INFLUENCE OF SIZING OF GLASS FIBER ROVING ON MECHANICAL PROPERTIES IN GLASS FIBER BRAIDED FABRIC REINFORCED PHENOLIC COMPOSITE

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SUMMARY: This study dealt with the influence of sizing of glass fiber roving on the resin impregnation and the mechanical properties in glass fiber flat braided fabric reinforced phenolic resin composite. The wetting behavior and the influence on the interfacial adhesion were evaluated for the glass fiber rovings with different amount of binder in sizing. The amount of binder affected both wetting and interfacial adhesion, and the optimum amount of binder existed in the roving. By using these rovings, the fabrics as reinforcement were braided, and the wetting behavior and composite properties were evaluated. In the braided fabrics, the wetting behavior was different from the case of roving. Such difference influenced the tensile strength of the composites. The tensile strength of the composites was improved with increasing the amount of binder. The higher amount of binder brought the strong binding of the roving and less damages of the fibers. These effects improved the tensile strength of the braided fabric reinforced phenolic composite.

KEYWORDS: Braided fabric, Phenolic resin, Sizing, Binder, Wetting behavior, Interfacial adhesion

INTRODUCTION

Fiber reinforced plastics have been widely used in various applications, and they have been applied to the structural materials in the transportation industries. In such application, the incombustibility and thermal resistance as well as high specific modulus and strength are important properties. Among the matrix resin for composites, phenolic resin is the most suitable resin to satisfy these properties. Phenolic resins can sustain temperatures of up to 200°C with very low toxicity and low smoke emission[1]. In addition, they are considered fire-restricting materials, as they can fulfill function in an established fire. However, phenolic resin produces H_2O during reaction process in curing and the nature of phenolic resins is brittle[2]. These undesirable natures lead poor mechanical properties of phenolic composites, and therefore it is difficult to apply phenolic resin as matrix of composites in transportation industries[3]. However, such undesirable natures may be avoided by finding the

optimum molding condition. One of the suitable molding techniques for phenolic composite is the resin transfer molding (RTM). In RTM the preformed glass fiber textile is often used as the reinforcement to get complex shaped structure. In order to get easily the complex shaped structure, the braided fabric is considered as one of the most suitable reinforcement for RTM process. However, phenolic resin does not have any chemical reactive function with silane coupling agent on the glass fiber, and therefore the resin impregnation into the glass fiber bundle should be one of the most important molding factors to achieve high mechanical properties and good quality of the product. The resin impregnation into the fiber bundle is affected by the sizing of the fiber bundle. Generally, the fiber bundle for braiding structure is treated by a great amount of binder in sizing in order to reduce the fiber damage during braiding process. The author has clarified that sizing of the fiber bundle affects the resin impregnation and fiber distribution in cross-sectional area[4]. Same effects may be also brought to the braided composite. In the previous paper, the authors discussed the influence of molding condition on the tensile properties of the flat braided fabric reinforced phenolic composite[5], and it was clarified that the good impregnation into the fiber bundle was important to get high tensile strength.

From these backgrounds, this study discussed the influence of sizing on the tensile properties of glass fiber flat braided fabric reinforced phenolic resin composite. The glass fiber rovings with different amount of binder in sizing were used as reinforcement, and the influence of binder on the wetting behavior and mechanical properties were investigated for both the roving and braided fabric reinforced composite.

EXPERIMENTAL

Three kinds of rovings with different in the amount of sizing (binder) were used as fiber bundle for flat braided fabrics. Table 1 summarizes the rovings used in this experiment. Filament diameter, tex and number of filaments were the same, and only the amount of binder was different. In Table 1 the amount of binder is indicated relatively against the amount for the Medium bundle. The roving with medium binder is a standard fiber bundle for braiding. Matrix resin used was novolac based phenolic resin (Shonol BRL-240, Showa High Polymer Co., Ltd., Japan).

In order to evaluate the influence of binder on resin impregnation, the wetting measurement was performed for the fiber bundle. Five fiber bundles were combined into one bundle in order to get enough amount of resin uptake. The wetting behavior was measured by the Wilhelmy type dynamic wetting instrument (WET-6000, Rhesca Co., Ltd., Japan). The

Table 1 Glass fiber rovings for braided fabric.

Type	Low	Medium	High
Filament diameter (μm)	17	17	17
Tex	575	575	575
Number of filament	1000	1000	1000
Relative amount of binder	0.1	1	2
Weight loss after burned-out (%)	0.05	0.4	0.8

combined fiber bundle advanced into the resin at a constant speed of 0.1mm/min. After reaching the dipped length of 9.9mm, the advancing was stopped and the fiber bundle kept dipping in the resin for 180sec. During these processes, the change of tension force was measured. After measuring the wetting behavior, the fiber bundle with resin was cured, and the resin impregnation into the bundle was observed by the optical microscope. In addition, the lateral compression test was performed for the resin impregnated unidirectional fiber bundle composite in order to discuss the relation between the resin impregnation and the mechanical property. The fiber bundle was dipped into the resin under vacuum condition to impregnate the resin. After impregnation process, the fiber bundle with resin was pulled into the PTFE tube mold with an inner diameter of 6mm covered by aluminum tube, and were cured at 80°C for 1hour. After complete curing, test specimens were cut from it to ϕ6mm × 6mm. In the lateral compression test, the load was applied perpendicular to the long direction of the specimen by using the Instron type universal testing machine (Autograph AGS-1000B, Shimadzu Corp., Japan) at the constant cross-head speed of 0.5mm/min. The lateral compressive strength (σ_c) was evaluated by

$$\sigma_c = \frac{P_c}{\ell \times d}$$

After testing, the cross-section was microscopically observed in order to discuss the fracture behavior.

The wetting measurement was also performed for the flat braided fabric made of three kinds of rovings listed in Table 1. Wetting behavior of the braided fabric was measured in the same manner with the case of the roving. In order to discuss the influence of the binder on the mechanical property of braided fabric reinforced composite, glass fiber flat braided fabric reinforced phenolic resin composites were fabricated for three kinds of the fabrics with different amount of binder. The resin was impregnated into the braided fabric under vacuum condition at room temperature for 20 minutes. After this process, the resin-impregnated fabric was placed in the silicon mold with top and bottom glass plates, and it was cured at 60°C for 2 hours. The geometry of the specimen was 220mm in length, 17mm in width and 1mm in thickness. The tensile test was performed by using the Instron type universal testing machine (Autograph AGS-1000B, Shimadzu Corp., Japan) at a constant cross-head speed of 1mm/min with the gauge length of 120mm at room temperature. During the tensile test, the acoustic emission (AE) activity was monitored by the AE instrument (7600 series, NF Corp., Japan). AE transducer with the resonant frequency of 140kHz was attached on the surface of the specimen, and the AE event count and the maximum AE amplitude were monitored during the tensile test. After tensile test, fracture surfaces of the specimens were observed by scanning electron microscope.

RESULTS AND DISCUSSION

Influence of Binder on Wetting and Compressive Strength in Glass Fiber Roving

Fig.1 shows the typical wetting tension diagrams for three kinds of glass fiber rovings. In advancing process, the tension force went down for all the fiber rovings, and the resin did not

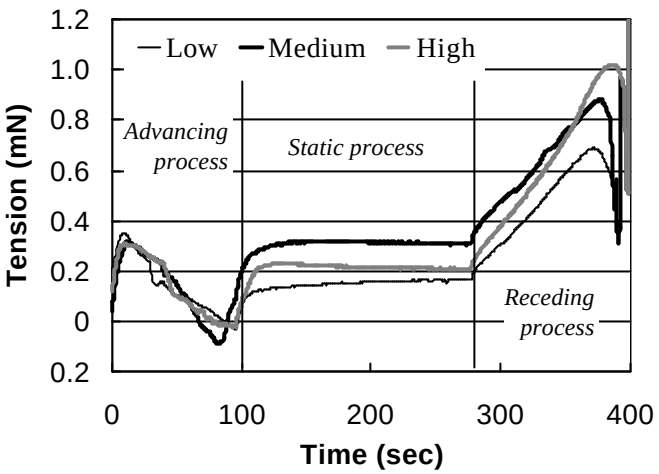


Fig.1 Wetting tension diagram for glass fiber rovings.

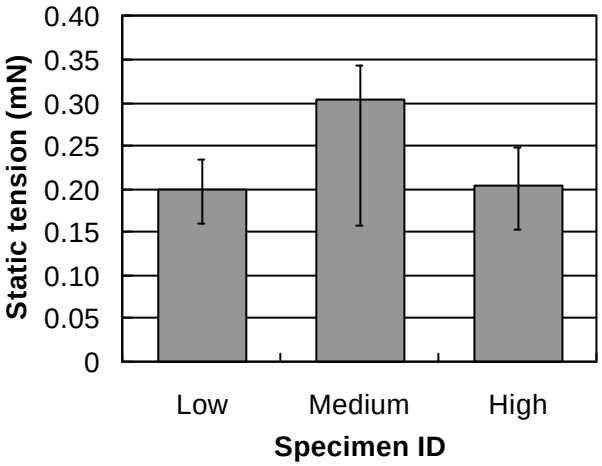
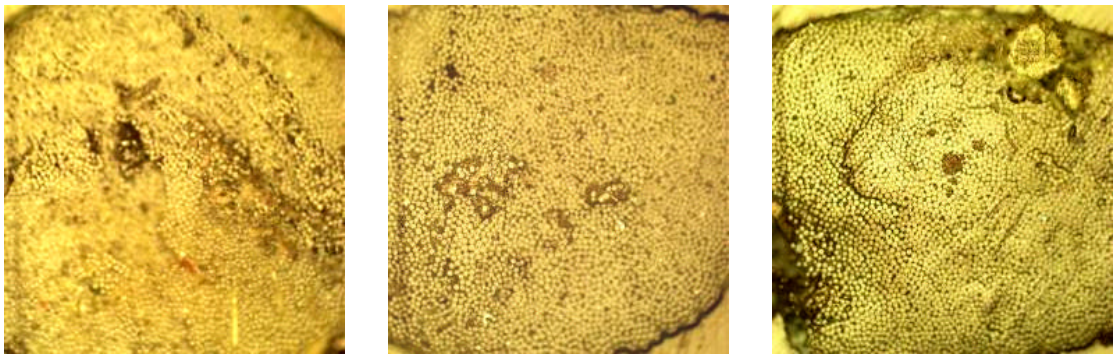


Fig.2 Static tension in the wetting measurement for glass fiber rovings.

impregnate into the rovings. In the Low roving, the tension force kept going down in the advancing process, and it began to go up from the beginning of static process. However, the increase of the tension was a very few. In the High roving, the tension force also kept going down in the advancing process, however, it increased rapidly from the beginning of the static process. The tension force soon reached the highest value in the static process, and it gradually decreased. On the other hand, the tension force began to increase from the last stage in the advancing process, and it showed remarkable increase to the static process. In the static process the tension force kept almost constant. In the receding process, the tension force increased remarkably for all the rovings, however, the amount of force increase depended on the amount of binder. The change of tension force in the wetting measurement means the resin impregnation into the roving. Therefore the resin impregnation into the roving was significantly influenced by the amount of binder. From the results of wetting measurement, the static tension was obtained as the difference between the maximum tension in the static process and the minimum value in the advancing process. Fig.2 shows the influence of amount of binder on the static tension in the wetting measurement. The static tension of the Medium roving showed the highest value, and the tension of the Low and the High rovings were almost same value. This result suggested that the suitable amount of binder existed for getting good resin impregnation into the roving.



(a) (b) (c)

Fig.3 Cross-sectional micrographs of the resin-impregnated rovings after wetting measurement; (a) Low, (b) Medium and (c) High rovings.

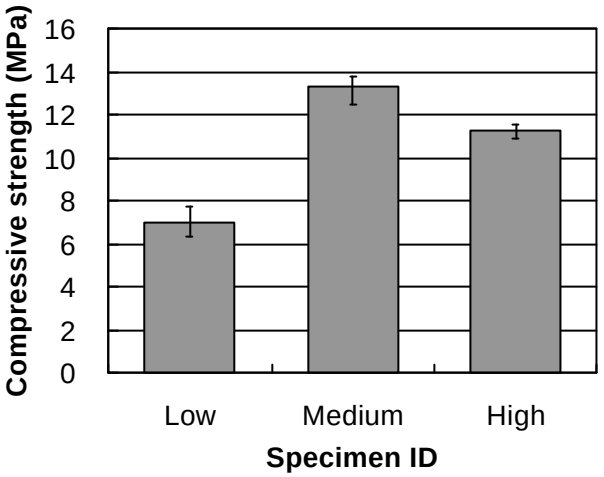


Fig.4 Comparisons of lateral compressive strengths of resin-impregnated fiber rovings.

In order to discuss the resin impregnation into the roving, the cross-section was microscopically observed for the resin-impregnated rovings after wetting measurement. Fig.3 is the cross-sectional micrographs of the resin-impregnated rovings after wetting measurement. In the Low roving, the resin did not impregnate well into each roving, and each roving was separated by the resin rich region. In addition, many voids could be observed on the cross-section. In the Medium and the High rovings, the resin was impregnated well into each roving, and few voids remained inside the. In the Medium roving the rovings dispersed into the filaments, and the filaments were distributed uniformly. Such distribution of the filaments brought the higher uptake of the resin during the wetting measurement in the Medium roving. In the High roving, however, the roving still kept strong binding, and boundary of each roving still remained. Such difference of the filament distribution caused the remarkable difference of the resin uptake between the Medium and the High rovings. From the results of wetting measurement and microscopic observation, the best resin impregnation could be obtained in the Medium roving.

Fig.4 shows the influence of the binder on the lateral compressive strength of the resin-impregnated roving specimens. The influence of the amount of binder on the lateral compressive strength was similar to that on the static tension in the wetting measurement. The strength of the Medium specimen showed the highest value among these specimens.

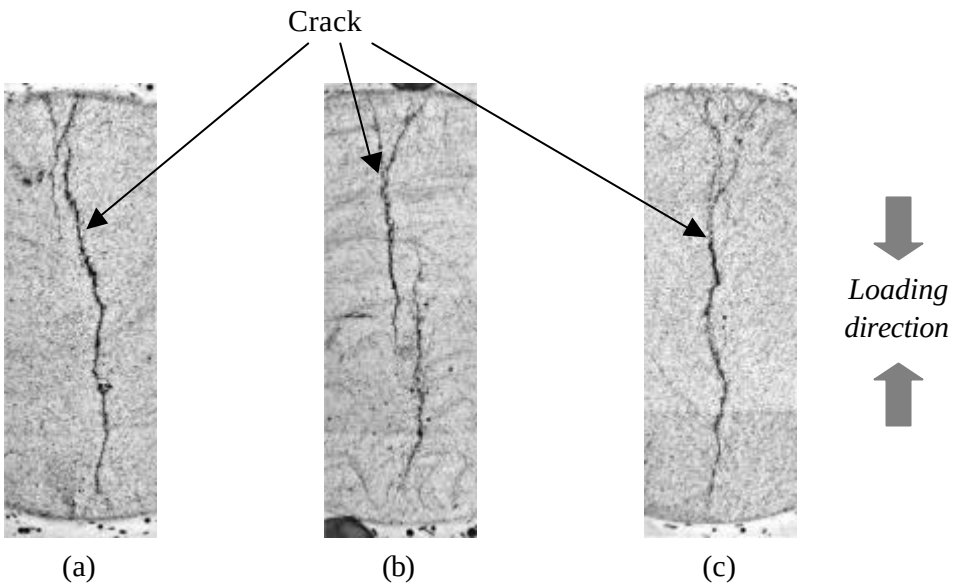


Fig.5 Fracture aspects in the lateral compression test;
(a) Low, (b) Medium and (c) High specimens.

However, the strength of the High specimen also showed higher value than that of the Low specimen. In the wetting measurement, the static tension of the High specimen was almost same with the Low specimen, however, this tendency was different in the compressive strength. The fracture aspects after the compression test are shown in Fig.5. In all the specimens, the cracks propagated from the center of the specimen parallel to the loading direction. It is considered that these cracks were caused by the tension force perpendicular to the loading direction and that these indicated the influence of binder on the interfacial adhesion. In the Medium specimen, which showed the highest strength, two main cracks initiated and propagated from the center of the specimen, and the scale of these cracks changed during propagation process. In addition, microcracks progressed to several ways as very short cracks during the main crack propagation. On the other hand, the single crack propagated from the center of the specimen in the Low specimen, and few separations of the microcracks could be observed. In the High specimen, the crack propagation behavior was looked like a combination of the Medium and the Low specimen. Such difference in the crack propagation caused the difference in the lateral compressive strength. From the results of lateral compression test, it is considered that the good interfacial adhesion could be obtained in the Medium specimen in this paper.

Influence of Binder on Wetting and Mechanical Properties in Braided Fabric Composite

Fig.6 shows the typical wetting tension diagrams for the glass fiber flat braided fabrics with different types of the rovings. In all the fabrics, the tension force decreased in the beginning of the advancing process, and it began to increase prior to the static process. In the Low fabric, the tension force showed remarkable decrease in the advancing process. In the static process, however, the tension force reached almost constant value, and it did not depend on the type of roving. The influence of the amount of binder appeared only in the advancing process, and this tendency was different from the roving shown in Fig.1. The static tension obtained from the wetting measurement is shown in Fig.7. The static tension of the Low fabric showed the highest value, and those of the Medium and High fabrics were almost the

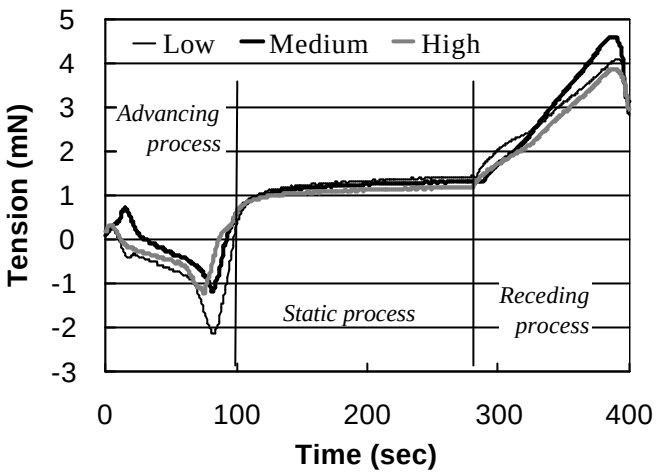


Fig.6 Wetting tension diagram for glass fiber flat braided fabric.

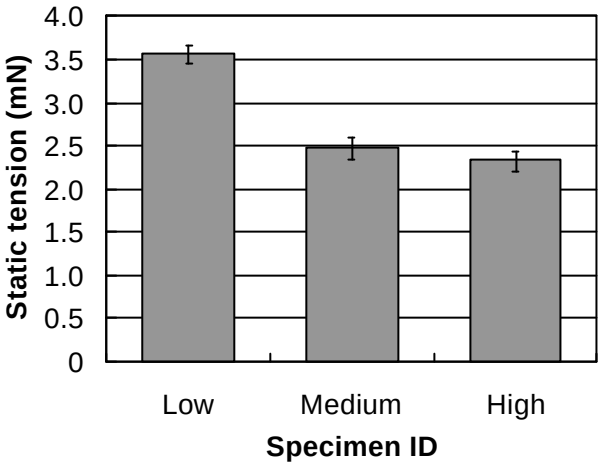


Fig.7 Static tension in the wetting measurement for the flat braided fabrics with different rovings.

same. This tendency was quite different from the result of rovings shown in Fig.2. The differences in the static tension of the fabrics were caused by the difference of the wetting behavior in the advancing process. In the Low fabric, the nap of the fibers could be observed after braiding, and it was caused by the braiding process. The binding force of the filaments in the Low roving was quite poor compared with the Medium and the High rovings, and the roving was easily napped in the braiding process. Such nappy fabric brought resisting force during the advancing process. However, napped fabric could hold the resin on the surface of the fabric, and it brought the higher static tension in the wetting measurement.

Fig.8 shows the comparison of the tensile strength for the glass fiber flat braided fabric composites with different type of roving. The influence of roving on the tensile strength was quite opposite to the static tension shown in Fig.7. The tensile strength increased with increasing the amount of binder, and it depended on the amount of binder. The strength of the Medium specimen was a little higher than that of the Low specimen, and that of the High specimen was much higher than that of the Medium specimen. However, the improvement of the strength was not proportional to the amount of binder. The amount of binder in the

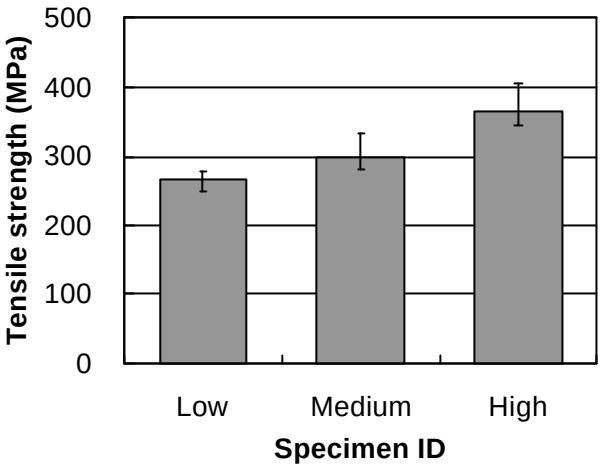


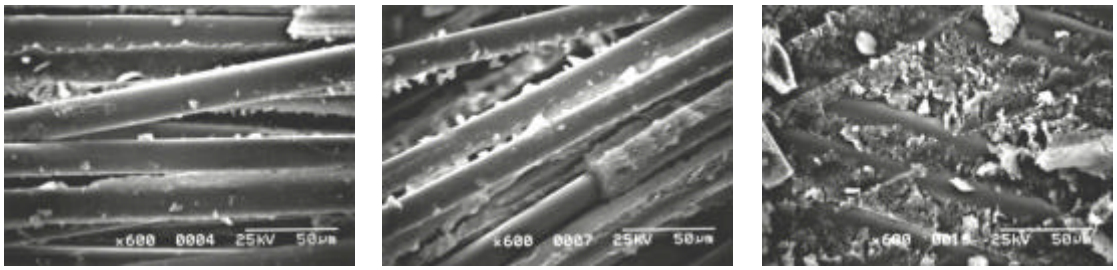
Fig.8 Comparison of the tensile strength for the glass fiber flat braided fabric composites with different rovings.

(a) (b) (c)

Fig.9 Fracture aspects of the flat braided fabric composites after tensile test; (a) Low, (b) Medium and (c) High specimens.

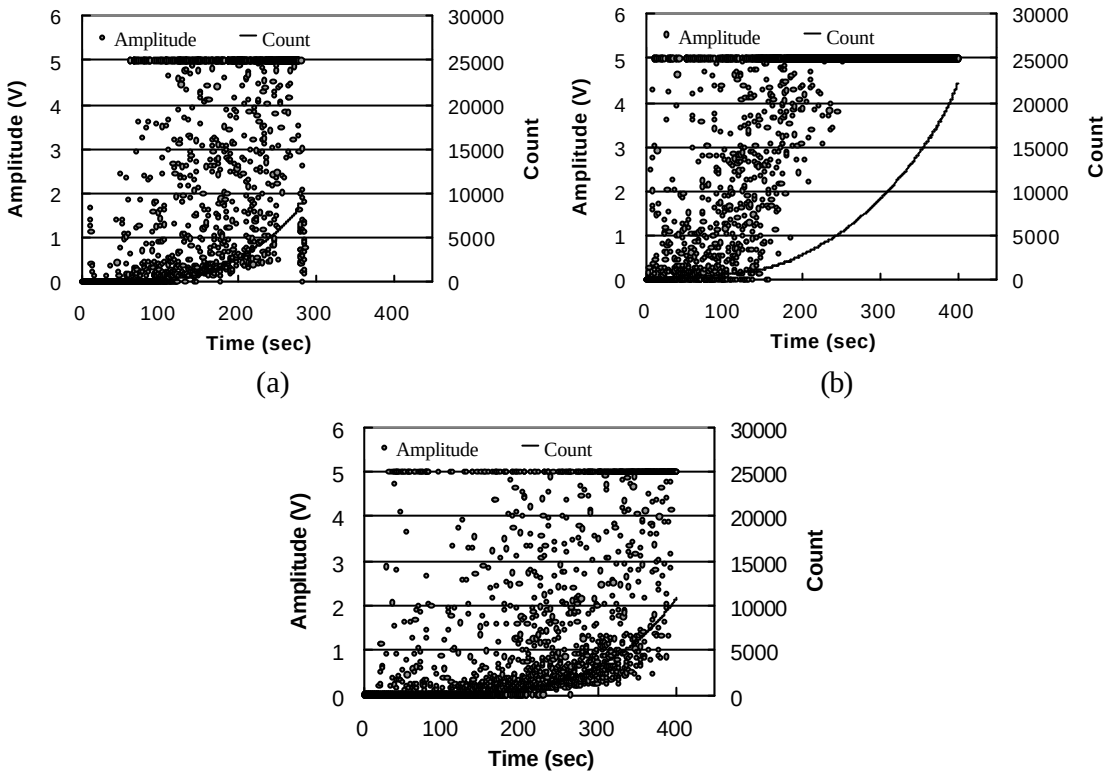
Medium specimen was ten times of the Low specimen, however, the improvement of the strength was a little. On the other hand, the amount of binder in the High specimen was only twice of the Medium specimen, however, the strength of the High specimen was fully improved compared with the Medium specimen.

In order to discuss the influence of the amount of binder on the fracture behavior, the fracture aspects of the specimens were observed after tensile test. Fig.9 are the fracture aspects of the specimens after tensile tests. In the Low specimen, the separation of the matrix resin could be observed on the surface of the specimen, and the long fibers were pulled out from the matrix resin. In addition, there was no resin on the fibers at the fracture surface, and the rovings were opened into filaments. The similar fracture surface was observed in the High specimen, however, the matrix cracks on the specimen surface was a few and the rovings still kept binding. In the Medium specimen, the fracture aspect was different from the others. The top of the fracture surface was quite flat, and the fabric still kept its shape. In addition, the length of the fiber pulled out from the resin was shorter than the others. Fig.10 are the scanning electron micrographs of the fracture surface for each specimen. In the Low



(a) (b) (c)

Fig.10 Scanning electron micrographs at the fracture surfaces of the tensile test specimens; (a) Low, (b) Medium and (c) High specimens.



(a) (b) (c)

Fig.11 Cumulative AE count and the maximum AE amplitude during tensile test; (a) Low, (b) Medium and (c) High specimens.

specimen, the surfaces of fibers were very smooth, and the roving was separated into each filament. This means the interfacial adhesion is quite poor in the Low specimen. With increasing the amount of binder, the amount of the resin on the fracture surface increased, and the rovings kept their binding of the filaments. Especially in the High specimen, a great amount of the resin remained on the fracture surface, and the adhesion between fiber and resin should be stronger than those of the Low and Medium specimens. Such difference in the interfacial adhesion induced the difference in the tensile strength. Fig.11 shows the AE activities during the tensile tests. In the Low and the Medium specimens, the AE events with high amplitude occurred from the beginning of the tensile tests. Generally, the AE event with high amplitude is caused by the fracture related to the fiber breakage. Therefore it is considered that the fiber fracture often occurred from the beginning of the tensile test in these specimens. Such fiber fracture is caused by the damage of the fiber during the

braiding process. On the other hand, the damage of the fiber in the High specimen was less than the Low and the Medium specimens due to the amount of binder, and therefore the fiber breakage was suppressed up to higher applied tensile stress. Such difference in the fiber damage also affected the tensile strength of the braided fabric composite.

CONCLUSION

This study dealt with the influence of sizing of glass fiber roving on the resin impregnation and the mechanical properties in glass fiber flat braided fabric reinforced phenolic resin composite. The wetting behavior and the influence on the interfacial adhesion were evaluated for the glass fiber rovings with different amount of binder in sizing. The amount of binder affected both wetting and interfacial adhesion, and the Normal roving showed the highest wetting behavior and the interfacial strength in the roving tests. This result suggested that the optimum amount of binder existed in the roving. By using these rovings, the fabrics as reinforcement were braided, and the wetting behavior and composite properties were evaluated. In the braided fabrics, the wetting behavior was different from the case of roving, and the less amount of binder brought the higher resin uptake in the wetting measurement. However, the composite with higher amount of binder showed higher tensile strength, and this tendency was opposite to the wetting behavior. The tensile strength was influenced by the interfacial adhesion in the braided fabric and by the fiber damage during the braiding process. The higher amount of binder brought higher binding force of the roving in the fabric and less damage of the fiber during braiding process. According to these desirable effects, the composite made of the fabric with higher amount of binder could achieve higher tensile strength.

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