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# Effect of Addition of PP-g-MA on the Tribological Properties of Hemp Fiber Reinforced Plant-Derived Polyamide1010 Biomass Composites

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**Abstract.** The aim of this study is to investigate the effect of addition of maleic anhydride grafted polypropylene (PP-g-MA) on the tribological properties of hemp fiber (HF) reinforced plant-derived polyamide 1010 (PA1010) biomass composites to develop the new engineering materials such as mechanical sliding parts (tribomaterials) based on inedible plant-derived materials. PA1010 was made from sebacic acid and decamethylenediamine, which are obtained from plant-derived castor oil. HF, which is one of the natural fibers, was used as a reinforcement fiber. HF was surface-treated by two types of surface treatment: (a) alkali treatment by sodium chloride (NaClO<sub>2</sub>) solution and (b) surface treatment by ureidosilane coupling agents (A-1160). PP-g-MA (2wt.%) was used as a compatibilizer. These biomass composites were extruded using a twin extruder, and injection-molded. Tribological properties such as frictional coefficient and specific wear rate by ring-on-plate type sliding wear testing were evaluated. The wear debris were observed by scanning electron microscope for understanding the wear mode. It was found that frictional coefficient and specific wear rate improved when filled with PP-g-MA. This may be attributed to the change in the mode of wear mechanisms according to PP-g-MA uses or not.

## INTRODUCTION

Natural fibers reinforced polymer biomass composites have been widely used as materials in various applications in recent years. Many reports have been presented on the processing, characterization, rheological and mechanical properties of these biomass composites [1-3]. To further enhance the eco-friendliness of materials, there is a strong need to use not only reinforcement fibers but also biopolymers obtained from plant-derived materials as the matrix polymer of these composites [4-6]. However, very few studies have been conducted on the use of biomass composites as tribomaterials, which are often applied for manufacturing mechanical sliding parts such as gears, bearings and seals [5-9]. In previous studies, we conducted the development of new polymeric tribomaterials made of 100% inedible plant-derived materials [4-6, 10-13]. In particular, we investigated the thermal, rheological, mechanical and tribological properties of various natural fiber such as hemp, sisal, jute and ramie fiber, reinforced plant-derived polyamide 1010 (PA1010) biomass composites. However, in order to develop further higher performance in these biomass composites, it is important to improve the interaction and interfacial adhesion of natural fiber and matrix polymer and the fiber dispersion in the composites. However, little interest has been paid to the effect of addition of compatibilizer such as maleic anhydride grafted polypropylene (PP-g-MA) on the tribological properties of these biomass composites. Therefore, it is strongly required to investigate the effect of addition of PP-g-MA on the tribological properties of these biomass composites. To develop a new polymeric tribomaterial made of only inedible plant-derived materials, this study aimed to experimentally investigate the effect of addition of PP-g-MA on the tribological properties of hemp fibers-reinforced plant-derived polyamide 1010 biomass composites.

## EXPERIMENTAL

The materials used in this study were hemp fiber-reinforced plant-derived polyamide 1010 biomass composites surface-treated using ureidosilane coupling agent and the blend of these composites and maleic anhydride grafted polypropylene. Plant-derived polyamide 1010 (PA1010, Vestamid Terra DS16, Daicel Evonic Ltd., Japan) was used as the matrix polymer. Maleic anhydride grafted polypropylene (PP-g-MA, Umex1010, Sanyo Chemical Industries, Ltd., Japan) was used as a compatibilizer. The composition of the blend of PA1010 and PP-g-MA was fixed as 98/2 by weight fraction. Hemp fiber (HF,  $\phi$ 50-100 $\mu$ m, Hemp Levo Inc., Japan) was used as a reinforcement fiber. HF was pre-cut into 5 mm pieces and surfaced-treated by two types of surface treatment: (a) alkali treatment by sodium chlorite (NaClO<sub>2</sub>) solution and (b) surface treatment by ureidosilane coupling agents [4, 13]. Alkali treatment by NaClO<sub>2</sub> was employed as follows: 5% NaClO<sub>2</sub> solution was placed in a stainless beaker. Chopped HFs were then added to the beaker and stirred well. This was kept at room temperature for 4 h. The fibers were then washed thoroughly with water to remove the excess NaClO<sub>2</sub> sticking to the fibers. The alkali-treated fibers were dried in the air for 24 h and in a vacuum oven at 80 °C for 5 h. The surface treatment of HFs with the concentration of 1 wt.% ureidosilane (3-ureidopropyltrimethoxy silane, A-1160, Momentive Performance Materials Inc., Japan) was carried out in 0.5 wt % acetic acid aqueous solution where the pH of the solution was adjusted to 3.5 and stirred continuously for 15 min. Then, the fibers were immersed in the solution for 60 min. After treatment, fibers were removed from the solution and dried in air for 24 h and in a vacuum oven at 80 °C for 5 h. The volume fraction of fiber  $V_f$  was fixed with 20 vol % in this study. The code and composition of various HF/PA1010/PP-g-MA used in this study are listed in Table 1.

**TABLE 1.** Code and composition of various HF/PA1010/PP-g-MA biomass composites used in this study

Code	PA1010 (vol.%)	PP-g-MA (vol.%)	HF (vol.%)	Surface treatment
PA1010	100	-	-	-
HF	80	-	20	-
S-HF	80	-	20	NaClO <sub>2</sub> + A-1160
HF/C	78.3	1.7	20	-
S-HF/C	78.3	1.7	20	NaClO <sub>2</sub> + A-1160

Various HF/PA1010/PA-g-MA biomass composites were extruded using a twin screw extruder and injection-molded [6, 14]. All the components were dried for 12 h at 80 °C in a vacuum oven beforehand until the moisture level was below 0.2%, then dry blended in a small bottle, and subsequently the melt was mixed at 85 rpm and 220 °C using a twin screw extruder (TEX-30HSS, Japan Steel Works, Ltd., Tokyo, Japan). After mixing, the extruded strands of various HF/PA1010/PP-g-MA biomass composites were cut into 5 mm pieces using a pelletizer and dried again at 24 h at 80 °C in a vacuum oven. HF/PA1010/PP-g-MA biomass composite samples of various shapes for various experiments were injection molded (NS20-A, Nissei Plastic Industrial, Nagano, Japan). The molding conditions were as follows: cylinder temperatures of 220 °C, mold (cavity) temperature of 30 °C, and the injection rate of 13 cm<sup>3</sup>/s. To maintain the dry conditions of the samples in all the measurements, they were kept in accordance with JIS K 6920-2 for at least 24 h at 23 °C in desiccators after molding.

The tribological properties were measured using a ring-on-plate type sliding wear tester (EFM-3-EN, Orientec, Co. Ltd., Tokyo, Japan) [6, 14]. The tests were conducted using plate-shaped test specimens (30 mm × 30 mm × 3 mm) with the surface ( $R_a = 0.4 \mu$ m) finished by No. 240 and No. 800 polishing paper at room temperature under dry conditions in accordance with JIS K 7218a. A carbon steel (S45C) ring ( $\phi$ 20 mm ×  $\phi$ 25.6 mm × 15 mm, apparent contact area  $A_a = 2 \text{ cm}^2$ ) with the surface ( $R_a = 0.4 \mu$ m) finished with the same polishing paper was used as a metal counterpart. The sliding wear test under constant normal and constant sliding velocity was conducted at the normal load  $P$  of 50 N (apparent contact pressure  $p_a = 0.25 \text{ MPa}$ ), the sliding velocity  $v$  of 0.2 m/s, and the sliding distance  $L$  of 600 m. Tribological properties were evaluated by the kinetic frictional coefficient  $\mu$  and specific wear rate  $V_s$ . The kinetic frictional coefficient was calculated from the average value of the test distance range of 400 m to 600 m in which the frictional coefficient is stable. The specific wear rate was calculated by the weight of the test specimen before and after test. The wear debris were observed using a SEM with osmium (Os) sputter coated.

## RESULTS AND DISCUSSION

The tribological properties of various HF/PA1010/PP-g-MA biomass composites using a ring-on-plate type sliding wear tester under constant normal load and constant sliding velocity, and under dry conditions are discussed. Figure 1 shows the relationship between the specific wear rate  $V_s$  and the frictional coefficient  $\mu$  of various HF/PA1010/PP-g-MA biomass composites with or without the alkali treatment by  $\text{NaClO}_2$  and the surface treatment by ureidosilane coupling agent. The  $\mu$  and  $V_s$  of neat plant-derived PA1010 (100%) improved when filled with HF and PP-g-MA and surface-treated by the combination of  $\text{NaClO}_2$  alkali treatment and ureidosilane coupling agent. The  $\mu$  and  $V_s$  of various HF/PA1010/PP-g-MA biomass composites decrease in the following order: PA1010  $\gg$  HF > HF-S > HF/C > HF-S/C. These results suggest that the combinations of PP-g-MA as a compatibilizer and the surface treatment of fiber, which is alkali treatment by  $\text{NaClO}_2$  and the surface treatment by ureidosilane, are the most effective method for the enhancement of tribological properties such as frictional coefficient and specific wear rate. These may be attributed to the change in the mode of wear mechanisms according to the addition of the PP-g-MA and according to whether surface treatment uses or not. In other words, these are considered to be due to change in the interaction and interfacial adhesion between fiber and matrix polymer and dispersion of fiber in the composites by the addition of PP-g-MA and the surface treatment of fiber. Thus, it is necessary to observe the wear debris in order to understand the mechanisms of tribological properties, because polymer composites are strongly influenced by their ability to form these [6,14]. Figure 2 shows the SEM photographs of the wear debris of various HF/PA1010/PP-g-MA biomass composites: (a) neat PA1010 (100%), (b) HF-S, and (c) HF-S/C, respectively. These examples of wear debris were collected from the outside of the sliding wear test to determine the wear mechanism of various HF/PA1010/PP-g-MA biomass composites. The shape and size of the wear debris changed when filled with HF and PP-g-MA. Those of PA1010 (Fig. 2(a)) were small and long filamentary (roll) particles [6]; those of HF-S (Fig. 2(b)) were many large flaky particles; and those of HF-S/C (Fig. 2(c)) were many thick granular particles. Therefore, these differences may support the change in the mode of wear mechanism when filled HF and PP-g-MA caused by good interfacial interaction between HF and PA1010 and good dispersion of fiber in the composites. However, it is necessary to further clarify the above mechanisms by carrying out detailed observations of the structural changes of frictional surface of various HF/PA1010/PP-g-MA biomass composites such as the metallic counterface and worn surface.

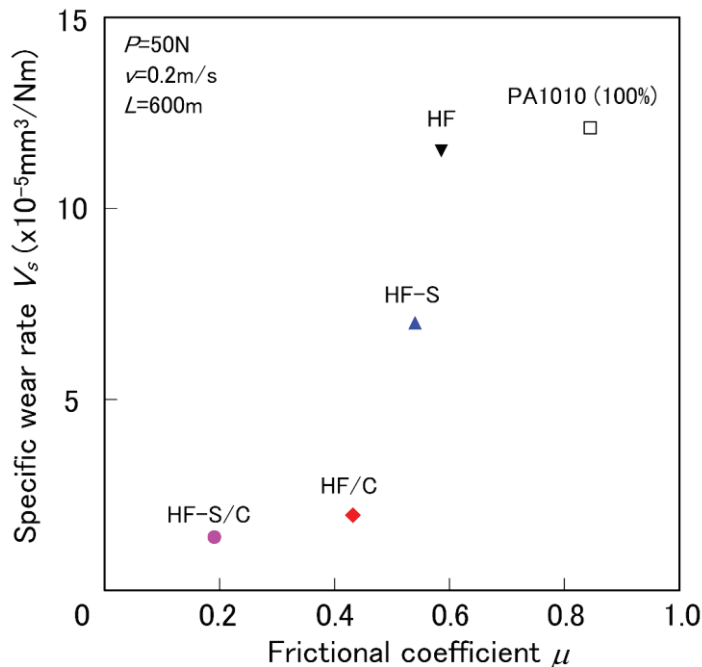
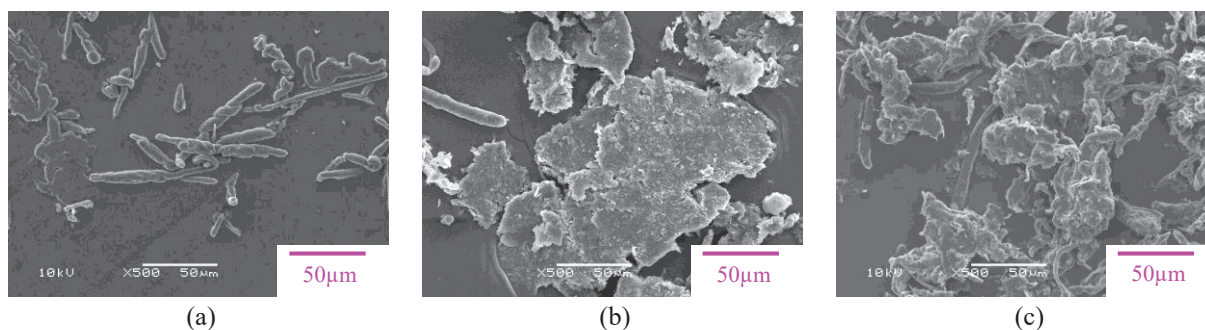


FIGURE 1. Relationship between specific wear rate and frictional coefficient of various HF/PA1010/PP-g-MA biomass composites.



**FIGURE 2.** SEM photographs of wear debris after sliding wear tests of various HF/PA1010/PP-g-MA biomass composites: (a) neat PA1010, (b) HF/S, and (c) HF-S/C.

## CONCLUSION

To develop a new polymeric tribomaterial made of only inedible plant-derived materials, this study aimed to experimentally investigate the effect of addition of PP-g-MA on the tribological properties of hemp fibers-reinforced plant-derived polyamide 1010 biomass composites. It was found that the frictional coefficient and the specific wear rate improved when filled with HF and PP-g-MA and the surface treatment of fiber, which is alkali treatment by NaClO<sub>2</sub> and surface treatment by ureidosilane coupling agent. In particular, the combinations of PP-g-MA as a compatibilizer and the surface treatment of fiber were the most effective method for the enhancement of tribological properties such as frictional coefficient and specific wear rate.

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