surface cracks and through-thickness cracks, respectively.

The surface textures of the RPC sheets fabricated by the 3D rolling technique with different jumping angles were examined by scanning electron microscopy (ABT-55 Tungsten SEM, TOPCON, Japan). The cross-sectional structures of the RPC sheets fabricated from the conventional rolling scheme and the 3D rolling technique were examined by hot stage polarizing microscopy (Laborlux 12 pol S, Nikon Cool PIX 990, HSPM, Japan).

1.2. Fabricating scheme

The conventional flat rolling machine associated with the use of the novel dynamic 3D roll mill was applied to calender the RPP into RPC sheets under different rolling conditions and processing schemes. Meanwhile, it is a cold rolling process with T_g of PVA being over ambient temperature. The flat rolling machine has the specification:roll = 216 × 303 (mm); power = 3.73 kW; operating speed = 0–120 rpm. The dynamic 3D roll mill is designed with the same specification as the flat rolling machine in the power supply and the size of roll. However, the surface of the cylindrical roll is machined into ring slots with desired depth, width, and pitch for the purpose of extruding material to flow in the axial direction of the rolls during the processing, shown in Fig. 1a. The rolls of the 3D rolling device is designed so versatile such that they can rotate (rotating motion) and jump (translating motion) with any desired angle of θ and amplitude S as well as associated with variable pulsating roll gap to fabricate different configurations of sandwich structure of composites, shown in Fig. 1b. Herein, the variable pulsating roll gap can be used to fabricate a dense/loose structure of the RPC sheet along the rolling direction, shown in Fig. 2b. The relationship between the maximum reciprocating displacements induced by the jumping rolls in the rolling direction (d_x) and the thickness direction (d_z) and the jumping angle θ can be express as $\tan\theta = d_x/d_z$. Obviously, the intensification of the skin and the core-material of the RPC sheet can be controlled while tuning the jumping angle θ. That is, the greater the d_x, the higher the skin density of the RPC sheet. By contrast, the lower the d_x or the larger the d_z, the better the densification of the core-material of the RPC sheet.

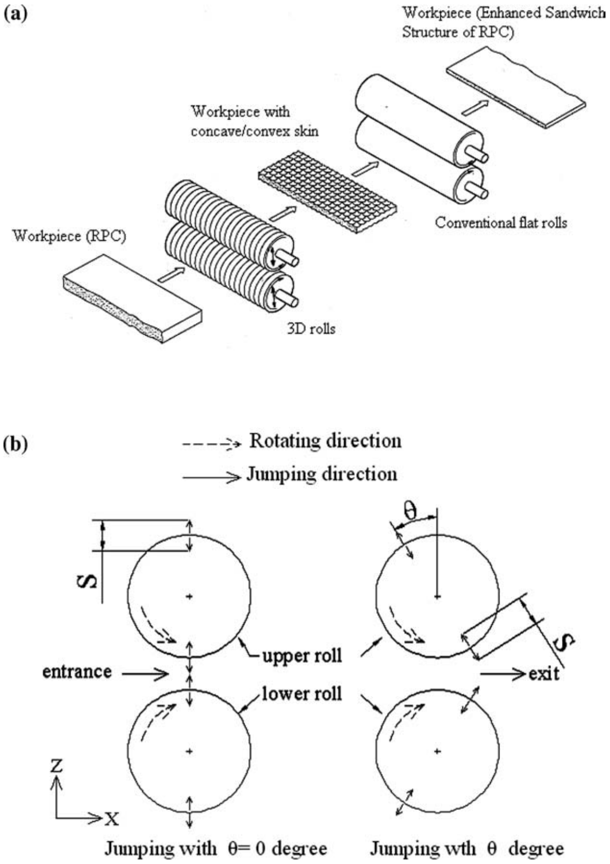

Figure 1 The schematic illustration of rolling an enhanced sandwich structure of RPC sheet: (a) by using the conventional rolling device associated with the novel three-dimensional roll mill installed and (b) with vibration rolls to generate synthesis motion of rotation and jumping translation.

2. Results and discussion

The dynamic jumping rollers of the roll mill shown in Fig. 1, which can be tuned to jump at any desired angle, provide alternating compression and extrusion forces to densify the workpiece along the in-plane and out-off-plane direction. During the rolling processing, the ring slots of the jumping rollers and the induced alternating rolling force in the rolling direction extrude the materials near the surface to flow along the axial direction of roller and rolling direction, and thus the density and the surface texture of the outer layers of the RPC sheet can be enhanced. Meanwhile, the alternating rolling force in the thickness direction provides the RPC sheet with locally and periodically intensified density of core-material. Therefore, the distribution of the intensified

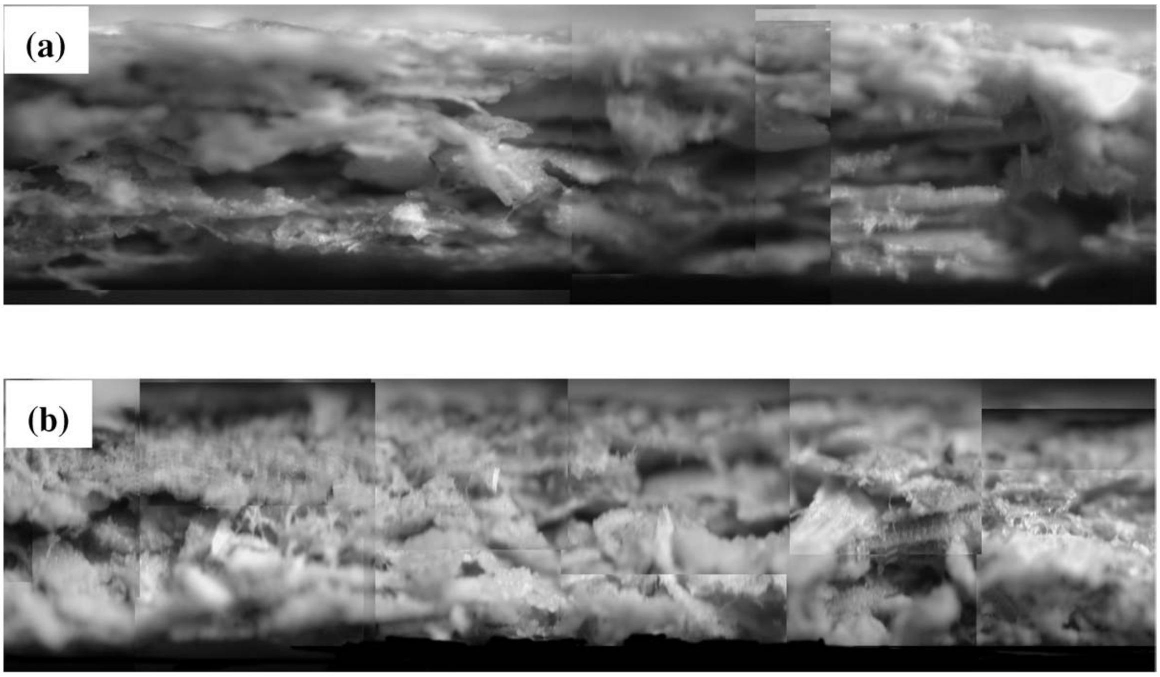

Figure 2 The optical micrographs of the cross-sectional structures of the RPC sheets (40×) fabricated: (a) with the conventional rolling technique and (b) the novel 3D rolling methodology.

density of the composites is controllable by means of the jumping modes of the rollers. Consequently, different types of enhanced sandwich structures of RPC sheets can be produced. Fig. 2b shows the optical micrographs of the enhanced sandwich structure of RPC sheet composed of highly densified skin and locally and periodically densified core-material made by 3D rolling technique. In the meantime, the RPC sheet with stringer porous-structure made from the conventional rolling (Fig. 2a) has been fabricated as that with near-globular porous structure (Fig. 2b). That is, the stringer voids contained in the RPC sheet have been deformed as oval or round voids embedded in the interlayer of the RPC sheet. It is also found that the highly densified skin and the locally densified core-material produced are closely cross-linked together, and the loose core materials are interlaced among the locally densified core-material. As a result, the enhanced sandwich structure of RPC sheet is essentially constructed as network structure formed from the intense densified material. Furthermore, as shown in Fig. 3, the surface finish of the RPC sheet made form the 3D rolling technique is much finer than that fabricated from conventional rolling method. In addition, the average net cross-sectional area of the RPC sheet is also increased, shown in Fig. 2b.

Theoretically, the higher the density or the lower the void content of the porous material is, the greater is its tensile strength. As stretching the RPC sheet with enhanced sandwich structure, the network structure will deflect along the direction of applied load, and thus the outer layers of the RPC sheet deform toward the core-material. Meanwhile, the porous core-material with appropriate void filling capacity provides proper space for the material flow as the final result that the flexibility of the enhanced sandwich structure can be greatly enhanced. That is the enhanced sandwich structure with network substructure can intrinsically provide for the need of improving both the stiffness and the flexibility, which are significantly related the mechanical properties and the flowability of the RPC sheet. In addition, the near globular porous structure and the fine surface finish are capable of offering the improvement of the bendability of the porous composites [12]. Consequently, the tensile strength, the failure strain, the bendability, and the flowability of the porous composites can be significantly enhanced.

Fig. 4 illustrates the enhanced mechanical properties of RPC sheets fabricated with different rolling schemes. It is found that the enhanced sandwich structure gives significantly effect on the mechanical properties. The magnitudes of the tensile strengths and the failure strains of the RPC sheets increase noticeably with the increasing of jumping angle from 0° to 30° and 0° to 60°, respectively. Three reasons lead to the increases of the tensile strength and the failure strain. First, proper deformations produced by extrusion in different directions are needed for enhancing the flow of material to intensify the density of RPC sheet controllably. Second, the bonding strength of RH/PVA powders related to mechanical properties can be significantly increased through alternating compression and extrusion. Third, the network structure is constructed from thin thickness of highly densified material with excellent bonding strength can provide for the need of the flexibility as well as the strength to the system, and the flexibility offers the enhancement of the ductility or the failure strain of the structurized porous composite sheet.

Accordingly, the densified outer layers or skins play a key role for enhancing the tensile strength of the powder-based porous composite sheet. The network structure fabricated from the interconnected periodically densified core materials and the densified outer layers provide not only for the improvement of tensile

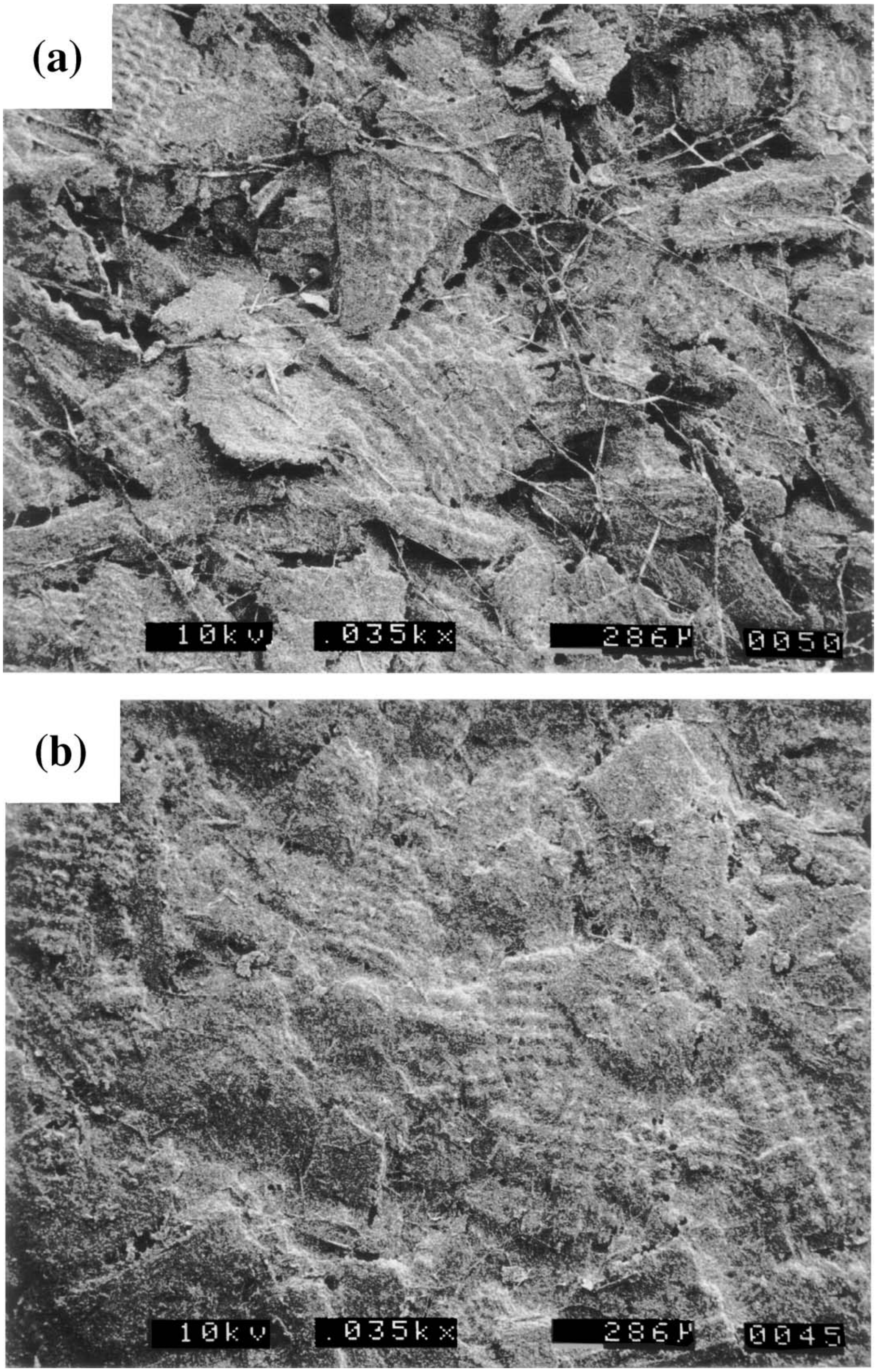

Figure 3 Scanning electron micrographs of RPC sheets fabricated: (a) with the conventional rolling method and (b) the novel 3D rolling technique associated with jumping angles of 0° and 45°, respectively.

strength but also for the ductility of the porous composite sheet. The porous core-material offers the flowability of the powders and the elastic deformation of network structure during the forming. As shown in Fig. 4a and b, the tensile strengths in the rolling direction (X-direction) and the axial direction (Y-direction) for the wet RPC sheets with 25% water content have been increased by 104 and 81%, respectively, and their failure strains are also increased by 56 and 48%. Correspondingly, the tensile strengths of the dry RPC sheet in the rolling direction and the axial direction are enhanced by 130 and 138%, respectively, shown in Fig. 4c, and the failure strains are increased by 53 and 43%, respectively, shown in Fig. 4d. In addition, the surface

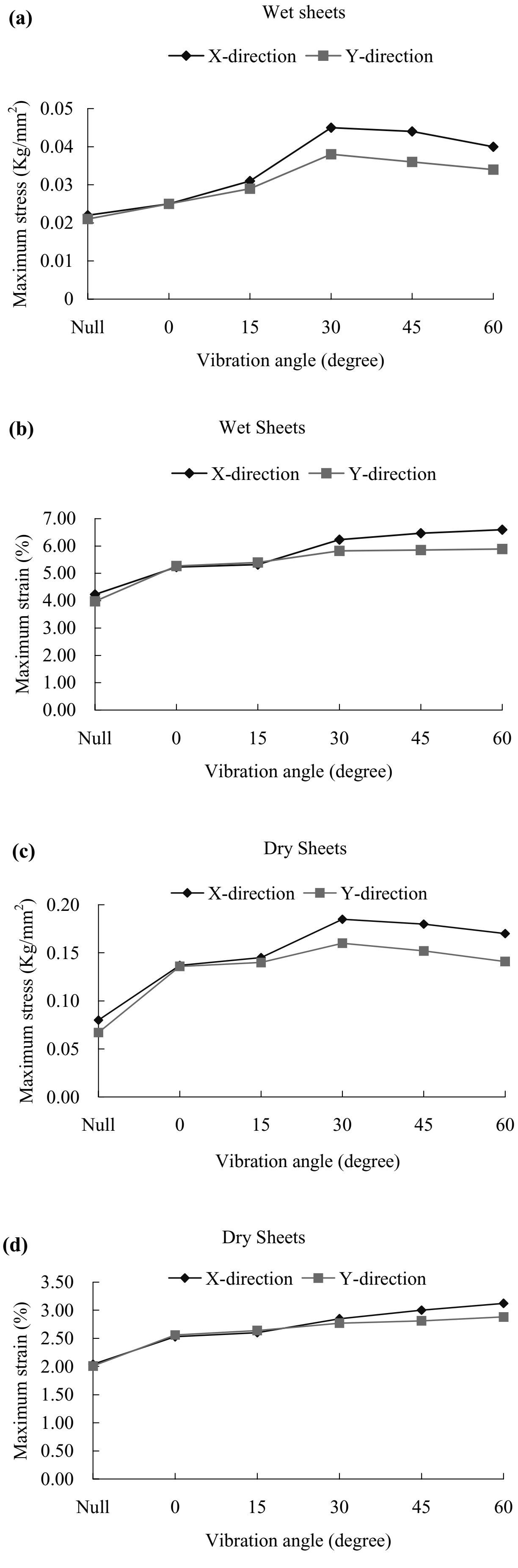

Figure 4 The variations of the maximum stresses and failure strains with the vibration angles of jumping rolls for (a, b) the wet RPC sheet and (c, d) the dry RPC sheet in the X-direction (rolling direction) and Y-direction (axial direction of rolls).

finish of the RPC sheet is enhanced with the increase of jumping angle shown in Fig. 3b. Essentially, as rolling the RPC sheet with different rolling phases, the higher the jumping angle of rollers, the smaller the thickness of densified skin and the greater the width of intense core-material are, and thus the higher the intensified density of skins and the lower the intensified density of core-material are generated. Obviously, the thickness of densified skin and the width of densified core-material of the network structure dominate the stiffness/strength and the flexibility/ductility of the RPC sheet. For obtaining higher failure strain of the structurized RPC sheet, thinner thickness of densified skin is intrinsically needed. By contrast, too thinner thickness of densified skin is not permitted for obtaining high tensile strength of RPC sheet.

There is an inverse relationship between bendability and the tensile reduction of area of the material. Therefore, to increase the bendability of RPC sheets, we may increase their tensile reduction of area by means of the highly densified outer-layer and the globular porous core-material of the enhanced sandwich structure. Experimental results show that the allowable minimum bend radius of RPC sheet will decrease to a certain extent with the increasing of skin densification. The best allowable minimum bend radius of RPC sheet can be obtained while setting the jumping angle of rollers at the range of 30–60° during the rolling. Herein, as bending these RPC specimens on a 3-mm diameter bar, it is found that they cannot be fractured completely and the inside compressive layer of the RPC sheet is still attained to good status although the outside tensile layer has been cracked near the interlayer. Obviously, as setting jumping angle to be higher than 30° during the rolling, a good densified skin of RPC sheet is produced, and thus its compressive outer-layer cannot be fractured during the bending test.

Bendability depends not only on the mechanical properties of the sheet but also on the edge (or surface) condition and the shape of inclusion (or porous) configuration. The finer the surface finishing is, the better is the bendability of a porous composite sheet. The porous composite sheet with globular porous structure has better bendability than that with stringer porous structure [12]. In addition, the void content of the RPC sheet offers the need of the space for the flow of material during the forming. Experimental result showed that the crinkles of the RPC sheet induced from the shrinkage around the circumference of the workpiece must be compressed out and new bonds are formed between the RH/PVA powders in these areas during deep drawing [9]. The entire wall of the workpiece is preferably densified and slightly increased in thickness comparing with the bottom of the sheet. In essence, globular porous structure gives more favorable flowability than the stringer-shaped porous structure as forming and shaping the specimen. Consequently, the RPC sheet with enhanced sandwich structure proposed improves both the mechanical and the forming properties.

The products made from RPC sheets, including (a) shallow depth of dish, (b) medium depth of container, and (c) high depth of container, are shown in Fig. 5.

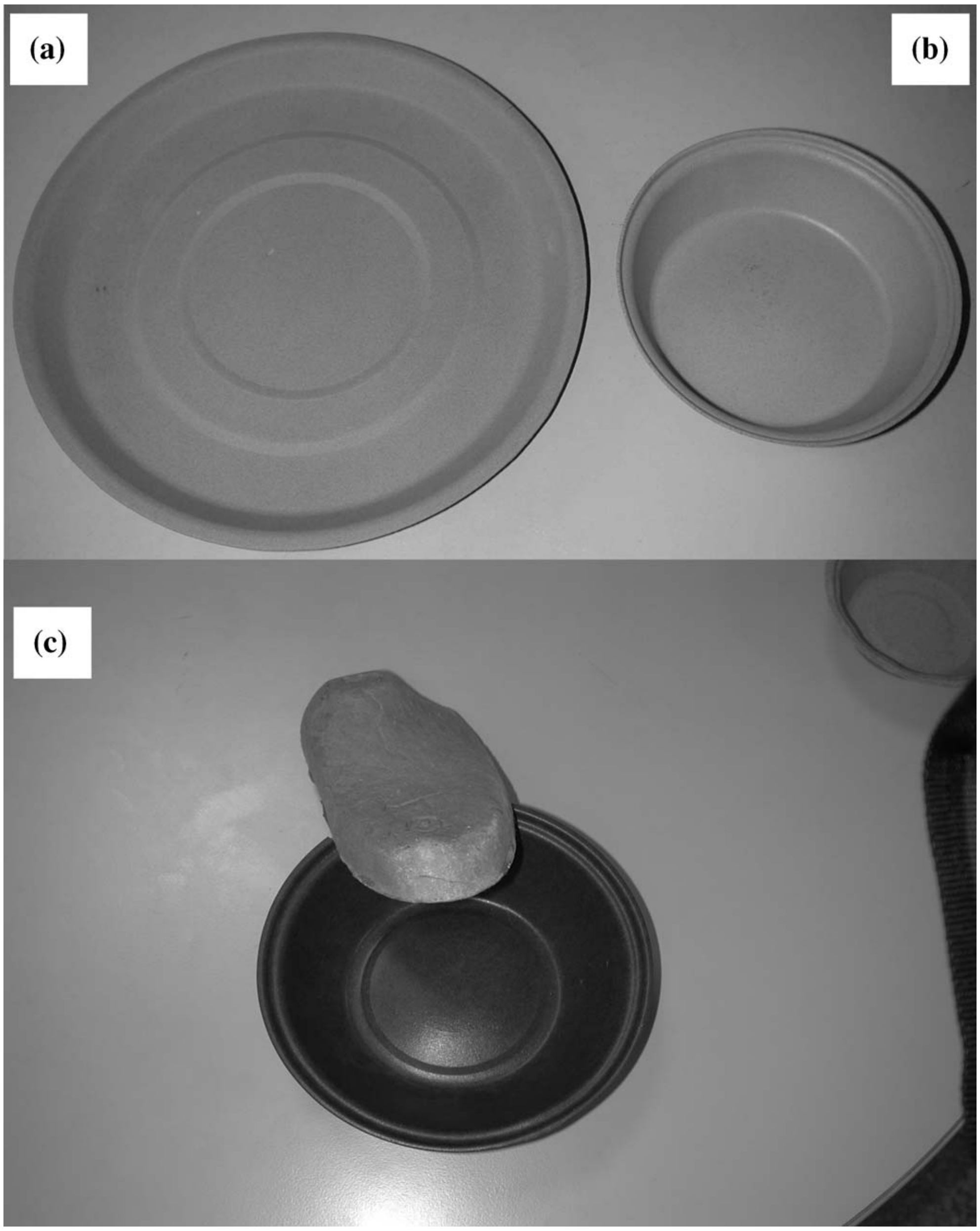

Figure 5 The products of RPC sheets: (a) shallow depth of dish, (b) medium depth of container, and (c) high depth of container.

Essentially, the RPC is typically a highly porous material, and the flaws on the near surface of wetted RPC sheet can be closed by compression and recovered by virtue of the rebinding capability of the plasticized PVA at room temperature.

3. Conclusions

The mechanical properties and formability of rice-husk/polyvinyl-alcohol composites (RPC) with enhanced sandwich structure have been successfully characterized. The highly densified skins closely cross-linked with the intense densified core-material to be formed as a network structure can provide for the reinforcements of the strength, the flexibility or ductility, and the bendability of the porous composites (RPC). The network structure of the porous composites exhibits a phenomenon akin to increased ductility and/or enhanced tensile strength. Different thicknesses of the densified skins and widths of the intensified core-material give dissimilar effects on the tensile strength, the failure strain as well as the bendability of the porous composites. The fine surface texture of the skins and the locally densified porous core-material with globular voids can intrinsically offer the enhancements of the resistance of surface crack initiation, the flowability, and the bendability of the composite sheet.

The desired configuration of enhanced sandwich structure can be fabricated by controlling the modes of deformation by applying alternating multiple forces associated with the use of the proper dimension of the slot of rollers. In essence, different types of enhanced sandwich structures for RPC sheet can be produced through the use of a novel versatile 3D rolling technique.

It has been acknowledged that the enhanced sandwich structure can provide for significant contributions to the enhancements of the mechanical properties and the forming properties of the wet RPC sheets needed for mold forming at room temperature. Correspondingly, the tensile strength and the failure strain of the dry RPC sheet are also greatly improved. Consequently, the proposed enhanced sandwich structure can be easily extended to the fabrications of other composites with relatively high volume fraction of powders or short fibers to boost their forming properties and fabricating efficiency.

Acknowledgements

Author is grateful for financial support from National Science Council in Taiwan on this work under the contracts NSC 90-2212-E-035-007 and NSC 91-2212-E-035-009.

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Received 26 December 2002
and accepted 17 October 2003