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Electrical and Mechanical Properties of Carbon/Glass Hybridized Long Fiber Reinforced Polypropylene Composites

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Abstract: The electrical percolation and hybrid effect that forms from long carbon fiber (LCF) and long glass fiber (LGF) hybridized composite in a polypropylene matrix were studied by investigating electrical and mechanical properties. As a process, the electrical and mechanical properties were investigated in terms of LCF loading at constant volume percentage in the total. The electrical resistivities of volume and surface were measured in order to learn the percolation threshold points, which were 8-9 and 10-12 volume percents of LCF loading individually. The mechanical properties, such as, were tensile and flexural modulus of the hybridized composite were obtained and compared with the prediction results using the rule of hybrid mixtures (RoHM) equation. The hybrid effect was observed in the result of the tensile modulus in the range of 6-10 volume percent of LCF loading whereas there was no hybrid effect in flexural modulus. The tensile and flexural strengths of LCF/LGF hybridized composite are 100 and 140 MPa at 20 vol% of LCF loading. The tensile and flexural modulus are approximately 22 and 14 GPa at 20 vol% of LCF. The interaction between reinforced fiber and the matrix was reported and analyzed by scanning electron microscopy (SEM) and heat depletion temperature (HDT). Through the process, the mechanical strength was more related to the interaction between fiber and the polypropylene matrix than the mechanical modulus.

Keywords: long carbon fiber, long glass fiber, hybridized composite, electrical resistivity, mechanical property.

Introduction

Long fiber reinforced thermoplastic (LFRT) composite has garnered much attention among composite researchers due to its high performance, mass processability, and eco-friendly properties. The approaches that are taken vary and range from experimental study to commercial use. LFRT composite exhibits greater strength, stiffness, and dimensional stability than conventional polymers.[1] Thus, the auto industry has extensively studied LFRT composite to take advantage of reinforcement with the fiber to solve challenges for improved fuel efficiency, weight reduction, and to tackle environmental issues. In China, air pollution from an increasing number of automobiles has become a critical issue and there have been many attempts to reduce carbon dioxide through vehicle technology improvements.[2] The U.S. environmental protection agency (EPA) has also set a target for passenger cars where emission levels need to be decreased by 15% compared to current 2012 levels through increased fuel efficiency.[3] These are some of the reasons the automobile industry has turned to fiber reinforced composite as an attractive candidate material with a lighter weight than metal. LFRT composites have been studied and developed for many years and continues to be a promising a high-performance material.

Long glass fiber can enhance composite properties better in terms of mechanical and thermal properties than short glass fiber thanks to the residual length of fiber in a mold. According to Thomason *et al*., fiber length and concentration are significant factors in determining the mechanical and thermal properties of the composites.[4-9] To improve the properties of LFRT composite, various efforts have been studied, including fiber type, modification of fiber surface,[11] and fiber orientation.[12] To be specific, various kinds of fiber such as aramid fiber, natural fiber, and carbon fiber have been examined to find better tensile and flexural properties. Among these fibers, carbon fiber is a key material that has been used as reinforcement in a thermoplastic matrix.

Generally, carbon fiber has been used as reinforcement in a thermoset matrix such as epoxy. The tensile strength and Young's modulus of carbon fiber are 4,000 MPa and 240 GPa. The values are 100% and 300% higher than conventional glass fiber. However, the density of carbon fiber is around 30% lower than glass fiber.[13] Additionally, carbon fiber has good electrical conductivity due to its chemical structure.[14] Thus, carbon fiber can be a potential material for

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future replacement of metals because of its exceptional mechanical and electrical properties. In spite of these important advantages, carbon fiber reinforced thermoplastic composite has not been intensively applied in manufacturing because costs so far have not been competitive. Therefore, reductions in cost and improving physical performance would be key factors to make it viable in a commercial market with mass processability. As such there are numerous studies looking for hybridized composites to fit this need.

For example, a study of short glass fiber (SGF) and short carbon fiber (SCF) reinforced thermoplastic composite was published by Mai *et al*., in terms of hybrid effects.[15] In the paper, the author discussed hybrid effects on tensile strength and failure strain. The tensile properties of SGF/PP and SCF/PP were also investigated by Hu *et al*.[13] Based on the results, composite strength is more dependent on fiber length or fiber aspect ratio, and fiber volume fraction is the more critical factor on composite modulus. Thomason *et al*.,[4] also have investigated theoretical and experimental values of fiber reinforced polypropylene in term of fiber length improvement and they have concluded that composite performance of both theoretical and experimental value are increasing with increasing fiber length as shown in Table I.

Although fiber length is known as a significant factor, the mechanical and electrical properties of long carbon fiber (LCF) and long glass fiber (LGF) hybridized composite has not been studied by many researchers.

In light of this, the goal of the study is to understand electrical percolation and hybrid effects in terms of electrical resistivity and mechanical properties. This paper investigates electrical resistivity by considering volume resistivity and surface resistivity as a function of LCF loading. The hybrid effect of LCF/LGF hybridized composite was also investigated in terms of mechanical properties. The role of the hybrid mixtures (RoHM) equation was applied to find the point that indicates a hybrid effect. The composites were prepared by an injection molding technique and characterized by thermal gravimetric analysis (TGA) and scanning electron microscopy (SEM). The hybrid effect was observed in the result of tensile modulus at a range of 6 to 10 volume of LCF loading, and the interaction between fiber and matrix was more important than fiber type.

Table I. Effect of Fiber Length on Composite Properties in Terms of Theoretical and Experimental Results[4]

Fiber Length Increasing (1.2 mm → 4.2 mm)		Strength (%)	Modulus (%)
Theoretical Data		+ 36	+ 9
Experimental Data	Tensile	+ 51	+ 3
	Flexural	+ 3	+ 6

Experimental

Materials. Commercially available LGFT PP was purchased from SAMBARK LFT Co. LTD with the trade name SUPRAN® PP 1360 which has 60 wt% of glass fiber content in a polypropylene matrix. SUPRAN® PP 1360 was used as a long glass fiber reinforced thermoplastic (LGFT). Long carbon fiber reinforced thermoplastic (LCFT), especially polypropylene, was produced from the same company. In order to improve the compatibility effect, commercially available maleic anhydride (MAH) grafted polypropylene (CM-1120) provided by Honam Petrochemical Corp. at around 1.0 wt% of MAH graft ratio was used as a compatibilizer in a LCFT process. Basic information of materials and fiber reinforced composite was listed in Tables II and III.

Sample Preparation. The hybridized composites of LGFT and LCFT were prepared by simple dry mixing with a given volume ratio as shown in Table IV. The density of LGFT/LCFT hybridized composite is listed in Table V.

Mechanical and electrical specimens were prepared using an injection-molding machine (Model: MD180W i6.5, Man-

Table III. Characteristics of Long Fiber Reinforced Thermoplastic Investigated

	Length of Fiber in Pellet (mm)	Aspect Ratio (L/D)
LGFT	10	580
LCFT	10	1430

Table IV. Sample Preparation and Name with LCF and LGF Volume Ratio

	LCF0	LCF6	LCF10	LCF14	LCF20
LCF vol%	0	6	10	14	20
LGF vol%	20	14	10	6	0
PP vol%	80	80	80	80	80

Table V. Density of Hybridized LCF/LGF Samples

	LCF0	LCF6	LCF10	LCF14	LCF20
Density	1.21	1.19	1.15	1.12	1.09

Table II. Characteristics of Base Materials Investigated

	Density (g/cm^3)	Diameter (μm)	Tensile Strength (MPa)	Tensile Modulus (GPa)	Melt Index (g/10min)
PP	0.9	N/A	34	0.8	34
Glass Fiber	2.6	17	2,500	73	N/A
Carbon Fiber	1.8	7	4,900	230	N/A

ufacture: UBE Machinery) following the ASTM standard for tensile test and flexural test.

Characterization. The tensile test was carried out in accordance with ASTM D638 and the crosshead speed was 10 mm/min. In order to measure flexural strength and modulus, the test was conducted in accordance with ASTM D790 and the cross head speed was 5 mm/min. The tensile and flexural tests were carried out using a universal testing machine (UTM) (Model: 5566, Manufacture: Instron).

The electrical resistivity (volume) of the samples was obtained in accordance with ASTM D4496 using a multimeter (Model: Fluke 187, Manufacture: Fluke Corp.). Silver paste was applied onto the flexural test specimen top and bottom which is perpendicular to resin flow direction. In order to reduce the large deviation of the electrical resistivity when four-terminal measurement was applied at each point, a two-terminal technique was used. The measurement was conducted at room temperature and the results were collected after waiting 5 min. The volume electrical resistivity was obtained by the equation:

$$\rho_v = \frac{RLW}{t} \quad (1)$$

where ρ_v is a volume electrical resistivity, R is a measured resistance, L and W are specimen length and width, and t is a thickness of the specimen.

The surface electrical resistivity of samples was obtained using a four-probe resistivity meter (Model: RS8-1G, Manufacture: DASOL ENG). The surface resistivity of the samples was measured on a rectangular flexural test specimen with ten measurements per sample to reduce the variation of the results.

In order to take microscopic images of distribution and size of LGF and LCF, a scanning electron microscope was used (Model: SM 701, Manufacture: TOPCON). The hybridized composites were immersed in liquid nitrogen for 5 min and then fractured. The samples were prepared by being sputter-coated with gold prior to SEM imaging. In addition to analyzing the distribution of LGF and LCF in the composite, X-ray maps were collected with an energy-dispersive X-ray spectroscopy (EDS) (Model: System 6, Manufacture: NORAN). The images were obtained from the fractured surface of the hybridized samples.

Thermal gravimetric analysis (TGA) was carried out using specialized equipment (Model: TGA 2950, Manufacture: TA Instrument). The total temperature ranges for the analysis were from 40 to 900 °C under nitrogen (40-750 °C) and under air (750-900 °C) with a heating rate of 10 °C/min to investigate the LCF content.

Results and Discussion

Characterization. In order to confirm the content of LCF in the composite, TGA was used to measure the exact weight of LCF under the N_2 condition in the composite. Generally, to measure the weight of reinforcement in LFT, the weight of residue after burning under air is calculated by following the ASTM D5630 method. However, in the case of LCF, it is possible to oxidize under air at high temperature. The weight percent of LCF is carefully measured by using TGA. The result shows that LCF content is approximately 45 wt% from the residue. In order to confirm LCF in the residue, air was applied after 750 °C as shown in Figure 1.

Electrical Resistivity. Figure 2 shows the influence of LCF loading in terms of volume resistivity using LCF/LGF hybridized composite. The measuring direction of the volume resistivity is perpendicular to the resin flow. The volume resistivity of LCF0 was not measurable because of lack of a conductive intermediate. At 6 volume percent loading of LCF in a LCF/LGF hybridized composite, volume resistivity is approximately 90 Ωcm, but the resistivity dramatically decreases with increasing LCF loading at 10 volume percent. In order to obtain more precise information of percolation threshold, three samples were prepared which were 8, 9, and 10 vol% of CF. From the results, there is a percolation point between 8 and 9 volume percent loading of LCF.

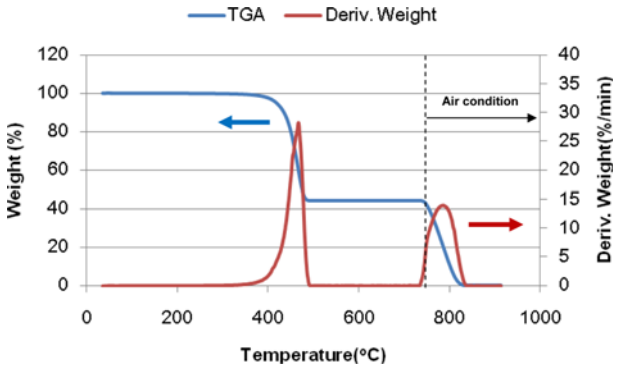

Figure 1. TGA curve and derivative weight loss curve of LCFT. Basically, TGA analysis was conducted under the N_2 condition below 750 °C and air was applied after the point to characterize CF.

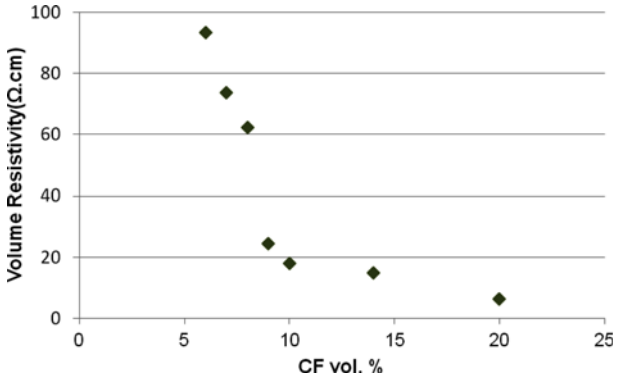

Figure 2. Volume resistivity as a function of LCF volume percentage for LCF/LGF hybrid samples. Volume resistivity of LCF0 sample is out of range.

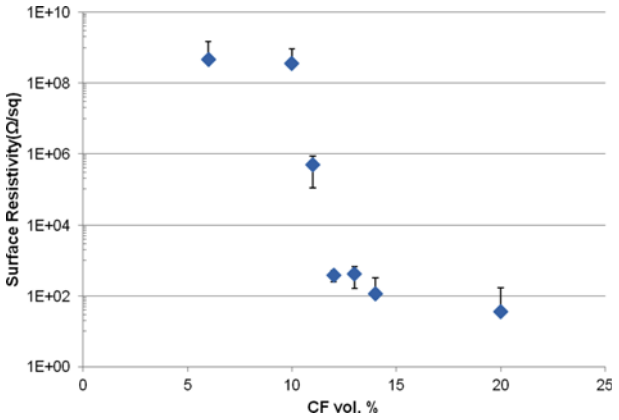

Figure 3. Surface resistivity of LCF/LGF hybridized samples as a function of LCF volume percentage. Surface resistivity of LCF0 sample is out of range.

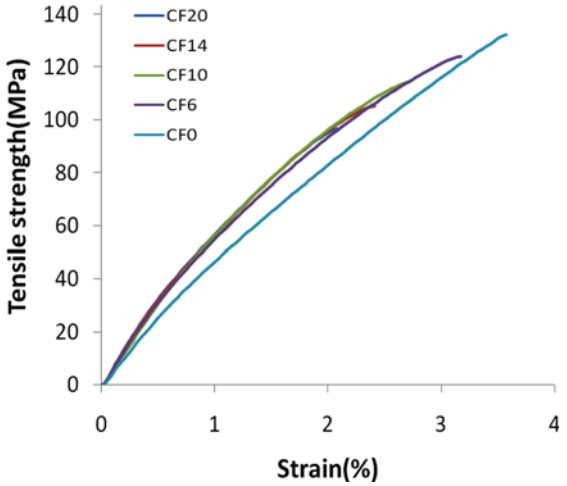

Figure 4. Tensile stress and strain curves of LCF/LGF hybridized composites.

In the case of short CF composite, the electrical percolation point is located between 6 and 9 volume percent of short CF.[16] In general, electrical conductivity is related to the percolation threshold, which is a function of the filler type and its content. The filler type indicates aspect ratio or original conductivity of the filler.[17] In comparison with our results, a similar range for percolation threshold has been shown. The aspect ratio value might affect the electrical conductivity, but it is not significantly changed in CF/GF hybridized composite because the long fiber can be broken during the injection molding process. The length differences between long fiber and short fiber after injection did not significantly affect the electrical properties. Further detailed study is in progress in terms of fiber direction, length, as well as compatibilizer content.

The electrical surface resistivity of LCF/LGF hybridized composite reinforced polypropylene as a function of LCF loading is shown in Figure 3. The surface resistivity for LCF0 was not measurable becuause of a lack of a conductive intermediate. From the results, there is a significant drop of surface resistivity between 10 and 14 volume percent of LCF loading in LCF/LGF hybridized composite and the precise percolation point is found by preparing three more samples which have 11, 12, and 13 vol% of CF. As a result, between 10 and 12 vol% of CF, the percolation threshold has been shown. According to Schutte *et al*., surface resistivity can be changed by injection pressure, mold temperature, and barrel temperature. These factors change CF length and concentration on the surface.[16] It might also be possible to increase surface resistivity in our system, but this result is evidence that there is a CF concentration gradient from the surface to the core. Additonally, O_2 plasma treatment was carried out to remove a polymer-rich surface to insure good contact between an electrode and sample surface.[18] Thus, the electrical conductivity on the surface which is a polymer-rich area can be lower than the core which is a CF-rich area.

Mechanical Properties. In general, the tensile stress-strain curve of polypropylene shows a ductile type of curve and strain at failure at around 500%.[13] However, the tensile stress-strain curves of LCF/LGF hybridized compostie exhibits brittle curves as shown in Figure 4. These hybridized composites occur first with linear deformation and then show non-linear deformation at high tensile stress. The failure of the composite follows several steps:[19]

1. Interfacial micro-failure occurs at the fiber end.
2. Micro-failure propagates along the fiber length direction.
3. Plastic deformation bands of the matrix occurs from the matrix.
4. Crack openings take places in the band and then the crack grows through the band.
5. Catastrophic crack propagation occurs through the matrix with a pulling out of fibers from the matrix.

The stress-strain curve, originally, shifts to the left meaning more brittle, but ultimate tensile strength is increased by increasing LCF volume faction because the strengh and modulus of CF are higher than GF. Tensile modulus of the composite containing LCF is higher than LCF0 composite, as expected, but the maximum tensile strength and strain of the composite decresases with increasing LCF volume fraction as shown in Figure 4. We discussed this issue in Fractography section.

In a hybrid combined with two homogeneus systems, the mechanical properties can be generelly predicted by the rule of hybrid mixtures (RoHM) equation:[15]

$$P_H = P_A V_A + P_B V_B \tag{2}$$

where, P_H is the predicted mechanical property, P_A and P_B is the property of the system A and B, respectively. V_A and V_B is the volume fraction of the system A and B, respectively.

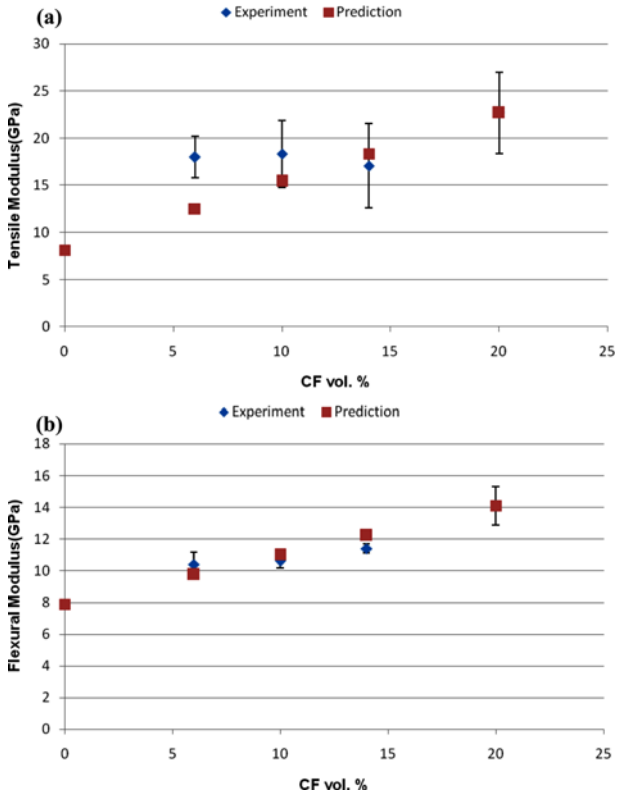

Figure 5. Tensile (a) and flexural (b) modulus results of LCF/LGF hybridized composites by increasing LCF volume percent. Blue diamonds indicate the experimental results and red rectangles indicate the predicted results using RoHM equation.

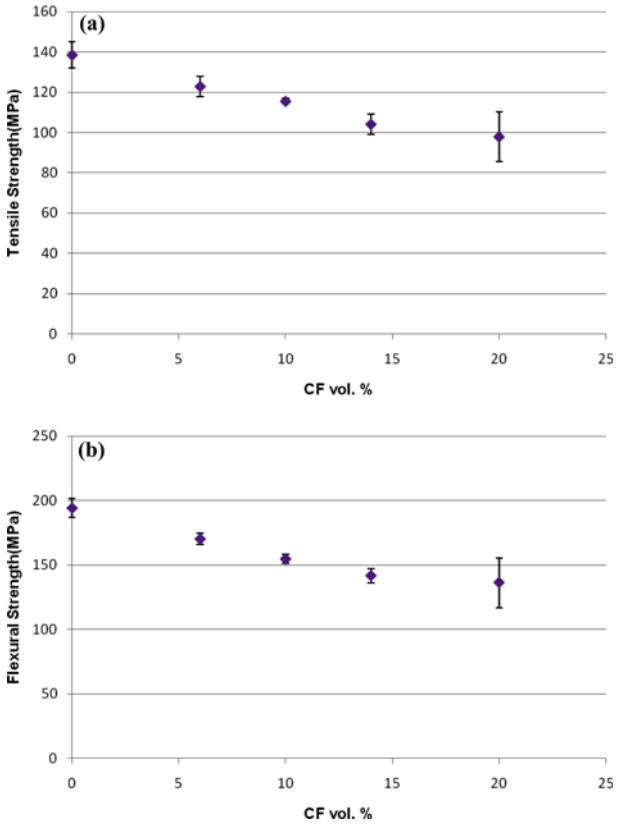

Figure 6. Tensile (a) and flexural (b) strength results of LCF/LGF hybridized composites by increasing LCF volume percent.

Figure 5 shows that the tensile and flexural modulus of the composite is increasing with increasing LCF volume fraction. Based on the results, the tensile and flexural modulus of LCF/LGF hybridized composite is enhanced by the addition of LCF and LGF. The maxium tensile and flexural modulus is approximately 22.7 and 14.1 GPa at 20 volume percent of LCF, which is similar to the tensile modulus of SCF composite which has 25 volume precent loading.[15] This means that the flexural modulus at the same volume fraction of LCF composite is higher than short CF composite.

The modulus of LCF/LGF hybridized composite was predicted using the RoHM equation and plotted in order to investigate the hybrid effect as shown in Figure 5. In the case of tensile modulus, the value is increasing with increasing LCF loading, but the experimental values are higher than the predicted values between 6 and 10 volume percent loading of LCF. This means that the LCF/LGF hybrid effect is observed at the points. In contrast, the flexural modulus of the hybridized composite was very close to the predicted value which indicates that there is no hybrid effect.

Figure 6 shows maximum tensile and flexural strength of LCF/LGF hybridized composite. It can be seen that the tensile and flexural strenght of LCF/LGF hybridized composite is improved by adding LCF and LGF in comparision with the strength of a polypropylene matrix. According to the RoHM equation, the maximum strength of a LCF/LGF hybridized compostie should increase with increasing LCF loading because of the tensile strength of CF.[15] However, based on these experimental results, the tensile and flexural strength of the composite is decreasing with increasing LCF loading. According to Nygard *et al*., bonding strength between LGF and polypropylene can be enhanced by surface treatment using silane compound as a sizing agent as well as PP-g-MAH as a competibilizer.[20] The LGF used in that experiment has a good bonding strength with the polypropylene matrix, whereas the LCF used in this experiment has poor interactioin with the matrix. Thus, the tensile and flexural strength are decreasing with increasing LCF loading even with high loading of LCF. In the short CF/GF hybridized composite, the maximum strength of the composite ranged from 50 to 58 MPa by increasing CF loading at constant 25 volume percent.[15] However, although there is no competibilizing effect for LCF and low volume loading of reinforcement, the maximum tensile and flexural strength of LCF/LGF hybridized composite were 100 and 140 MPa, respectively at maximum volume percent of LCF loading as shown in Figure 6(a). The value is approximately 2 to 3

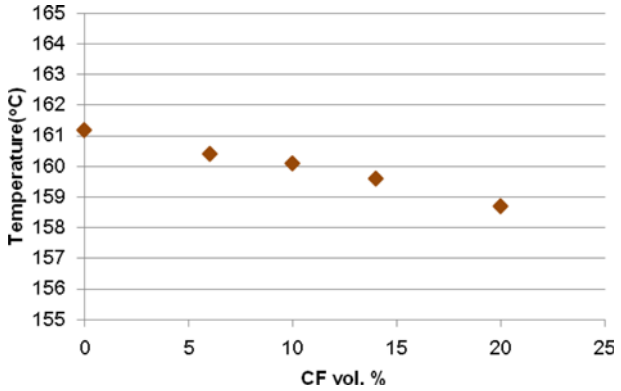

Figure 7. Heat deflection temperatures (HDT) of LCF/LGF hybridized composites at different volume factions.

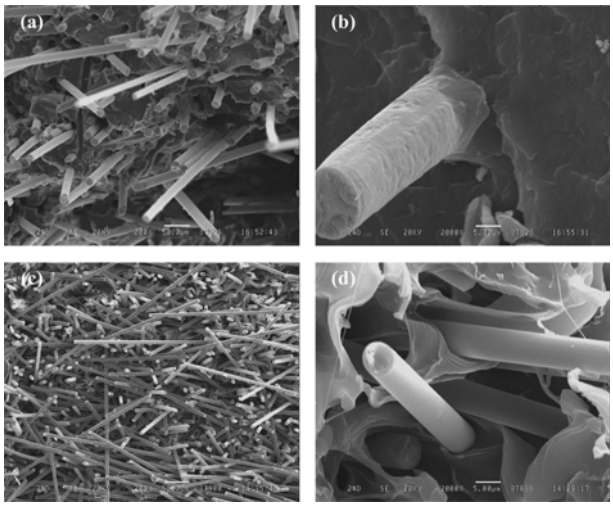

Figure 8. SEM images of the composites with 20 volume percent of LCF and LGF. (a) and (b) are LGF composites with a magnification of ×200 and ×2000. (c) and (d) are LCF composites with a magnification of ×200 and ×2000, respectively.

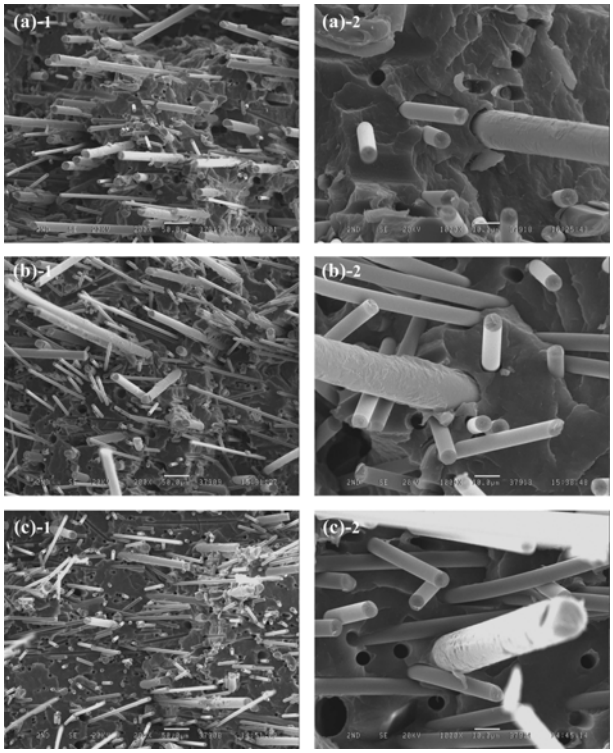

Figure 9. SEM images of LCF/LGF hybridized composites at a magnification of ×200(1) and ×1000(2); (a) LCF6, (b) LCF 10, and (c) LCF 14, respectively.

times higher than that of a short CF/GF hybridized composite. This indicates that fiber length effect is more significant than surface interaction in LCF/LGF hybridized composite.

Heat deflection temperature (HDT) is one essential property in a composite which is related to applications requiring thermal resistivity. Based on a HDT property, the composite can be evaluated as a material for high temperature applications. According to Ke *et al*., HDT is closely related to the motion of the matrix polymer chain.[21] As shown in Figure 7, HDT is decreasing with increasing LCF volume fraction. This indicates that the movement of a polypropylene molecular chain is increased with increasing LCF content. In general, surface area is inversely proportional to the diameter of fiber at the same volume. This indicates that a fiber which has a smaller diameter has a high possiblility for preventing the motion of a matrix chain. Since the diameter of CF is around 7 μm, HDT, generally, is increasing as LCF volume fraction is increasing. However, HDT is decreasing with increasing LCF volume fraction in our results. This is related to the surface interaction between LCF and the matrix polypropylene. Surface property will be discussed in the next session.

Fractography. Figure 8 shows the SEM micrographs of the fracture surface of LCF and LGF composite in terms of magnifications. At higher magnification, it is clear that LGF and PP matrix has a good interaction resulting from PP-*g*-MAH sizing. However, the LCF surface shows poor interaction between LCF and the matrix. This indicates that PP-*g*-MAH is able to react with a LGF sizing agent, but the LCF sizing agent is not matched to PP-*g*-MAH. Although the basic strength of CF is higher than GF, the tensile strength of the composite having the same volume fraction is lower than the LGF composite. However, in the case of modulus, it comes from the basic property of CF. As a result, it is increasing with increasing LCF volume fraction, even if LCF has poor interaction with the matrix.

Figure 9(a)-(c) show SEM images obtained from the fracture surfaces of the LCF/LGF hybridized composites. From high-magnification images, the diameter of CF and GF was measured as ~7 and ~17 μm, respectively. In addition, the figures show that LGF has a good interaction with the polypropylene matrix because of the rough surface of the LGF. However, the surface of LCF is smooth which means there is no interaction between the LCF surface and polypro-

pylene matrix as shown in all samples. The interaction between filler and matrix is a factor in changing mechanical properties.[22] According to Rong *et al*., the mechanical properties of a composite is enhanced if an inorganic particle is incorporated with the matrix, resulting from the surface modification of the inorganic particle.[23] Based on our experimental results, there is evidence of tensile and flexural strength decrease with increasing LCF volume percent. By decreasing LGF content, the interaction between fiber and matrix is decreasing so that the strength value is decreased even if the dispersion properties are good.

Figure 10 shows X-ray maps taken by the fracture surface of hybridized LCF/LGF composite using energy-dispersive X-ray spectroscopy (EDS) in order to investigate the distribution properties of LCF and LGF in polypropylene matrix. Although LCF surface doesn't have any interaction with the matrix, LCF is well dispersed in the matrix as shown in Figure 10(a-3), (b-3), and (c-3). In the case of nanoparticles, physical mixing is not enough to combine two or more different materials. However, it is possible to distribute a micro-reinforcement well in a matrix using physical mixing such as an extrusion, injection, and dry blending. The composition of Si and C is imaged in the second and third column in Figure 10. According to X-ray map images, it is clear that

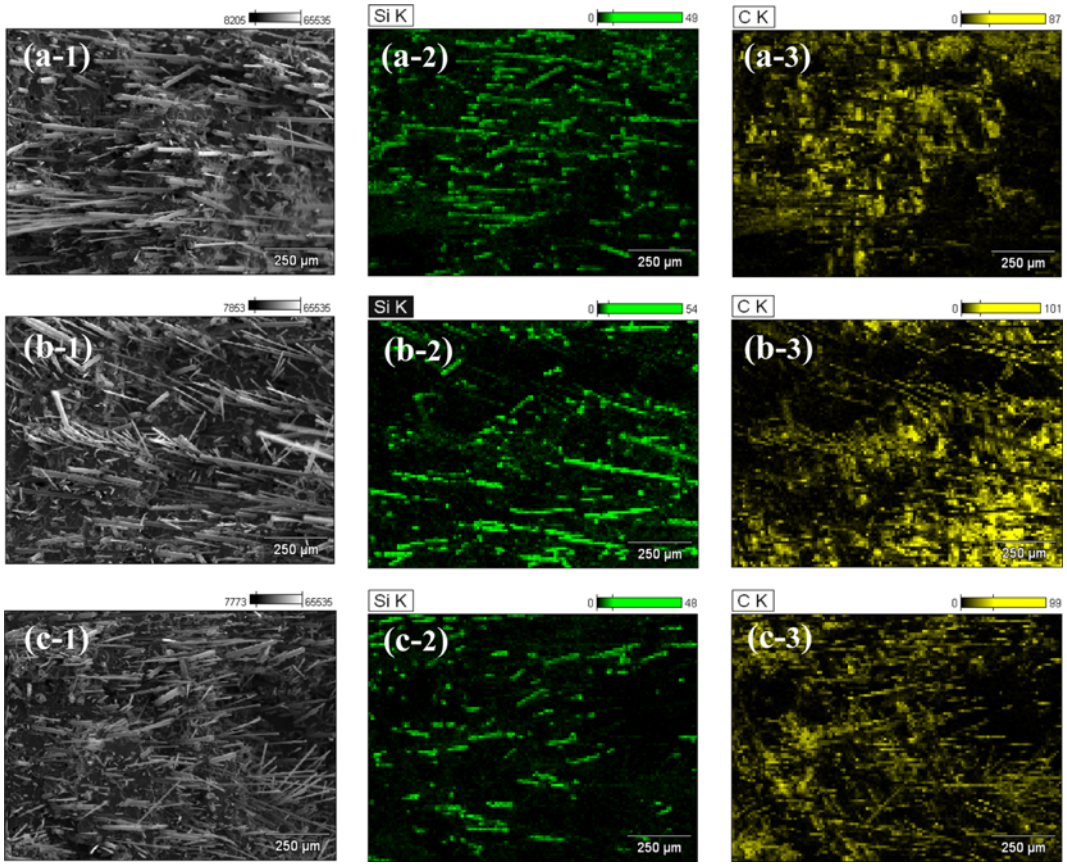

Figure 10. X-Ray maps of the hybridized LCF/LGF composites: a, b, and c indicate LCF6, LCF10, LCF14, respectively. 1 indicates topographical image. 2 and 3 indicate Si and C concentration images.

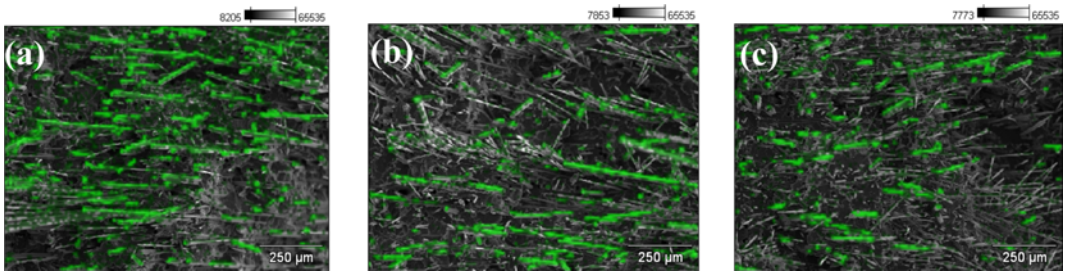

Figure 11. Overlapped X-ray map which is Si concentration image on topographical image: (a), (b), and (c) indicate LCF6, LCF10, LCF14, respectively.

LCF and LGF are dispersed well and the concentration of Si (green color in the map) is decreasing with increasing LCF volume in a composite matrix. In comparison with a topological image, the X-ray map of Si was overlapped with the SEM image as shown in Figure 11.

Conclusions

The electrical and mechanical properties of LCF/LGF hybridized composites have been investigated in order to understand electrical percolation and the hybrid effect for industrial application in automobiles, aerospace, and home appliances. The volume resistivity of the electrical property results show that the percolation threshold point is between 6 and 10 volume percent of LCF loading in LCF/LGF hybridized composite. The surface resistivity data shows that the significant drop appears between 10 and 14 volume percent of LCF loading. The results of tensile and flexural modulus of mechanical property have been studied as a function of LCF loading. In comparison with RoHM equation, the hybrid effect has been observed in tensile modulus results around 6 and 10 volume percent of LCF loading whereas the result of flexural modulus is very close to the predicted result. This means that physical percolation threshold can be related to not only electrical properties but also mechanical modulus. However, the tensile and flexural strength of the composite is decreasing with increasing LCF loading because of the poor interaction between LCF and polypropylene matrix. The interaction study between LCF/LGF and matrix polypropylene has been investigated by SEM and HDT test. As a result, the interaction between fiber and polypropylene matrix can affect mechanical strength more significantly than mechanical modulus.

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