The properties of various lignocellulose flour-plastic composites with high filler contents

Masahiro TAKATANI*

** Department of Advanced Bioscience, Kindai University, 3327-204, Nakamachi, Nara 631-8505*

Synopsis

Lignocellulose flour-plastic composites with 80% filler contents were experimentally made from lignocellulosic materials such as rice husks, apricot seeds, and wood flour of agriculture and forestry product origin. Compounds containing lignocellulose flour, thermoplastic polymer (PP), and compatibilizer were made with a Henschel type mixer, and then the composites were manufactured by compression molding and twin-screw extrusion molding under the following conditions: temperature of 200°C, pressure of 11 MPa, time of 10 min, and temperature of 180°C, pressure of 6.5 MPa ~17 MPa, respectively.　The fracture strength and water resistance of composites were evaluated.　The mechanical properties of boards made by compression molding were maximal for Asada (Japanese hardwood flour)　: PP : MAPP = 80 : 20 : 2, showing bending strength; 40 MPa , and thickness swelling (TS) after 24hr immersion in water; 6%, and for boards made by extrusion molding were maximal for commercial wood flour ：PP ：MAPP = 80 ：20 ：2, namely bending strength; 73 MPa and TS; 1.2%. The water resistance of rice husk flour-plastic composites showed particularly good performance.

Keywords: lignocellulose, filler, compression molding, twin-screw extrusion molding

1. Introduction

The term wood flour-plastic composite (WPC) is a very general term for many different blends and formulations in USA and Europe[1]. Wood flour has been successfully blended with PVC, HDPE, PP, PS, and ABS[2, 3]. Studies on lignocellulosic fiber-plastic composites have been concentrated on wood–based flour and grass fiber, and have been reported by many researchers. Furthermore, many natural fiber materials other than wood flour and/or wood fiber have been blended with these thermoplastics. Plant fibers are currently being evaluated as environmentally friendly and low-cost alternatives for E-glass fibers. Natural fibers have a number of advantages over E-glass fibers; they are renewable, cheap, lightweight, biodegradable, non-abrasive, and CO_2 neutral. Also natural fibers can be incinerated with energy recovery, there is less concern with safety and health, and they have good mechanical properties[4].

In this study, we tried to manufacture environmentally-friendly lignocellulose flour-plastic composites for the purpose of obtaining better properties despite the high filler contents of 80% using lignocellulosic materials such as rice husk, apricot seed, and wood flour of agriculture and forestry product origin. We then evaluated the performance of these various lignocellulose flour-plastic composite boards made by two processing methods; compression molding and twin screw extrusion molding.

2. Materials and methods

2.1 Materials

The materials used in this study were three kinds of wood flour, rice husks, and apricot seeds as fillesr of lignocellulosic flour, polypropylene (PP) as a thermoplastic polymer, and MAPP which is polypropylene modified with maleic anhydride as a compatilizer. Wood flours were a commercial product from soft wood (C300G, Rettenmaier Co.), and two waste wood flours, asada and lauan. Waste agriculture products used as fillers were rice husks supplied by JA Ishikawa and apricot seeds supplied by Daiei Kigyo Co. The size distribution of lignocellulosic flours was lauan > rice husk> asada > C300G > apricot as shown in **Table 1**. The thermoplastic polymer was a random PP (PM930V, density 0.9 g/cm^3, melting point 150-153°C, MFR 55g/10min at 200°C, Sun Allomer Co.). The compatibilizer was MAPP (H1100P) obtained from Toyo Kasei Kogyo Co.

2.2 Blending procedure

All materials of lignocellulosic flour, PP, and MAPP could be successfully blended using a Henshel type mixer (SMV-20A type, Kawata Co.). The mixer was operated as the temperature increased from ambient temperature to 180℃ and a blade speed of 1380 rpm to prepare homogenized compounds. The composite materials were compounded with two ratios of lignocellulosic flour : PP : MAPP, 80 : 20 : 0 and 80 ： 20 ： 2, by weight.

2.3 Molding process

Compression molding

The compounds were used after drying at 60℃. The composite boards were made using the compounds of two ratios of lignocellulosic flour : PP : MAPP, 80 : 20 : 0 and 80 : 20 : 2 by compression molding with an oil hydraulic hot press (Model F type, Sinto Kinzoku Kogyosyo Co.). Namely, the compounds of 24 g were compression-molded using steel mold (10 x 80 x 100 mm) by the hot press at 200℃ and 11 MPa for 10 min, before being cooled to room temperature. Teflon films were used to avoid adhesion of compounds to the stainless surface of the mold. The board was cut into three pieces of 25 x 100 mm, which were used as test pieces. Six standard specimens (dimensions, 2.5 x 25 x 100 mm, density, 1.2 g/cm^3) were prepared.

Extrusion molding

The compounds were put into a conical twin screw extruder (Taitan 68, Taiyo Gosei Co.) with an outlet of 60 x 7 mm. The composite materials were prepared with two ratios of C300G : PP : MAPP, 80 : 20 : 2 and C3000G : Rice husk : PP : MAPP, 40 : 40 : 20 : 2. Process conditions for extrusion are shown in **Table 2**. The composite boards were successively obtained with the outlet of 60 x 7 mm. The long composite board obtained was cut into test pieces of 7 x 25 x 140 mm. The board density of a sample was about 1.2 g/cm^3. Six specimens were used to evaluate the performance of composite boards.

2.4 Mechanical test and water absorption test

The performance of composite boards was evaluated by bending test and water absorption test. The bending strength and water resistance of test pieces were determined according to Japanese Industrial Standard method, JIS A 5741 for wood plastic recycled composite[5)] and JIS A 5908 for particleboards[6)]. The bending strength was measured by a three point bending strength test machine (Model TCM-500, Shinko Tushin Co.). The water resistance was determined by measuring the swelling and water absorption of test pieces after immersing the sample into water at room temperature (22±2℃).

3. Results and discussion

3.1 Properties of composite boards by compression molding

Good compounds were obtained by blending with lignocellulosic flour, granular PP, and MAPP flour in the Henschel type mixer. Results of bending tests and water absorption tests on composites boards made with different kinds of lignocellulosic flour by compression molding are shown in **Fig.1～4**. The error bars represent one standard deviation. The modulus of rupture (MOR), modulus of elasticity (MOE) and dimensional stability of all specimens of composite boards were improved by addition of MAPP. The composite boards made with the ratio of asada : PP : MAPP＝80 : 20 : 2 showed the maximal MOR of 40 MPa, and better water resistance, thickness swelling (TS) after 24 hr

Table 1 Size distribution of lignocellulosic flour (%)

	10~20mesh	20~60mesh	60~100mesh	100mesh~
C300G	9.8	57.5	17.7	14.5
Asada	7.9	66.9	15.3	5.3
Rice husk	34.5	52.7	4.5	4.2
Apricot	ND	22.7	42.7	32.1
	~7mesh	7~10mesh	10~20mesh	20mesh~
Lauan	32.8	37.5	24.2	5.4

Table 2 Process conditions for extrusion in the twin-screw extruder

	Temperature (℃)								
Filler type	Screw	Cylinder 1	Cylinder 2	Cylinder 3	Cylinder 4	Adapter	Die 1	Die 2	Die 3
C300G	125	205	195	172	170	170	172	135	60
C300G/Rice husk	130	225	220	175	172	172	178	140	56

Filler type	Screw speed (rpm)	Feeder speed (rpm)	Resin temp. (℃)	Resin pressure (MPa)
C300G	1.3~3.0	0.5~1.5	174	14.2~17.0
C300G/Rice husk	2.0	1.5	177	6.5~16.6

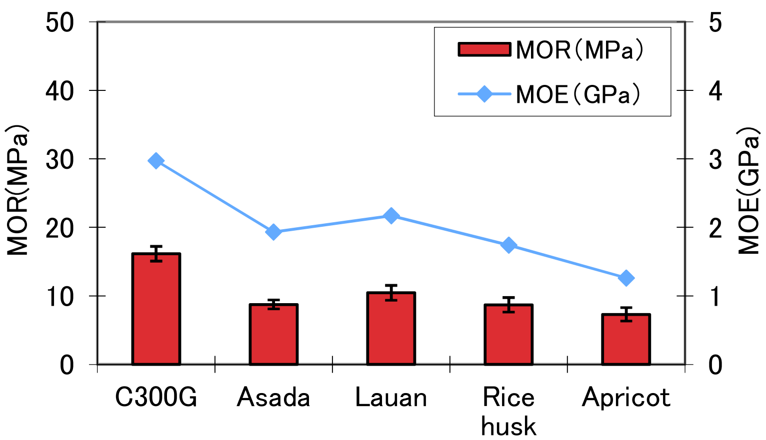

Fig. 1 Bending strength of wood flour-PP composite boards using different kinds of lignocellulosic flour (Lignocellulosic flour : PP =80 : 20).

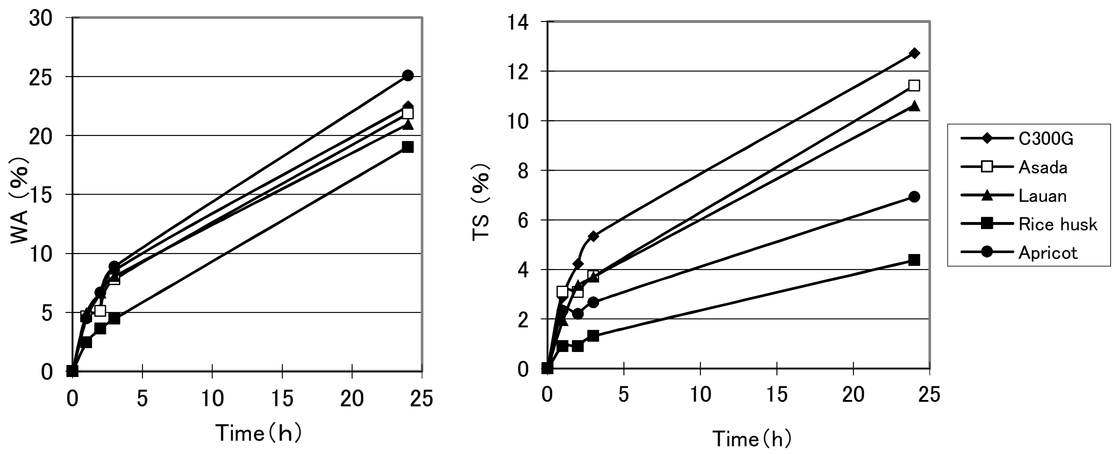

Fig.2 Water resistance of wood flour-PP composite boards using different kinds of lignocellulosic flour (Lignocellulosic flour : PP = 80 : 20).

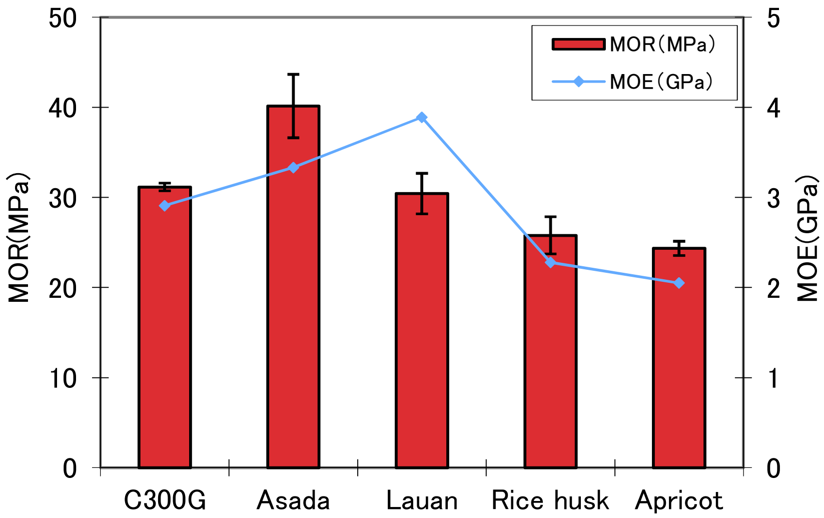

Fig.3 Bending strength of wood flour-PP composite boards using different kinds of lignocellulosic flour (Lignocellulosic flour : PP : MAPP = 80 : 20 : 2).

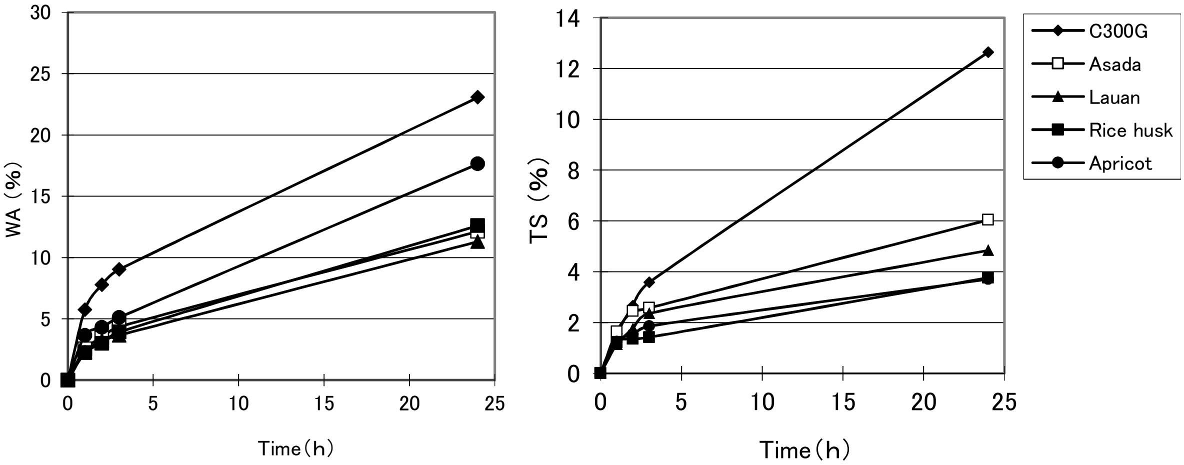

Fig.4 Water resistance of wood flour-PP composite boards using different kinds of lignocellulosic flour (Lignocellulosic flour : PP : MAPP = 80 : 20 : 2).

immersion in water; 6%. But none of the specimens passed the level (less than 10%) of water absorption for JIS A5741 (**Fig. 4**).

3.2 Properties of composite boards by extrusion molding

The results of bonding test and water absorption test of composite boards prepared with differential levels by extrusion molding are shown in **Fig. 5 and 6**. The performance of composite boards made with extrusion molding was improved by addition of MAPP even more than that of composite boards made with compression molding, and significantly passed the standard levels of bending strength (more than 20 MPa) and water resistance (less than 3% water absorption, 12% thickness swelling, and 3% linear expansion for JIS standard).

The boards made by extrusion molding were superior with the following ratio; commercial wood flour：PP：MAPP = 80：20：2, namely MOR; 73MPa, WA; 2.0%, and TS; 1.2 %. The MOR of commercial wood flour-plastic composites was similar with its of bamboo flour-plastic composites[7]. Also, the water resistance of rice husk flour-plastic composites showed good performance, WA; 0.8% and TS; 0.7%. These superior properties of rice husk flour-plastic composites might be the result of hydrophobic silicate in rice husk[8].

4. Conclusions

The lignocellulose flour-plastic composites with 80 % filler contents were experimentally made from lignocellulosic materials such as rice husks, apricot seeds, and wood flour of agriculture and forestry product origin. The compounds containing lignocellulose flour, thermoplastic polymer (Polypropylene, PP), and compatibillizer were made with a Henschel type

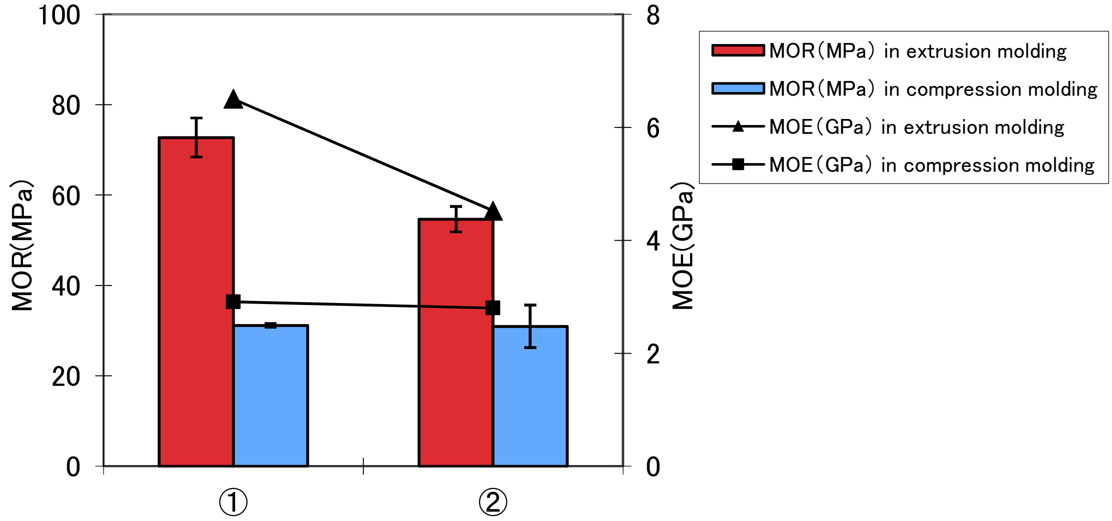

Fig.5　Bending strengths of wood flour-PP composite boards

① C300G : PP : MAPP = 80 : 20 : 2

② C300G : Rice husk :PP : MAPP = 40 : 40 : 20 : 2

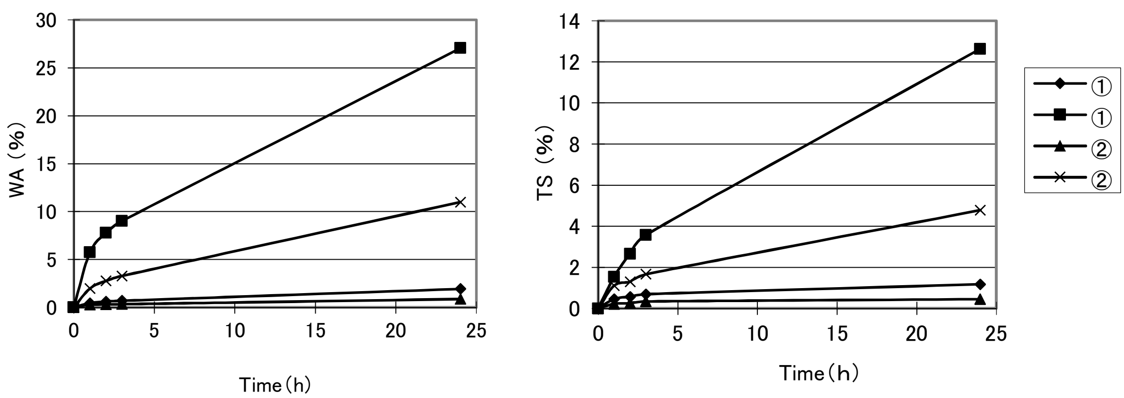

Fig.6　Water resistance of wood flour-PP composite boards

① C300G : PP : MAPP = 80 : 20 : 2

◆ extrusion molding, ■ compression molding

② C300G : Rice husk :PP : MAPP = 40 : 40 : 20 : 2

▲ extrusion molding, × compression molding

mixer (Kawata Co.), and then the composites were manufactured by compression molding and twin-screw extrusion molding under the following conditions: temperature of 200°C, pressure of 11 MPa, time of 10min, and temperature of 180°C, pressure of 6.5 ~ 17 MPa. The fracture strength and water resistance of composites were evaluated. The mechanical properties of boards made by compression molding were maximal for the following ; Asada (Japanese hardwood flour) : PP : MAPP = 80 : 20 : 2, namely bending strength; 40 MPa , and thickness swelling (TS) after 24 hr immersion in water; 6 %. The performance of boards made by extrusion molding was maximal for the following ratio; commercial wood flour : PP : MAPP = 80 : 20 : 2, namely bending strength; 73 MPa and TS; 1.2%. Notably, the water resistance of rice husk flour-plastic composites showed particularly good performance. And larger area composites (a cross section, 50 mm x 500 mm) by extrusion molding were made. The composite boards were made with the ratio of wood flour : PP : MAPP = 80 : 20 : 3 and its density was 1.2 g/cm^3. The speed of molding was about 700 cm/hr. Mirmehdi et al. reported the effect of filler content and filler type on lignocellulose-polyethylene composite properties[9]. The findings described in that report support the results obtained in this study.

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高フィラー含有量のリグノセルロース/プラスチック複合体の特性

高谷　政広*

*　近畿大学農学部バイオサイエンス学科

要約

フィラー含有量 80%のリグノセルロース/プラスチック複合体が農林産物のモミガラ、アンズの種、および木粉から実験的に試作された。リグノセルロース粉末、熱可塑性樹脂（PP）、および相溶化剤を含んだコンパウンドをヘンシェル型ミキサーで作製後、複合体は熱圧縮成形法と二軸押出し成形法によりそれぞれ次のような条件、200℃、11 MPa、10 分間と 180℃、6.5 MPa～17 MPa で試作され、複合体の強度と耐水性が評価された。熱圧縮法で作製された複合体では、アサダ（広葉樹木粉）: PP : MAPP＝80 : 20 : 2 の比率が優れ、曲げ強度 40 MPa、水中浸漬 24 時間後の厚さ膨張率（TS）6%であった。また押出し成形法で作製された複合体では、市販木粉（C300G）：PP：MAPP＝80：20：2 の比率が優れ、曲げ強度 72 MPa、水中浸漬 24 時間後の厚さ膨張率 (TS) 1.2%であった。特に良好な耐水性を示した複合体は、モミガラの場合であった。