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Low-cost Activated Carbon Materials Produced from Used Coffee Grounds for Electric Double-layer Capacitors

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Abstract— Activated carbon materials were produced from used coffee grounds by KOH and CO₂ activation. Physical properties including pore-size distribution, specific surface area using Brunauer-Emmett-Teller theoretical calculations (S_{BET}), micro pore surface area (S_{micro}), mesopore surface area (S_{meso}), micro pore volume (V_{micro}) and mesopore volume (V_{meso}) of the prepared coffee ground carbon (CGC) materials were examined. A maximum specific surface area (S_{BET}) value of 1971 m²·g⁻¹ was observed for the prepared CGCs. Polarized electrodes were fabricated using CGC materials in order to examine the physical and electrochemical influence of CGCs on the characteristics of electric double-layer capacitors (EDLCs). Electrochemical measurements were carried out in 0.8 M (C₂H₅)₄NBF₄/PC and 0.5 M H₂SO₄ electrolytic media for characterization of EDLCs using cyclic voltammetry (CV) and electrochemical impedance spectroscopy. Specific capacitance values as high as 204 F·g⁻¹ were observed in aqueous electrolyte and 127 F·g⁻¹ in organic electrolyte. The cost-performance ratio was also calculated considering all manufacturing costs, transportation cost and maintenance cost. As the result, the cost-performance of EDLCs electrodes was calculated as 0.444 Yen (JPY) ·F⁻¹ in the case of using organic electrolyte. This study highlights the potential of high-performance and low-cost EDLCs by applying CGCs derived from abundant and inexpensive used coffee grounds.

Index Terms— activated carbon, battery, electric double-layer capacitor, coffee ground.

I. INTRODUCTION

In recent years, the study of electric double-layer capacitors (EDLCs)[1] as energy storage devices in place of chemical batteries, or to provide additional power or life cycles in battery systems, has become a notable area of research on a global scale. EDLCs utilize the double layer formed at the interface between the polarized electrode and the electrolyte solution. The polarized electrode is prepared by mixing an activated carbon, a conducting material and a binder. Various types of activated carbon materials, for example, coconut shell [2], activated carbon fiber[3], and carbon nanotubes[4] were used in EDLC. The activated carbon typically occupies 80 wt% of the polarized electrode, and the cost of activated carbon represents 30 % of the total cost of EDLCs.[5] Therefore, the cost of EDLC fabrication can be significantly reduced through the use a low-cost activated carbon source. A significant drawback of using EDLCs is their low energy density compared to batteries. This property can be improved by increasing the withstand voltage of the electrolyte and increasing the surface area of the polarized electrodes. Withstand voltage of electrolytes are decided by solvent, for examples, 1.0 V in the case of using aqueous electrolyte and 2.5-3.0 V in the case of using non-aqueous electrolyte. Given these limitations it is very important to choose a carbon material, which can absorb large amounts of charge if energy density is to be high. In recent years, the use of coffee husks and coffee endocarp - an abundant source of activated carbon - has generated significant global research interest in terms of materials chemistry applications.[6-8] In Japan 100,000 tons per year of used coffee grounds are exhausted. Some beverage companies tried to use coffee grounds as fertilizers. However, the amount of coffee ground importation and consumption gradually increase year by year. It is necessary to further develop effective utilization of used coffee grounds. Many studies reported the characterization of activated carbons from coffee grounds and application to EDLCs. Baquero et al. reported activated carbons were derived from coffee bean husks in presence of H₃PO₄ and the surface areas were 1402 m² ·g⁻¹. [9] Rufford et al. reported activated carbons were produced from used coffee grounds by treatment with ZnCl₂ and the surface areas were about 1020 m² ·g⁻¹. [10-11] Takahara et al. reported KOH activated carbons were obtained from waste coffee beans and they clarified relationships between weight ratio of KOH to carbon material and specific surface area. [12] Kikuchi et al. reported KOH activated carbons were obtained from coffee grounds and they clarified relationships between concentration of KOH and specific surface area. They obtained high



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specific surface area of $2549 \text{ m}^2 \cdot \text{g}^{-1}$ when the concentration of KOH was 4.5 M.[13] In the present study, relatively high surface area ($1971 \text{ m}^2 \cdot \text{g}^{-1}$) activated carbon was derived from used coffee grounds with high concentration of KOH (8 M) and low weight ratio of KOH to carbon material (0.5:1). Furthermore, the cost-performance of EDLCs using activated carbons derived from used coffee grounds was calculated.

II. EXPERIMENTAL

A. Preparation and characterization of activated carbon (CGCs)

Used coffee grounds were provided by a Japanese beverage company. Original coffee grounds were not crushed and grain diameter was about 1–3 mm. Used coffee grounds were dried at $150 \text{ }^\circ\text{C}$ for 1 h. After drying, the grounds were carbonized in a tube furnace (TMF-500N, As One Co., Japan) under N_2 gas flow at $600 \text{ }^\circ\text{C}$ for 1 h at a heating rate of $5 \text{ }^\circ\text{C} \cdot \text{min}^{-1}$. The KOH activating process required soaking of the carbonized coffee grounds in 8 M KOH over 24 h. The weight ratio of KOH/carbonized coffee grounds was 0.5/1.0. It was the problem that corrosion of tube furnace by using KOH because KOH is alkaline chemicals. In this experiment, maintenance cost by corrosion could be saved using low amount of KOH. After drying, the materials were activated in a furnace under N_2 gas flow at $900\text{--}950 \text{ }^\circ\text{C}$ for 1–3 h and $1000 \text{ }^\circ\text{C}$ for 2 h. For CO_2 activation, the materials were activated in a furnace under CO_2 gas flow at $1000 \text{ }^\circ\text{C}$ for 1–3 h. Ten CGCs were prepared in this manner by varying the activation method and activating temperatures and times used. Table 1 shows activating conditions employed for the prepared CGCs. The CGCs were further purified by washing in 0.6 M aqueous HCl to remove impurities, followed by further rinsing with distilled water until pH of the wastewater reached 7. The resultant materials were dried further at $150 \text{ }^\circ\text{C}$ for 1 h. The morphologies of CGCs were analyzed by scanning electron microscope (SEM; S-5500, Hitachi High-Technologies Co., Japan) and transmission electron microscopy (TEM; JEM-2010M, JEOL Ltd., Japan). Small particles observed by microscopy were analyzed using particle size analysis (FPAR-1000, Otsuka Electronics Co. Ltd., Japan). X-ray photoelectron spectroscopy (XPS; S-3300, Shimadzu, Japan) was performed to determine the types of oxygen-containing functional groups. The XPS spectra were recorded using Mg $K\alpha$ radiation through background processing. The analysis of the spectrum was performed using a nonlinear least-squares fitting program with a symmetric Gaussian function. The surface composition of the samples was estimated based on C-C, C-O, C=O and O-C=O bond peaks and the appropriate sensitivity factors.[14] The prepared CGCs were dried at $150 \text{ }^\circ\text{C}$ over 12 h under high vacuum (10 Pa). Surface area and pore-size distributions of CGCs were analyzed by Brunauer-Emmett-Teller (BET)[15], t-plot[16-18] and Barrett-Joyner-Halenda (BJH)[19] analysis methods using an automatic specific surface area/pore size distribution measuring equipment (Tristar 3000, Shimadzu, Japan).

B. Synthesis and electrochemical characterizations of CGCs-mixed polarized electrodes

Polarized electrodes were fabricated for electrochemical characterization experiments. Prepared CGCs, PTFE (polytetrafluoroethylene) as binder and Ketjenblack (EC-600JD; Lion Co. Ltd., Japan) as conducting materials were mixed in a mass ratio of 8.5:0.5:1.0. Mixed samples were placed in a mold and polarized electrodes were fabricated using a heat press machine (5 MPa) into a diameter of 10 mm and thickness of $550 \text{ }\mu\text{m}$. Capacitances of the prepared polarized electrodes were measured by an electrochemical measurement system (HZ-5000, Hokuto Denko Co. Ltd., Japan). For evaluation of the organic electrolyte medium, an electrolyte solution was prepared by dissolution of 0.8 M tetraethylammonium tetrafluoroborate ($(\text{C}_2\text{H}_5)_4\text{NBF}_4$) in propylene carbonate (PC) obtained from Tomiyama Pure Chemical Industries Ltd., Japan. A 200 mm^2 Pt counter electrode and an Ag/Ag⁺ reference electrode were used in the cyclic voltammetry (CV) measurement. For evaluation of aqueous electrolyte medium, an electrolyte solution (0.5 M H_2SO_4) was obtained from Wako Pure Chemical Industries Ltd., Japan. A 200 mm^2 Pt counter electrode and a Ag/AgCl reference electrode were employed in CV measurements. Durability tests were performed for 3000 cycles in aqueous electrolyte. The measurement conditions were set at a sweep rate of 1 to $100 \text{ mV} \cdot \text{s}^{-1}$ with a voltage range of 2.5 V in organic electrolyte, and 1.0 V in aqueous electrolyte. The temperature of the electrolyte was kept at $20 \text{ }^\circ\text{C}$ using a low-temperature incubator (CSB-900N Series-2; Asone Co. Ltd., Japan). Capacitances were calculated from CV measurements. We assumed that when the potential (V) applied to the working electrode was swept anodically or cathodically from V_1 to V_2 at a constant scan rate (r_s), the current, i , as a function of time, t , flowed to yield a total charge, $-Q$ or $+Q$, in each scan. The total charge was given by half the area S_c of the rectangular i -V curve which was then scaled by the mass of the activated carbon materials, m (g). Hence, the capacitance C ($\text{F} \cdot \text{g}^{-1}$) of the EDLC could be evaluated by Eq. (1) using the total one-way scan time t_s (s):



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$$C(F \cdot g^{-1}) = \frac{\int_0^{t_2} i dt}{m \int_{V_1}^{V_2} dV} = \frac{\frac{1}{z} \int_{V_1}^{V_2} i dV}{m \int_{V_1}^{V_2} dV} = \frac{\frac{1}{z} S_t}{m(V_2 - V_1)} \quad (1)$$

The complex impedance of the EDLC cells was measured with an impedance measurement system (FRA5020, Hokuto Denko Co. Ltd., Japan), using an AC two electrode method. A 0.2 mA AC current was applied across the EDLC cells with a frequency range of 10 mHz to 20 kHz.

III. RESULTS AND DISCUSSION

A. Characterization of activated carbon (CGCs)

The resultant surface morphologies of KOH-activated CGCs and CO₂-activated CGCs consisted of an amorphous surface structure. Graphitization was not observed in XRD pattern measurement. Particle sizes of KOH-activated CGCs were under 10 μm and those of CO₂-activated CGCs were either distributed around and under 30 μm in bigger ones or 400 nm in small ones (Fig. 1(a-b) and Fig. 1(e)). TEM images indicate surface morphologies of KOH-activated CGCs and CO₂-activated CGCs, and the typical distance between the graphene layers (002) was ~0.34 nm (Fig. 1(c-d)). N₂ adsorption-desorption isotherms of K900-1, K950-3, K1000-2 and C1000-2 CGCs at -196 °C are shown in Fig. 2. The N₂ adsorption isotherms of all synthesized CGCs displayed a typical type I isotherm according to the IUPAC classification. A type I isotherm is observed in micro porous solids having relatively small external surface, the limiting uptake being governed by the accessible micro pore volume rather than by the internal surface area.[17] Two distinct events were observed for the adsorption-desorption isotherms. The initial rapid adsorption at P/P₀ ~ 0.3 observed in each isotherm was mainly due to N₂ adsorption in micro pores. The subsequent gradual adsorption over a wide range of relative pressures (0.2 < P/P₀ < 0.96) was likely due to capillary condensation of N₂ in mesopores. Slight interaction between N₂ and the mesopores by desorption isotherm was observed in C1000-2 CGCs. These phenomena matched with the results of a previous report demonstrating that CO₂-activation promoted the development of mesopores.[13] In addition to the adsorption-desorption isotherms described above, data were also obtained for micro pore-diameter distribution curves (Fig. 3). BET surface area (S_{BET}), micro pore surface area (S_{micro}), mesopore surface area (S_{meso}), micro pore volume (V_{micro}), and mesopore volume (V_{meso}) data were also obtained (Table 2). The yields are also listed in the Table 2. Yields were calculated from CGCs mass divided by dried raw materials mass. The BET surface areas of the K950 series were higher than those of the K900 series at the same activation time. The K950-3 CGCs had S_{BET} = 1971 m²·g⁻¹, the largest BET surface area of all samples tested. All samples were found to have a high micro pore volume. Total pore volume of K950-3 CGCs is slightly larger than that of C1000-2 CGCs. In addition, the obtained BET surface areas, mesopore surface area, micro pore volume and mesopore volume in the case of temperature at 950 °C showed a tendency to increase and the yields showed a tendency to decrease with longer KOH activation times. On the other hand, CO₂-activated CGCs had lower micro pore surface areas and yields than KOH-activated CGCs, and C1000-2 CGCs had S_{meso} = 950 m²·g⁻¹, the largest mesopore surface area of all samples tested. It was reported that a reaction of carbon atoms with KOH formed micro-sized pores, while the reaction of carbon atoms with CO₂ formed meso-sized pores.[13] The amount of pore volume was determined by the activation method and temperature and time. However, high temperature and long time activation decreased the BET surface areas, for example, K1000-2 CGCs had S_{BET} = 1436 m²·g⁻¹ and C1000-3 CGCs had S_{BET} = 1467 m²·g⁻¹. Micro pore diameter distribution curves of K900-1, K950-3, K1000-2 and C1000-2 CGCs are shown in Fig.3. K900-1 CGCs possessed a pore diameter of 0.4-1.4 nm and K950-3 CGCs possessed a pore diameter of 0.4-1.8 nm. C1000-2 CGCs pore diameter showed a range of 0.8 to 1.8 nm. These tendencies indicated high temperature and long activating time increased pore diameters and high pore volume led to an increase the BET surface areas and micro pore volumes, except for K1000-2. K1000-2 CGCs pore diameter has sharp peak at around 0.6-0.7 nm. It was considered that topical pore diameters were generated by chemical reaction of carbon atoms and KOH at high temperature of 1000 °C and low pore volume over 0.7 nm led to decrease the BET surface areas.

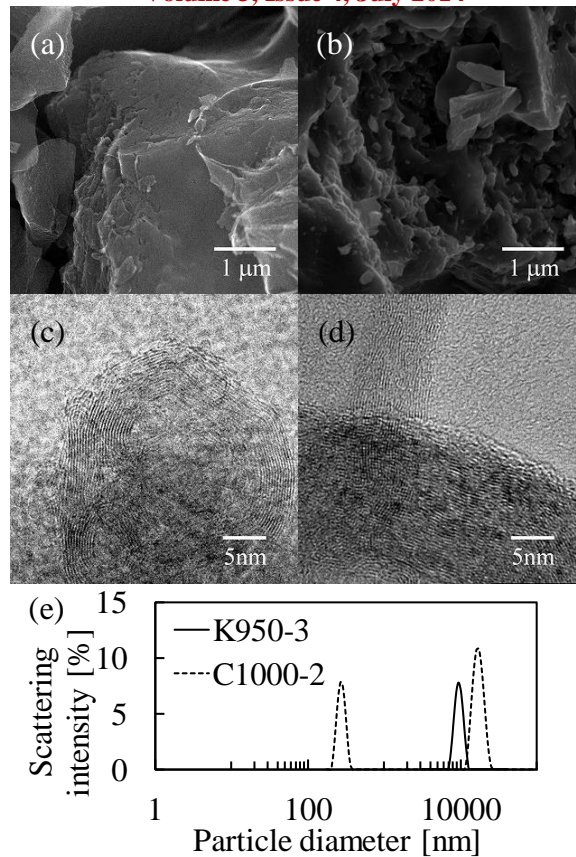


Fig 1. Microscope images of prepared CGCs. (a) SEM image of 950 °C and 3 h with KOH-activated CGCs; (b) SEM image of 1000 °C and 2 h in CO₂-activated CGCs; (c) TEM image of 950 °C and 3 h with KOH-activated CGCs; (d) TEM image of 1000 °C and 2 h in CO₂-activated CGCs. (e) Particle size analysis of 950 °C and 3 h with KOH-activated CGCs and 1000 °C and 2 h in CO₂-activated CGCs.

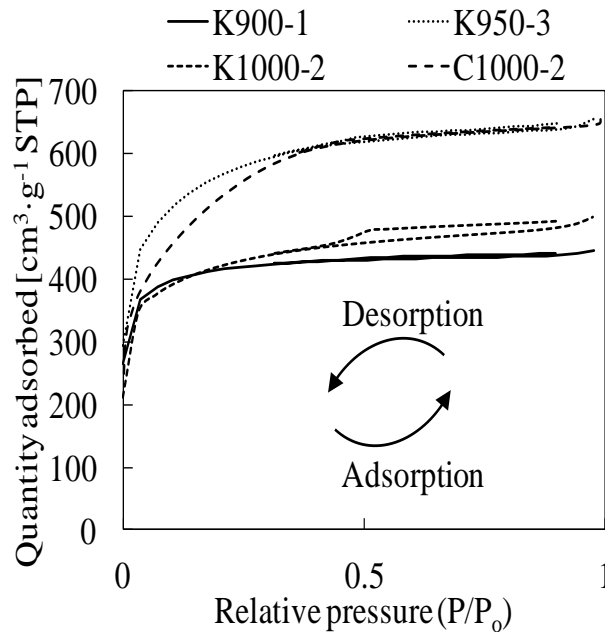


Fig. 2. N₂ adsorption-desorption isotherms of K900-1, K950-3, K1000-2 and C1000-2 CGCs.2

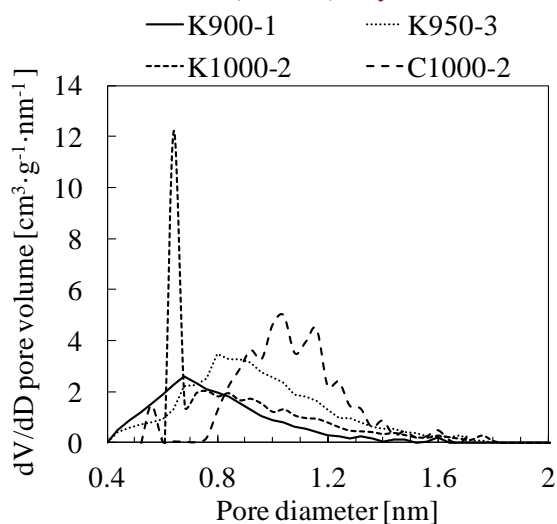


Fig. 3. Micro pore diameter distribution curves of K900-1, K950-3, K1000-2 and C1000-2 CGCs.

B. Electrochemical characterization of CGCs-mixed polarized electrodes

Fig. 4(a) show the CVs of the polarized electrode mixed with K900-3 or K950-3 CGCs at $10 \text{ mV}\cdot\text{s}^{-1}$ in $0.8 \text{ M } (\text{C}_2\text{H}_5)_4\text{NBF}_4/\text{PC}$. The CVs show that the electrolytic ions were inserted into and removed from the working electrode as voltage changes, with the nearly rectangular shape of the current-voltage curves suggesting a predominantly capacitive response. As shown in Fig. 4(a) in the case of K950-3, current densities became larger by increasing anion (BF_4^-) and cation ($(\text{C}_2\text{H}_5)_4\text{N}^+$) adsorption-desorption compared to K900-3 CGCs. The specific capacitance of the best sample in organic electrolyte was $127 \text{ F}\cdot\text{g}^{-1}$ using K950-3 CGCs (Table 3). K950-3 CGCs, such a large volume with relatively high surface area of micro pores and mesopores would contribute to an increase of amount of ion adsorption. It is said that a double layer capacitance of EDLC is dependent on the surface area of the electrode, and the surface area is influenced by micro pore volume. These results suggest that the polarized electrode with high micro pore volume and high micro-mesopore surface area can absorb more anions and cations in the organic electrolyte. Fig. 4(b) shows Nyquist plot of the mixed K900-3 and K950-3 CGCs polarized electrode. Each impedance spectrum is composed of a semicircle in the frequency range of 2–20 kHz and a linear part in the low-frequency range of 10 mHz to 2 kHz. The starting point of the semicircle represents the bulk resistance of the polarized electrode. The semicircular path diameter indicates the interface resistance. An equivalent circuit of the EDLCs is comprised of an interface resistance (R_i) and double layer capacitance connected in parallel and one section with bulk resistance (R_b) connected in series, as shown in Fig. 4 (b). Results of internal resistance (R_b+R_i) analysis by electrochemical impedance spectroscopy are also shown in Table 3. KOH-activated carbons had lower internal resistances than those of CO_2 -activated carbons and a low internal resistance of 2.95Ω was obtained using K950-3 CGCs. Fig. 4(c) shows capacitance vs. scan rate characteristics of K950-3 CGCs electrode in $0.8 \text{ M } (\text{C}_2\text{H}_5)_4\text{NBF}_4/\text{PC}$. It was confirmed that the capacitance of the K950-3 CGCs electrode with a scan rate of $100 \text{ mV}\cdot\text{s}^{-1}$ was decreased by 30.7% compared to the same material with a scan rate of $1 \text{ mV}\cdot\text{s}^{-1}$. Fig. 5(a) shows the CVs of the polarized electrode mixed with K900-1 and C1000-2 CGCs at $10 \text{ mV}\cdot\text{s}^{-1}$ in $0.5 \text{ M } \text{H}_2\text{SO}_4$. The specific capacitance of the best sample in aqueous electrolyte was $204 \text{ F}\cdot\text{g}^{-1}$ using K900-1 CGCs (Table 3). In aqueous solution, the responses were not only due to the surface area of the activated carbon, but also have been shown to be due to redox reactions of the functional groups on the surface of the CGCs. Oda et al. reported that a carboxyl group has a strong polarity that causes leakage current due to a catalytic effect and the Faradaic current causes a pseudo-capacitance.[20] Therefore, it was believed that the wettability of the electrodes was influenced by oxygen-containing surface functional group generated. The results of XPS spectra of K900-1 CGCs are shown in Fig. 5(b). The C-C, C-O, C=O and O-C=O bond spectra of the CGCs were analyzed by deconvolution of the obtained spectrum. The acquired spectrum was deconvoluted into four-component spectra with peak energies of 284.9, 286.3, 287.7, and 289 eV using a nonlinear least-squares-fitting program with a symmetric Gaussian function. The spectrum at around 284.9 eV corresponds to a C-C bond; the spectrum at around 286.3 eV is due to a C-O bond, which indicates the presence of a phenolic hydroxyl group; the spectrum at around 287.7 eV is due to a C=O bond; the spectrum at around 289 eV is due to a O-C=O bond, which indicates the presence of carboxyl groups. O 1s spectra at around 532 eV were also observed, which indicates the presence

of an oxygen-containing functional group. Table 3 includes the estimated O-C=O/C-C ratios on the surface of the obtained CGCs. It is clearly seen that KOH activation (K-samples) generates high O-C=O/C-C ratios (0.22-0.38) compared to those of CO₂ activation (C-samples). The specific capacitance of the best sample was 204 F·g⁻¹ using K900-1 CGCs, which has S_{BET} = 1389 m²·g⁻¹ showing 30 % lower surface area compared to the highest surface area sample, K950-3. On the other hand, O-C=O/C-C ratio of the K900-1 CGCs was 0.35. Compared to the nearly characterization sample, K950-1 CGCs had a specific capacitance of 170 F·g⁻¹ with O-C=O/C-C ratio of 0.22. It is observed that a high O-C=O/C-C ratio leads to increased specific capacitances. The specific capacitances in aqueous electrolyte were gradually increasing with increase of O-C=O/C-C ratios, as calculated by using Table 3 data. The presence of a carboxyl group resulted in an increase of redox reactions with wettability of the electrodes and increased pseudo-capacitance. Results of internal resistance (R_b+R_i) analysis by electrochemical impedance spectroscopy are also shown in Table 3. There were no significant differences in internal resistances of both KOH-activated CGCs and CO₂-activated CGCs and a low internal resistance of 0.800 Ω was also obtained using K900-1 CGCs. It is well known that the aqueous system shows higher capacitances compared to organic one in carbon electrodes because of high relative permittivity and ion size. Capacitance deterioration after long cycles is not uncommon; therefore, cycle tests were performed in order to investigate the electrochemical durability of the K900-1 for long cycles. Fig. 5(c) shows the electrochemical stability of the K900-1 and C1000-2 CGCs electrodes, which indicate the capacitance under 3000 cycles for CV tests with a scan rate of 10 mV·s⁻¹. These capacitances of K900-1 and C1000-2 CGCs were clearly stable, remaining in the range of 216–222 and 131–150 F·g⁻¹, respectively. The durability tests suggest that the CGCs have long-cycle reliability for frequent charging and discharging.

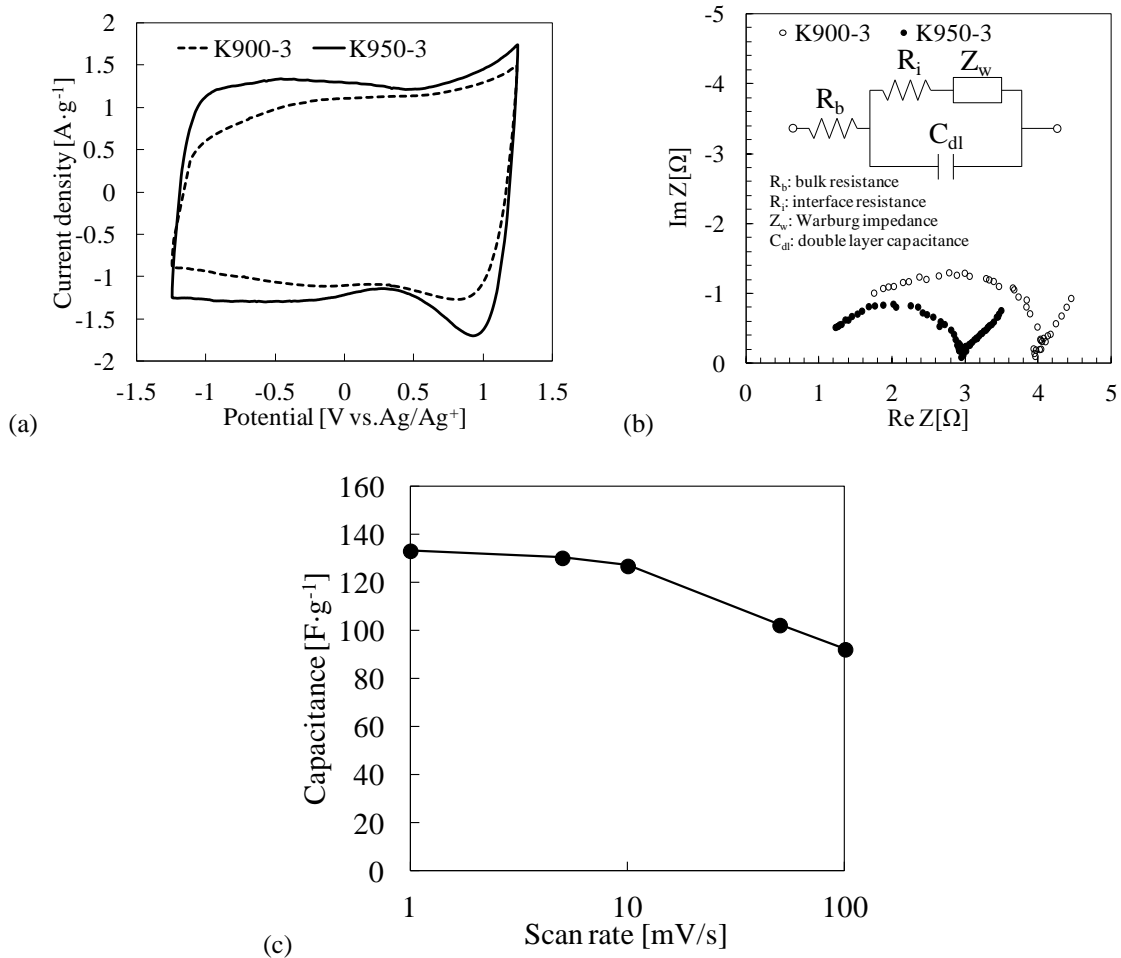


Fig. 4. (a) Cyclic voltammogram of the mixed K900-3 and K950-3 CGCs polarized electrode at 10 mV·s⁻¹ in 0.8 M (C₂H₅)₄NBF₄/PC. (b) Nyquist plot of the mixed K900-3 and K950-3 CGCs polarized electrode. (c) Capacitance vs. scan rate characteristics of K950-3 CGCs electrode in 0.8 M (C₂H₅)₄NBF₄/PC.

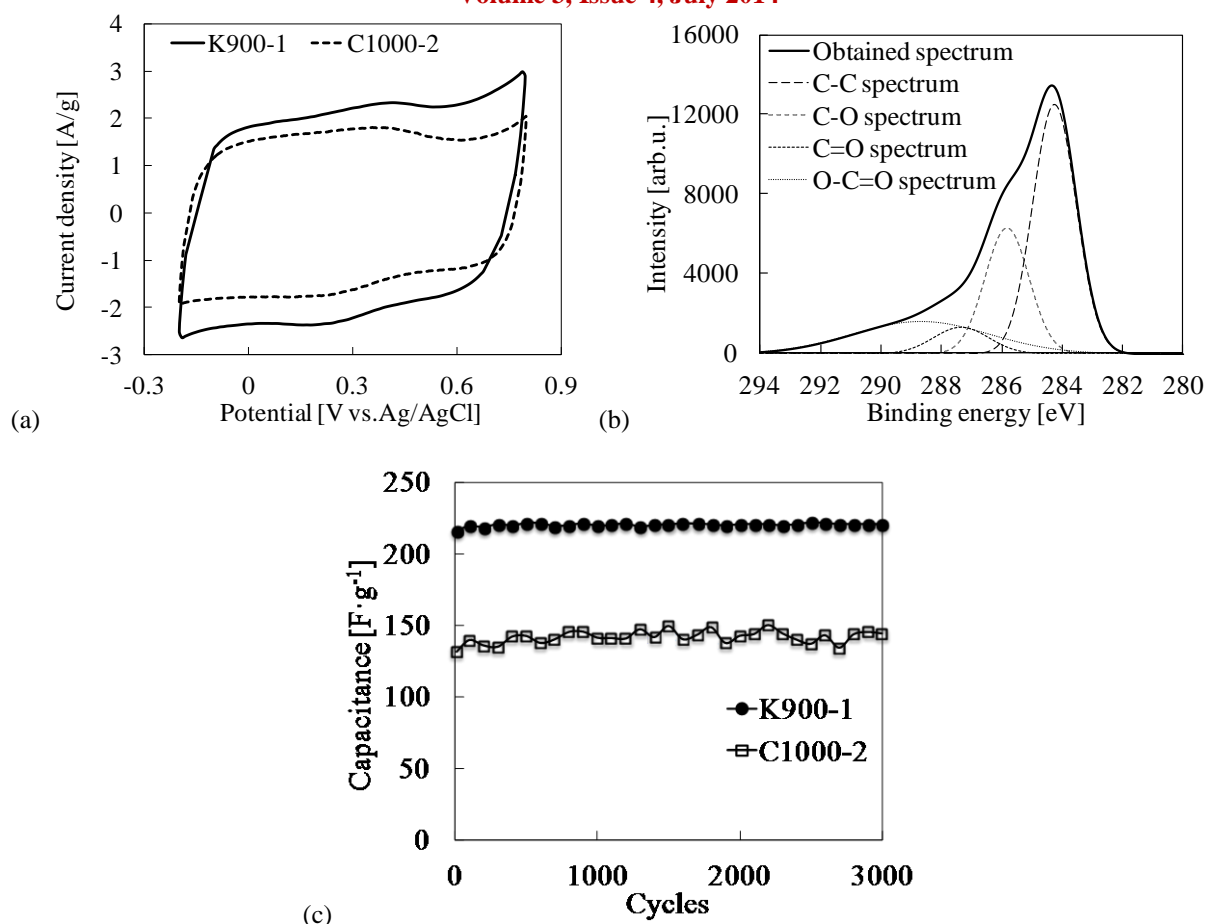


Fig. 5. (a) Cyclic voltammogram of the mixed K900-1 and C1000-2 CGCs polarized electrode at $10 \text{ mV} \cdot \text{s}^{-1}$ in $0.5 \text{ M H}_2\text{SO}_4$. (b) XPS spectra of K900-1 CGCs. (c) Durability test (3000 cycles) of K900-1 and C1000-2 CGCs electrodes at $10 \text{ mV} \cdot \text{s}^{-1}$ in $0.5 \text{ M H}_2\text{SO}_4$.

C. Cost analysis of activated carbon materials

The activated carbon used in general EDLC applications accounts for 30% of all manufacturing costs. NEDO (New Energy and Industrial Technology Development Organization) in Japan open free access to after evaluation report of “Carbon Nanotube Capacitor Development Project” in 2013.[21] In this report, the cost-performance ratio of EDLCs is 1.5-2.5 Yen (JPY) $\cdot F^{-1}$ in the case of large size EDLCs (estimated using organic electrolyte). In this work, actual cost of KOH and HCl as washing to remove impurities were taken into consideration and the manufacturing cost of CGCs were calculated as $16.4 \text{ Yen} \cdot \text{g}^{-1}$. Next, actual cost of transportation from the beverage company to the University of Miyazaki (about 50 km) was 910 Yen by train for 100 kg of used coffee grounds (dry materials). Maintenance cost was estimated as 44,100 Yen (actual cost of alumina tube) for every 100 kg used coffee grounds. The total cost of CGCs made by the University of Miyazaki was $16.9 \text{ Yen} \cdot \text{g}^{-1}$. The largest capacitance of $127 \text{ F} \cdot \text{g}^{-1}$ in high surface area CGCs was obtained in organic electrolyte. The cost-performance of the activated carbon was calculated as $0.133 \text{ Yen} \cdot F^{-1}$. The activated carbon accounts for 30% of all manufacturing costs, and the cost-performance of EDLCs was calculated as $0.444 \text{ Yen} \cdot F^{-1}$. The CGCs decreased the cost-performance ratio compared to activated carbon materials currently on the market.

IV. CONCLUSIONS

Activated carbon materials derived from used coffee grounds were employed in the fabrication of polarized electrodes for EDLC applications. The influence of the materials on surface morphologies, capacitance behaviors and internal resistances were then examined. The obtained KOH-activated CGCs had very high surface areas and high micro pore volumes. These combined features contributed to observed capacitances of $127 \text{ F} \cdot \text{g}^{-1}$ using K950-3 in $0.8 \text{ M (C}_2\text{H}_5)_4\text{NBF}_4/\text{PC}$ and $204 \text{ F} \cdot \text{g}^{-1}$ using K900-1 in $0.5 \text{ M H}_2\text{SO}_4$ electrolytic media. Low internal



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resistance of 2.95Ω in $0.8 \text{ M } (\text{C}_2\text{H}_5)_4\text{NBF}_4/\text{PC}$ and 0.80Ω in $0.5 \text{ M } \text{H}_2\text{SO}_4$ electrolytic media were also obtained. The cost-performance ratio of EDLCs was calculated as $0.444 \text{ Yen}\cdot\text{F}^{-1}$ for organic electrolyte. CGC materials derived from used coffee grounds were demonstrated to be a highly valuable source of low-cost activated carbon for electrochemical applications.

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