Improvement of Strength and Conductivity in Composite Materials with Rice-Hull Silica Carbon

M.Shishido*, Y.Kurita**, M.Ishikawa** and H.Iizuka**

 Doctor's Program of System and Information Engineering, Graduate School of Science and Engineering, Yamagata University, 4-3-16 Jonan, Yonezawa 992-8510, Japan Fax: +81-238-26-3212, e-mail: dhd91773@dipfr.dip.yz.yamagata-u.ac.jp
**Faculty of Engineering, Yamagata University, 4-3-16 Jonan, Yonezawa 992-8510, Japan

Rice hull, which is one of the agricultural wastes in Japan, is required to be reused from a viewpoint of the recycling. The rice-hull silica carbon (RHSC) powder is prepared by mixing the rice hull particles with a phenol resin, and then carbonizing in nitrogen gas atmosphere at high temperatures. One of the effective utilization of the RHSC powder is as a filler for plastics and rubbers to improve the mechanical strength and the electrical conductivity. In this study, the improvement of bending strength of the RHSC/Polyphenylenesulfide (PPS) composites and electric conductivity of the RHSC/Etylene propylene diene methylene linkage (EPDM) composites were investigated by kneading the RHSC powder with PPS and EPDM, respectively. The bending strength was slightly improved and the conductivity was gradually improved with the amount of the RHSC powder. Especially, the dependence of the conductivity on the amount of the RHSC powder is utilizable for the production of antistatic polymers and rubbers.

Key words: Rice-Hull, Carbonization, Composite Materials, Conductivity, Mechanical Strength

1. INTRODUCTION

Rice hull is a residual product of the rice, which amount is about 2.6 million tons per year in Japan [1]. Namely, the rice hull is one of the agricultural wastes, and is required to be reused from a viewpoint of the recycling. The rice hull contains about 20mass% of inorganic constituent and 80mass% of organic constituent. The main inorganic constituent is silica, which amount is more than 95mass% [2]. The authors have investigated a new industrial material that was manufactured from the rice hull [3,4].

The rice-hull silica carbon (RHSC) powder is manufactured by impregnating the rice hull with a phenol resin, and then carbonizing in nitrogen gas atmosphere at high temperatures. The RHSC powder is a hard particle, since it contains about 30mass% of silica from the rice hull and about 70mass% of the glass-like carbon that is made from phenol resin.

The molded material of the RHSC can be applied to a dry sliding element for a linear motion bearing, because it has excellent water resistance, frictional [5] and abrasion resistance property under unlubricating conditions. Moreover, the RHSC powder is expected to be used as a filler for the plastics and rubbers, since it provides new functions for the composite material. Especially, the mechanical strength and the frictional property are expected to be improved with its high mechanical performance [6]. Moreover, the RHSC is considered to have a unique feature for the electrostatic property because the RHSC contains high conductive carbon, low conductive Si and many vacancies in the structure. The electrostatic property is important for the production facility of an electronic device [7].

In this study, the improvement of mechanical strength

and conductivity were investigated by kneading the RHSC powder with polyphenylenesulfide (PPS) and ethylene propylene diene methylene linkage (EPDM).

2. EXPERIMENTAL PROCEDURE

2.1 Manufacturing process

Fig.1 shows the manufacturing process of the RHSC powder and RHSC composites. At first, the rice hull is impregnated with a thermosetting phenol resin (25mass%), and then carbonized in nitrogen gas atmosphere at 900°C. The RHSC was then pulverized by a mill to obtain the RHSC powder. The RHSC powder was sieved by 106 μ m meshed filter to obtain the powder of 60 μ m median diameter. In addition, the RHSC powder of 1.8 μ m median diameter is obtained by milling the RHSC for 10 hrs by a vibrating mill.

Fig.2 (a) shows the natural porous structure of the rice hull. The diameter of the unit cell is about $10\mu m$. The macrostructure of the RHSC powder is shown in Fig.2 (b). The natural porous structure is retained after the carbonization and the pulverization.

2.2 Testpieces

The PPS is known as a high molecular weight material and widely used as the machine elements such as valves and gears. The RHSC powder of 1.8µm median diameter was dispersed in the PPS matrix. In order to improve dispersibility, the surface of the RHSC was treated by an aluminate coupling agent. The testpiece was obtained by kneading with an uniaxial kneading machine, and by forming with an injection molding machine. The untreated testpiece was also prepared by the similar



Fig.1 Manufacturing process of RHSC powder and RHSC composites



Fig.3 Dispersion of RHSC particles of RHSC/PPS composite (treated)

manufacturing procedure in order to confirm the effect of surface treatment on the dispersibility.

Fig.3 shows the dispersion of the RHSC particles of the RHSC/PPS composites with the surface treatment. The RHSC powder is almost homogeneously dispersed in the PPS matrix.

The EPDM is widely used as the industrial materials for its low cost and high weather resistance. The RHSC powder of 60μ m median diameter was dispersed in the EPDM matrix kneaded with a roll kneading machine with various additives. The amount of the RHSC powder was increased



(b) Testpiece for fracture strength



from 50mass% to 400mass% in steps of 50mass%. After kneading, the material was vulcanized, and then molded at $170\pm5^{\circ}$ C in vacuum. The amount of the RHSC powder was increased from 40phr to 160phr in steps of 10phr. The rubber composite material with CarbonBlack (GPF-HS, Asahi carbon Co.Ltd., Japan) was also prepared for the reference.

2.3 Mechanical tests

The testpieces for the mechanical strength are shown in Fig.4. Fig.4 (a) shows the testpiece for bending strength. The bending test was carried out referring to the Japanese Industrial Standards of JIS-K7171. The test was carried out using a universal testing machine with 5kN maximum load. The crosshead speed was 10mm/min. Schematic figure of the three-point bending for fracture strength measurements is shown in Fig.4





Fig.6 Effect of RHSC powder addition on bending strength of RHSC/PPS composites

composites		
	Elastic modulus [GPa]	Yield stress [MPa]
PPS	2.57	101
10wt.% (untreated)	2.64	107
10wt.% (treated)	2.72	105
20wt.% (treated)	2.7	110
30wt.% (treated)	2.98	112

Table I Mechanical properties of RHSC/PPS

(b). The notch was introduced by using a thin diamond wheel. The radius of the wheel-tip curvature was 0.5mm.

Fig.5 shows the testpiece of the composite materials for the measurement of surface resistivity and volume resistivity. The surface resistivity was measured with referring to the Japanese Industrial Standards of JIS-K7194 with nine measureing sites.

3. EXPERIMENTAL RESULTS

3.1 Mechanical properties

Fig.6 shows the effect of the RHSC powder addition on the bending strength of the RHSC/PPS composites with the surface treatment. The ductility decreases with increasing the amount of the RHSC powder. The mechanical properties of the







Fig.8 Effect of surface treatment on bending strength of RHSC/PPS composites

RHSC/PPS composites are listed in Table I. The bending strength and the elastic modulus are improved by the adding of the RHSC powder.

Fig.7 shows the effect of the RHSC powder addition on the fracture strength of the RHSC/PPS composites with the surface treatment. The fracture strength decreases with adding the amount of the RHSC powder. The fracture strength usually decreases with the adding of second phase particles, because the interface usually de-bonds and provides the fracture initiation sites [8].

Fig.8 shows the effect of the surface treatment on the bending strength of the RHSC/PPS composites. The fracture ductility is largely improved by the surface treatment, although the maximum bending strength of the surface treated testpiece is almost equal to that of the untreated testpiece.

Fig.9 shows the effect of the surface treatment on the fracture strength of the RHSC/PPS composites. The fracture strength is not affected by the surface treatment. It is considered that the adhesive strength between the particles and the matrix is not changed and the dispersion of the particles is also not changed even under the untreated condition.

3.2 Electric characteristics

Fig.10 shows the surface resistivity of the RHSC/EPDM composites at the nine measuring sites.



Fig.9 Effect of surface treatment on fracture strength of RHSC/PPS composites



Fig. 10 Relationship between measuring site and surface resistivity of RHSC/EPDM composites

The surface resistivity increases with increasing the filler content. Moreover, since all the surface resistivity is almost homogeneous at each site, it is confirmed that the RHSC powder is uniformly dispersed.

Fig.11 shows the comparison of volume resistivity between the RHSC powder and the GPF-HS. The resistivity volume gradually decreases with increasing the amount of the RHSC powder in the RHSC/PPS composite. Under the favor of the gentle slope of the characteristic curve, it is easy to obtain the designed value for the volume resistivity by controlling the amount of the RHSC powder. By contrast, the volume resistivity drastically decreases with increasing the amount of the GPF-HS. Namely, the CarbonBlack induces the percolation phenomenon, in which the volume resistivity drastically decreases with increasing the amount of filler [9]. Therefore, it is necessary to precisely control the amount of filler to obtain a designed value of the volume resistivity.

4. CONCLUSION

The mechanical strength and conductivity were investigated for the composite materials of the RHSC powder adding in the PPS and the EPDM, respectively. The summary of the obtained results was described as follows.

(1) The bending strength of the PPS was improved by the adding of the RHSC powder.



Fig.11 Comparison of volume resistivity between RHSC powder and GPF-HS

- (2) The volume resistivity gradually decreased with increasing the amount of the RHSC powder in the EPDM. Therefore, it is easy to obtain the designed value of the volume resistivity by controlling the amount of the RHSC powder.
- (3) The RHSC powder was considered to disperse almost uniformly in the PPS and EPDM, respectively.

ACKNOWLEDGEMENTS

The authors wish sincerely to express their gratitude to Sanwa Yushi Co. Ltd. and Miyasaka Polymer Co. Ltd. for the preparation of the testpieces.

REFERENCES

- [1] T.Ishitani, K.Ohtsubo, "The Science of Rice (in Japanese)", Asakura Shoten, Tokyo (1995) p.172.
- [2] S.Chandrasekhar, K.G.Styanarayana, P.N.Pramada, P.Raghavan, "Processing Properties and Applications of Reactive Silica from Rice Husk", J. Mater. Sci., 38, 3159-68 (2003).
- [3] H.Iizuka, G.Kato, K.Igarashi, S.Shikano and T.Takahashi, "Mechanical Properties of Porous Carbon Materials made from Rice Bran", J. Soc. Mater. Sci., Jpn, 49, 625-630 (2000).
- [4] M.Shishido, S.Kubo, T.Takahashi and H.Iizuka, "Mechanical Properties of Porous Carbon Materials made from Rice Hull", Trans. Mater. Res. Soc. Jpn., 31, 993-996 (2006).
- [5] K.Yoshida, H.Iizuka, Y.Shibata, H.Unuma, T.Nakamura and S.Shikano, "Water Resistance of Porous Carbon Materials made from Rice Hull", Int. J. Mater. Eng. Resour. 13, 2, 49-53 (2006).
- [6] H.Miyairi, "Omoshiroi-hukugou-zairyou-no-hanashi (in Japanese)", Nikkan Kogyo Shinbunsha, Tokyo (1997) pp.120-125.
- [7] K.Tatebayashi, "Gouseigomu-shinpan (in Japanese)", Nikkan Kogyo Shinbunsha, Tokyo (1966) p.369.
- [8] J.Fujiwara, "The toughness of aromatic polyamide resin", doctoral thesis, Yamagata University (2006) p.89.
- [9] T.Noguchi, "Gomu-no-jiten (in Japanese)", Asakura Syoten, Tokyo (2000) p.424.

(Received February 16. 2007; Accepted July 12, 2007)