Preparation and properties of activated carbon/carbon composites with coal tar pitch-based COPNA resin

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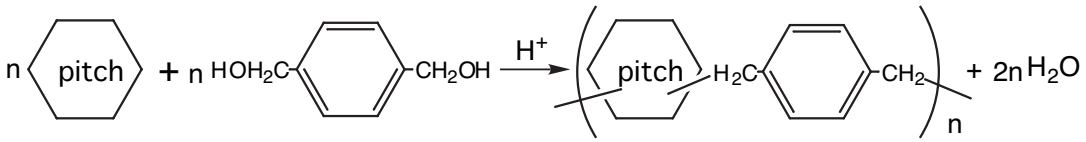
Introduction

Activated carbons are strongly indispensable and interesting for environmental pollution control materials, electrode materials, and other application materials. In recent years, porous carbon materials with high specific surface area have attracted considerable attention as electrode materials of electrochemical double layer capacitors (EDLC) with high energy density. Activated carbons are available in terms of typical porous carbon materials for EDLCs[1].
Electric resistance between activated carbon particles or between activated carbon particles and current collectors would, however, increase by the use of the particles. The formation of the thin firm molding bonded to current collector will contribute to practical application such as rechargeable electrical power sources in cellular phone and portable personal computer.
On the other hand, we have reported that coal tar pitch based thermosetting resin, named as COPNA resin (Scheme 1), was suitable for the preparation of C/C composite materials [2] without the pretreatment process such as removal process of sizing agent on carbon fiber.



Scheme 1

The present report considers the preparation and properties of activated carbon/carbon composites with coal tar pitch-based COPNA resin.

Experimental

Materials. Coal tar pitch (PK-QL, Kawasaki Steel Corp., Softening point: 85℃, Free Carbon: 54%, Ash<0.01%, Toluene Insoluble Component: 17%, Quinoline Insoluble Component <0.01%) was ground to powder under 100 meshes just before its use. Other reagents, activated carbon powder, 1,4-benzene dimethanol, 1-methylnaphthalene, and p-toluene sulfonic acid were used without further purification.

Instrumentation. Fourier transform infrared (FT-IR) spectra were obtained by the KBr disk technique on a Shimazu spectrometer. Curing temperature of the coal tar pitch-based thermosetting resin was estimated by thermal analysis with a SII TG/DTA 6200 (Seiko Instrument Inc.). The surface and cross section of the sample were observed with a scanning electron microscope (SEM, JSM-5600LV, JEOL Co.). The X-ray powder diffraction measurements were taken on a Rigaku RINT-2100.

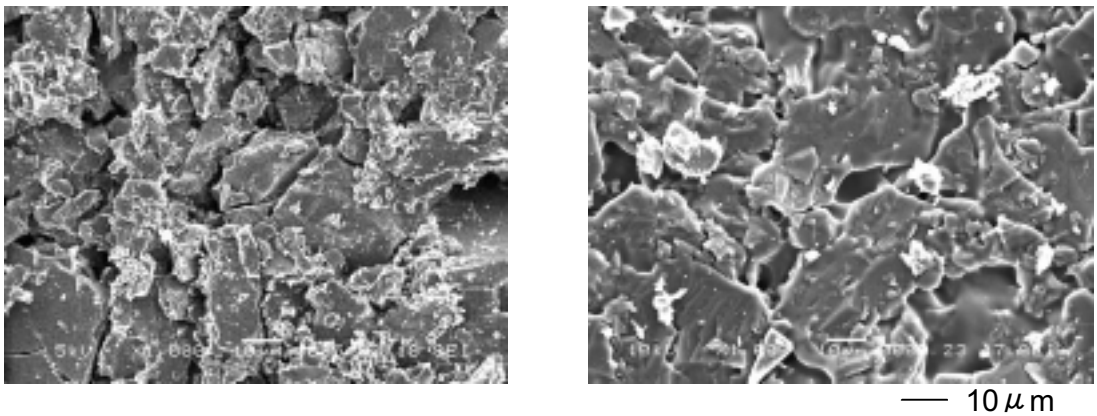
Preparation of activated carbon/carbon composites with coal tar pitch-based COPNA resin. Coal tar-pitch based COPNA resin was prepared to heat the mixture of coal tar pitch, 1-methylnaphthalene, p-xylylene glycol and acid catalyst at 130 ℃.

Activated carbon powder was mixed with coal tar pitch-based COPNA resin. The mixtures were pressed with the preheated mold at 180 ℃ for 1 h and then at 200 ℃ for 1 h by compression molding method to 450 kgf/cm^2. The moldings were heated at 1000 ℃ for 1 h in inert atmosphere. Graphite-based current collectors were mounted in the heat-treated molding (AC/COPNA) with paste prepared from coal tar pitch-based COPNA resin. The assembled samples were heated at 1000 ℃ for 1.

Results and Discussion

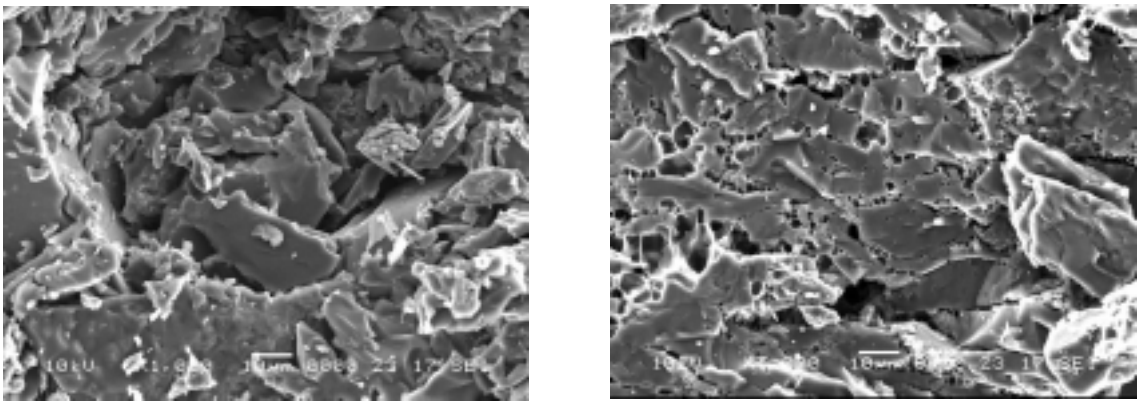
The moldings heated at 1000 ℃ were well firm and no problems occurred during the mounting process of current collectors on the moldings. The uneven holes (Fig.1 a) with various sizes were observed on surface of the carbonized moldings and the small spaces in the cross section (Fig.2 a) were observed. These holes and spaces will be expected to be effective for adsorption of electrolyte solution. In this case, the sample adsorbed 30wt% of electrolyte solution and showed the charge-discharge behavior as electric double layer capacitor. The moldings similar to those prepared from coal tar pitch-based COPNA resin were also made from phenolic resin and activated carbon powders as a control experiment. SEM photographs of phenolic resin based samples that the surface and cross section of the carbonized moldings (Fig. 1 b and 2b) were covered with thin deposit like pyrolytic carbon.

Specific surface area was estimated to be 1593 m^2/g for AC/COPNA. The size of capacitor is ca. 30mm like that of 500-yen coin as shown in Fig. 3.



(a) AC/COPNA (b) AC/phenolic resin

Figure 1 Surface of the carbonized moldings.



(a) AC/COPNA (b) AC/phenolic resin

Figure 2 Cross section of the carbonized moldings.

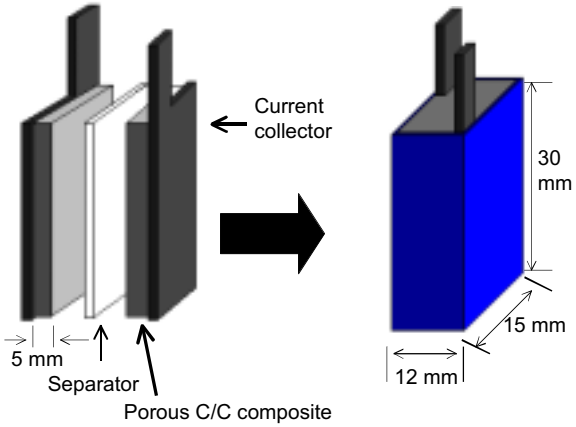


Figure 3. Assembling of a capacitor.

Capacitance values of 30 F/g at a constant current density of 10 mA/g at 25 ℃ was obtained by charge-discharge measurement in $LiClO_4$-propylene carbonate electrolyte. The effect of large specific surface area on capacitance value was not so large because micropore characteristic of activated carbon would be dead space in the charge-discharge measurement.

Conclusions

The molding prepared from activated carbon/carbon composites with coal tar pitch-based COPNA resin was well firm and small-sized assembling of a capacitor was successful. The surface area effective to capacitance measurement seems not to be so large because of dead space resulted in micropore characteristic of activated carbon.

References

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[2] Ota M., Aoyagi Y., Miyazaki S., Shibasaki M., 2000 International Chemical Congress, Hawaii, INOR 970(2000).