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Evaluation of Electrical Properties of Planar Supercapacitor with Substrate Made of Cellulose

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Abstract

The object of this study was to evaluate the electrical properties of the planar supercapacitor, which added polyimide powders into cellulose film as base materials. The pattern for the electrical circuit was formed on the prepared cellulose film by irradiating a blue semiconductor laser to fabricate the supercapacitor, where the cellulose film was modified with polyimide powders. Electrical resistance and specific capacitance of a planar supercapacitor were measured to evaluate the effect of the addition of polyimide powders on the addition rate of polyimide powders. Test results showed that the electrical resistance decreased with an increase in the addition rate of polyimide powders and then the specific capacitance was increased with an increase of the addition rate of polyimide powders. This study suggested that for obtaining high capacitance and low electrical resistance, the most suitable conditions of fabrication of a supercapacitor were 1wt% of the addition rate of polyimide powders and 400 mW of laser power.

Keywords

Cellulose, Polyimide, Planar Super Capacitor, Electrical Resistance, Specific Capacitance

1. Introduction

Flexible printed circuits (FPC) were focused as an electrical circuit board for wearable devices and small electronic equipment [1] [2]. Recently, the subtractive method [3], removing copper around circuit parts due to the etching process with photo mask, and the semi-active method [4], forming circuits due to copper plating on the film, have been employed for the fabrication method of FPC. These methods have a high environmental burden due to the etching process and copper plating, which require about the new fabrication methods of FPC that have low environment burden. Previous studies proposed the pretend-electro technics method [5], forming the circuits on the base materials by printing technology using the ink with silver powders, and the forming methods of circuits that need no auxiliary materials due to selective carbonization of the wood by laser [6]. However, the problem in these methods was that the auxiliary materials are expensive, and the electrical resistance of circuits is high.

In this study, cellulose film, which was modified with additional materials, was focused as the base material of FPC. Modified cellulose film was prepared by adding polyimide powders into cellulose, in addition, circuits pattern was formed by irradiating cellulose film with blue semiconductor laser. The object of this study was to evaluate the electrical properties of the planar supercapacitor which added polyimide powders into cellulose film as base materials.

2. Experimental Methods

2.1. Materials

Bacterial cellulose was prepared as the base material for the capacitors. Polyimide powders (P84@NT1-HCM superfine, Polyplastics-Evonik corp.) were also added to the matrix as shown in **Figure 1**.

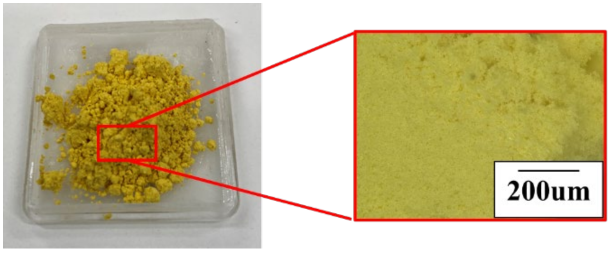


Figure 1. Polyimide powders.

2.2. Fabrication Methods of Cellulose Film Added with Polyimide Powders

Figure 2 shows the fabrication method of cellulose film. Cellulose suspension was

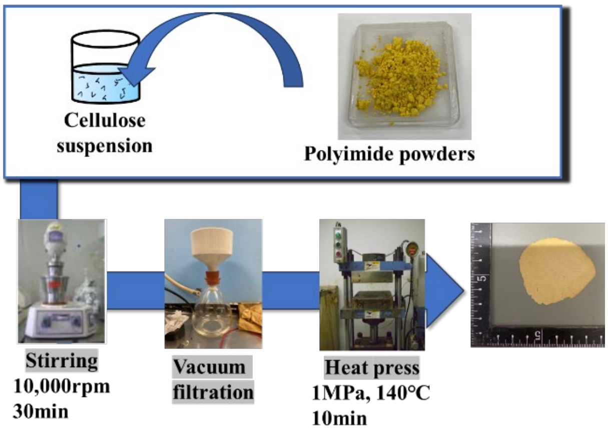


Figure 2. Fabrication methods of cellulose film.

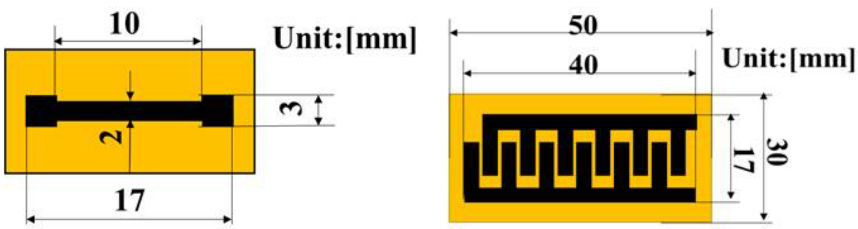
prepared by dispersing the mixtures with polyimide powders, bacterial cellulose and purified water at 10,000 rpm for 30 min by mechanically stirring machine. Cellulose film was fractured by vacuum filtering the suspension and preheating at 140 deg-C under 1 MPa for 10 min by the heat pressing machine. The addition rate of polyimide powders was approximately 0.3wt%, 0.5wt% and 1wt%.

2.3. Drawing Method of Carbonized Pattern

Carbonized patterns were drawn on cellulose film specimen by irradiating with blue-semiconductor laser (ATOMSTSCK, A510W). Table 1 shows the test conditions of the laser. Shapes of carbonized pattern were employed with a dumbbell shape for electrical resistance measurement, with comb shape for specific capacitance measurement, respectively, shown in Figure 3.

Table 1. Irradiating conditions of a blue semiconductor laser.

Laser power [mW]	200, 300, 400
Scanning rate [mm/min]	600
Wavelength [nm]	405



(a) Measurement of electrical resistance. (b) Measurement of specific capacitance.

Figure 3. Pattern shapes for measurement of electrical resistance and specific capacitance.

2.4. Fabrication Method of Planar Supercapacitors

Figure 4 shows the fabrication method of the capacitors. Collecting electrode was put on the prepared electrode pattern, and then a copper tape as tab electrode was also put on the pattern. Next, phosphoric acid/PVA electrolyte was dropped for about 1 mL/min onto the electrode and solidified in a desiccator by drying at room temperature for 12 hours. Solid electrolytes had about 0.2 mm of average thickness. The phosphoric acid/PVA electrolyte was prepared by adding 1 g of PVA and 1.3 mL of phosphoric acid to 18.7 mL of purified water and then stirring at 95˚C for 15 minutes. Finally, a copper tape was placed on the solidified electrolyte as a tab electrode for the reference electrode.

2.5. Measurement for Electrical Resistance of Circuit Pattern

The electrical resistance of the pattern was measured by 4-terminal method with digital multimeter (R6871E, Advantest). Volume resistivity ρ was calculated with the obtained electrical resistance based on Equation (1). At least 3 specimens were tested.

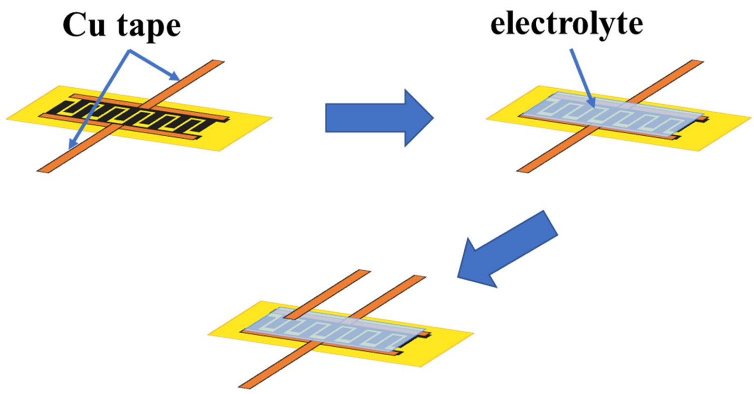


Figure 4. Fabrication method of supercapacitor.

$$\rho = \frac{A_c}{l} \times R \tag{1}$$

Here, A_c, l and R were cross section area of carbonized part, gage length and electrical resistance between gage lengths, respectively.

2.6. Measurement for Specific Capacitance of Planar Supercapacitors

Specific capacitance of the planar supercapacitor was measured by cyclic voltammetry test. Response current of the specimen was measured by applying voltage to the specimen. Test conditions of cyclic voltammetry test were shown in Table 2. Specific capacitance C_s was calculated from obtained current-voltage curve based on Equation (2).

$$C_s = \frac{\int IdV}{\Delta V \cdot \upsilon}$$

Here, I, V, ΔV and v were response current, voltage, window and scan rate, respectively.

Table 2. Irradiating conditions of blue semiconductor laser.

Sweeping volage range [mV]	0~1000
Scan rate [mV/min]	600

2.7. X-Ray Diffraction Analysis of Carbonized Particles

Carbonized parts by irradiating with blue semiconductor laser were analyzed due to x-ray diffraction. This analysis was investigated with X-ray diffraction instrument (SmartLab) which tube voltage and tube current were 40 kV and 40 mA, respectively.

2.8. Raman Spectroscopy Analysis of Carbonized Particles

Raman spectrum of carbonized parts was investigated with the Raman spectrometer (Anton Paar, Cora: Compact Raman Analyzer). The wavelength and power of laser were 532 nm and 50 mW, respectively.

3. Results and Discussions

3.1. Electrical Resistance of Circuit Pattern

Figure 5 shows the relationship between the electrical resistance of the specimen and the addition rate of polyimide powders. Results of specimens that added no polyimide powders were not shown in this figure because it was not conductive. Electrical resistance of specimens decreased with an increase of the addition rate of polyimide powders and decreased with an increase of laser power.

Figure 6 shows one of the cross-sectional observation results in the gage length

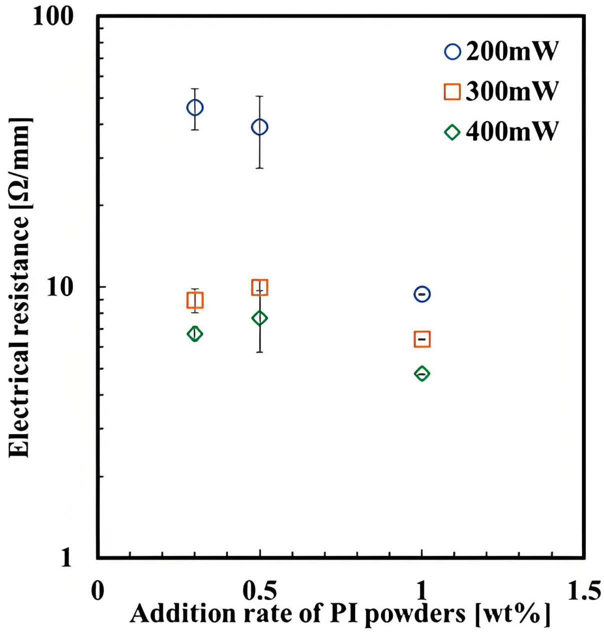


Figure 5. Electrical resistance with respect to the addition rate of polyimide powders.

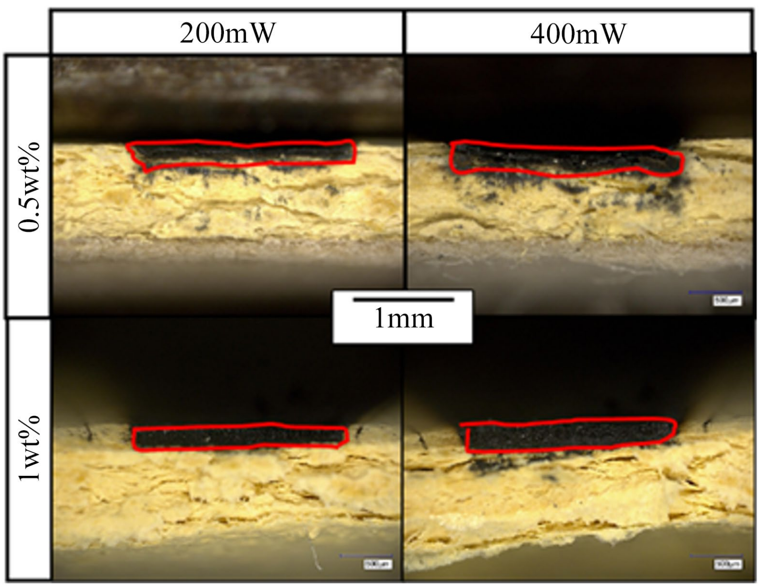


Figure 6. Cross-section images of carbonized parts.

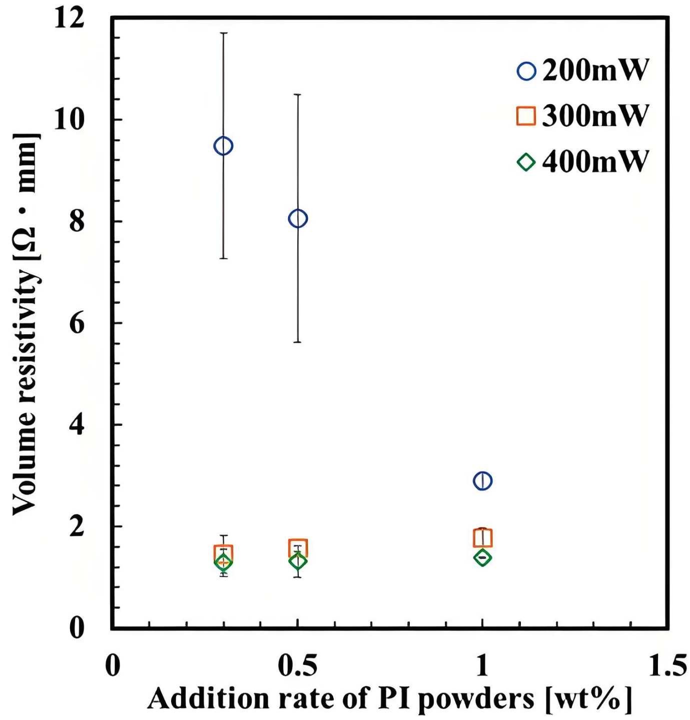


Figure 7. Volume resistivity with respect to addition rate of polyimide powders.

section. Blackened areas by laser irradiation were defined as a cross-section area of carbonized part, and the areas of carbonized part A_c was calculated using trapezoidal approximation and image analysis. The average thickness of cross section area was about 0.3 mm and slightly increased with an increase in laser power. Figure 7 shows the relationship between volume resistivity of the carbonized part and the addition rate of polyimide powders. In condition of 200 mW of laser power, the volume resistivity was decreased with an increase in the addition rate of polyimide powders. In contrast, the volume resistivity was almost the same regardless of the addition rate of polyimide powders. Therefore, it found that about 1wt% of the addition rate of polyimide powders and 300 mW of laser power that the stable conditions to obtain circuit pattern that the volume resistivity was low.

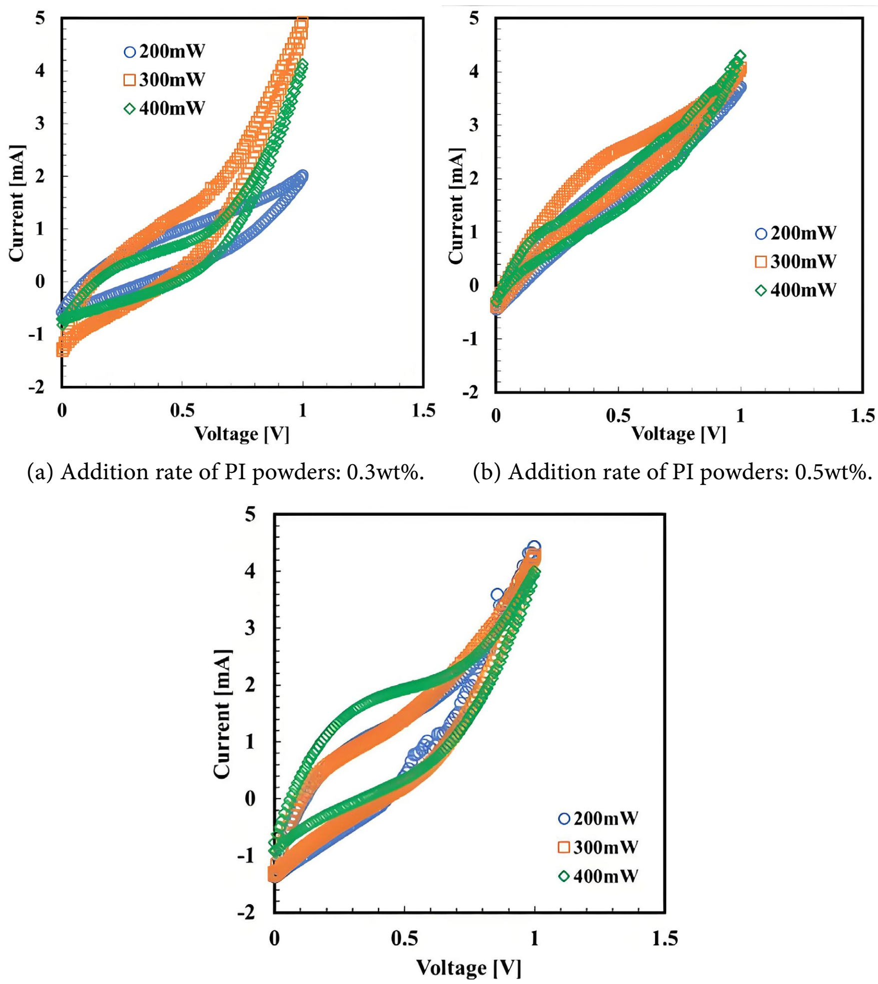
3.2. Current-Voltage Curves of Planar Supercapacitor

Figure 8 shows the current-voltage curves of the specimen obtained by cyclic voltammetry test. When the addition rate of polyimide powders was 0.3wt%, the area surrounded with the current-voltage curves was little difference regardless of the laser power. When the addition rate of polyimide powders was 0.5wt%, the maximum response current increased with an increase in laser power. When the addition rate of polyimide powders was 1wt%, in any conditions of the laser power, the maximum value of responded current was high, and the gradient of the curves during applying the voltage increased with an increase in the laser power.

3.3. Specific Capacitance of Planar Supercapacitor

Figure 9 shows the relationship between specific capacitance of the specimen and laser power. Specific capacitance shown in figure was calculated to divide total capacitance into the area of pattern. When the addition rate of polyimide powders was 0.3wt%, specific capacitance was no significant difference regardless of laser power. When the addition rate of polyimide powders was 0.5wt%, specific capacitance was slightly improved. When the addition rate of polyimide powders was 1wt%, specific capacitance was significantly improved by irradiating with 400 mW of laser power compared with other laser power conditions.

In comparison with the results of previous studies [7]-[9], the electrical performance of super capacitors made of cellulose showed little difference compared to that of a supercapacitor which irradiate laser with flexible polyimide film. In other words, this study found that supercapacitors made of cellulose have not only low environmental load, but also almost the same performance as conventional supercapacitors.



(a) Addition rate of PI powders: 0.3wt%. (b) Addition rate of PI powders: 0.5wt%.

(c) Addition rate of PI powders: 1wt%.

Figure 8. Current-voltage curves of specimens.

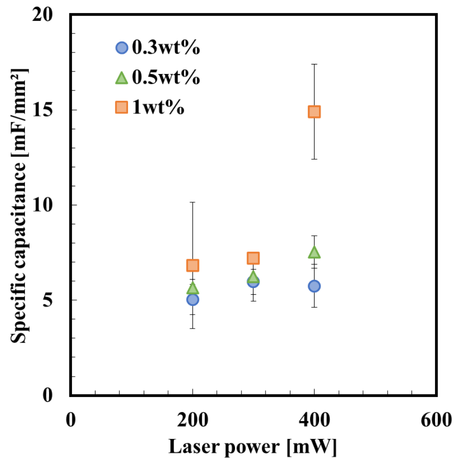


Figure 9. Specific capacitance of supercapacitor with respect to laser power.

3.4. X-Ray Structural Analysis Results of Carbonized Part

Figure 10 shows the relationship between the diffraction intensity and diffraction angle of the specimens that add 0.3wt% of the polyimide powders and irradiate the laser at 200 or 400 mW. In any condition, the significant peak was confirmed to be around 24 deg and 78 deg of the diffraction angle. In addition, the height of peaks increased with an increase in laser power. The peaks around 24 deg and 78 deg of the diffraction angle were C002 peak and C110peak, respectively, which implied that carbon crystal structure with high regularity was generated on cellulose film. Previous studies [10] [11] also reported that the laser induced graphene was generated by the irradiation of laser on polyimide film, therefore this study was

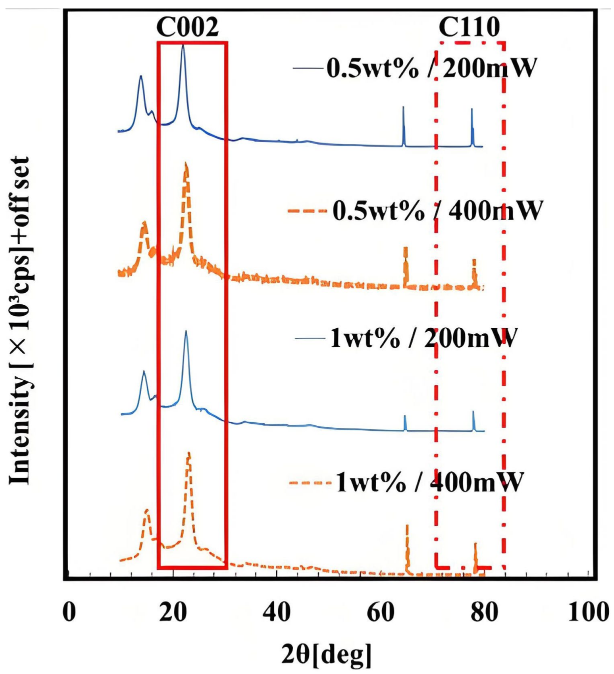
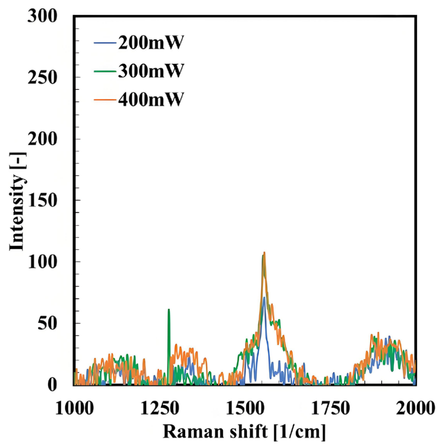


Figure 10. X-ray intensity with respect to diffraction angle.

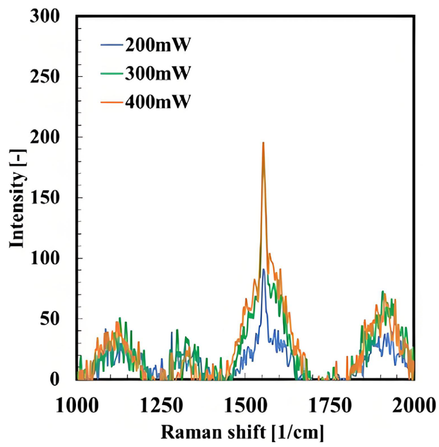
also suggested that the laser induced graphene was also generated. These results showed that the laser induced graphene was generated on the film by adding the polyimide powders and carbonizing, which prevented the formation of amorphous carbon and stabilized the formation of the crystal structure of carbonized parts.

3.5. Raman Spectroscopy

Figure 11 shows the relationship between Raman shift and intensity. When the polyimide powders were added 0.5 wt% and 1 wt%, the peaks of Raman shift were confirmed to be around 1580 cm^{-1} and 1350 cm^{-1}. The peak around 1580 cm^{-1} was also increased significantly with an increase in the laser power. These results show that the peak around 1580 cm^{-1} and 1350 cm^{-1} were G-band, based on the graphite structure, and D-band, based on the disorder and defects in the graphene structure, respectively. In addition, the G-band peak with adding 1wt% of polyimide powders was significantly improved compared with that with adding 0.5wt% of polyimide powders.



(a) Addition rate of PI powders: 0.5wt%.



(b) Addition rate of PI powders: 1wt%.

Figure 11. Relationship between intensity and Raman shift.

Therefore, this study found that more graphene was generated by adding 1wt% of polyimide powders and irradiating with 400 mW, which obtained low and high volume resistivity and specific capacitance, respectively.

4. Conclusions

In this study, to investigate the effects of the addition of polyimide powders on the electrical properties of planar supercapacitor made of cellulose, planar supercapacitor was prepared by irradiating blue semiconductor laser on cellulose specimen and carbonizing it, and the electrical resistance and specific capacitance of the planar supercapacitor were measured. The following conclusions were obtained:

1) When the laser power was 200 mW, the volume resistivity of carbonized parts decreased with an increase in the addition rate of polyimide powders. In

contrast, when the laser power was 300 or 400 mW, the volume resistivity was little difference regardless of the addition rate of polyimide powders.

2) When the addition rate of polyimide powders was 0.3wt%, the specific capacitance of planar supercapacitor was no significant difference regardless of the laser power. In contrast, when the addition rate of polyimide powders was 0.5wt%, the specific capacitance was slightly increased with an increase in the laser power. When the addition rate of polyimide powders was 1wt%, the specific capacitance of planar supercapacitor which carbonized at 200 or 300 mW was almost the same, however, that which carbonized at 400 mW was significantly increased compared with other conditions.

3) In this study, the most suitable conditions obtained low electrical resistance and high specific capacitance were 1wt% of the addition rate of polyimide powders and 400 mW of the laser power.

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Conflicts of Interest

The author declares no conflicts of interest regarding the publication of this paper.

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