

Characteristic of Thermally Reduced Graphene Oxide as Supercapacitors Electrode Materials

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Abstract. We investigated graphene like material named reduced graphene oxide (RGO) as an electrode material by employed graphene oxide (GO). Thin film of GO was prepared on the indium thin oxide (ITO) substrate by spin-coating method using varied concentration of GO that dispersed in water. In order to remove its oxygen contained, GO film was thermally reduced at 200 °C for 1 hour. We used cyclic voltammetry to measure its CV characteristic and estimated its specific capacitance. We obtained the highest specific capacitance of 6.53 mF g⁻¹ that measured from 4 mg ml⁻¹ RGO thin film at scan rate 25 mVs⁻¹.

1. Introduction

Supercapacitor is known as a complement of other power sources such as batteries and fuel cells, and it can have long life cycles and can be rapidly charged and discharged at high power densities due to its highly reversible charge storage process [1]. Therefore supercapacitor is a potential candidate as an alternative energy storage devices.

Graphene has a potential application as an electrode material for electrochemical energy storage since it has very high electrical conductivity and a high specific surface area (2630 m² g⁻¹) [2]. It is reported an intrinsic capacitance of graphene of 21 μF cm⁻² [3]. Graphene oxide (GO) is a most common starting material for graphene based applications since it can be produced in large quantities [4]. Since GO contains various oxygen functional groups, it is necessary to remove them in order to recover its conjugated structure [5, 6]. Thermal reduction using heat treatment is a common way to remove oxygen contained groups of GO [7].

In this paper, we report some results on measurement of specific capacitance value of reduced graphene oxide (RGO) films using cyclic voltammetry at varied scan rate in 1 M H₂SO₄ electrolyte.

2. Experiments

We used a commercial GO suspension (Graphenea) that dispersed in water. The GO dispersion was sonicated using ultrasonic cleaner (Branson 1800) for 1 hour at ambient temperature to exfoliate GO before used for film preparation. Indium Tin Oxide glass (ITO glass) was employed as a conductive substrate. The substrate was cleaned with teepol, rinsed with milli-Q water, then sonicated in ethanol



at 50 °C and dried. The GO films was prepared on ITO glass using spin-coating method (Chemat Technology Spin-coater KW-4A) at 1000 rpm and dried at 30 °C. The GO films were prepared using varied concentration from 1 to 4 mg ml⁻¹. In order to obtain RGO films, the GO films were thermally reduced by heating at 200 °C for 1 hour.

Cyclic voltammetry (CV) measurements were carried out using a potentiostatic (Metrohm Autolab) with a platinum as a counter electrode, an Ag/AgCl as a reference electrode, and RGO film as a working electrode. The measurements were done at ambient temperature in 1 M H₂SO₄ aqueous electrolyte. Cyclic voltammetry scans were recorded from -0.2 V to 0.8 V in varied scan rates.

3. Results and Discussions

The cyclic voltammetry curves (CV) of RGO samples that prepared using varied concentration of 1, 2, 3 and 4 mg ml⁻¹ on ITO glass substrates that were measured in a three-electrode configuration using 1 M H₂SO₄ as the electrolyte at varied scan speed are showed in Figure 1. The CV curves indicate oxidation and reduction behavior of RGO samples that the oxidation and oxidation currents depend on the film thickness, the thicker sample leads to higher oxidation and reduction currents.

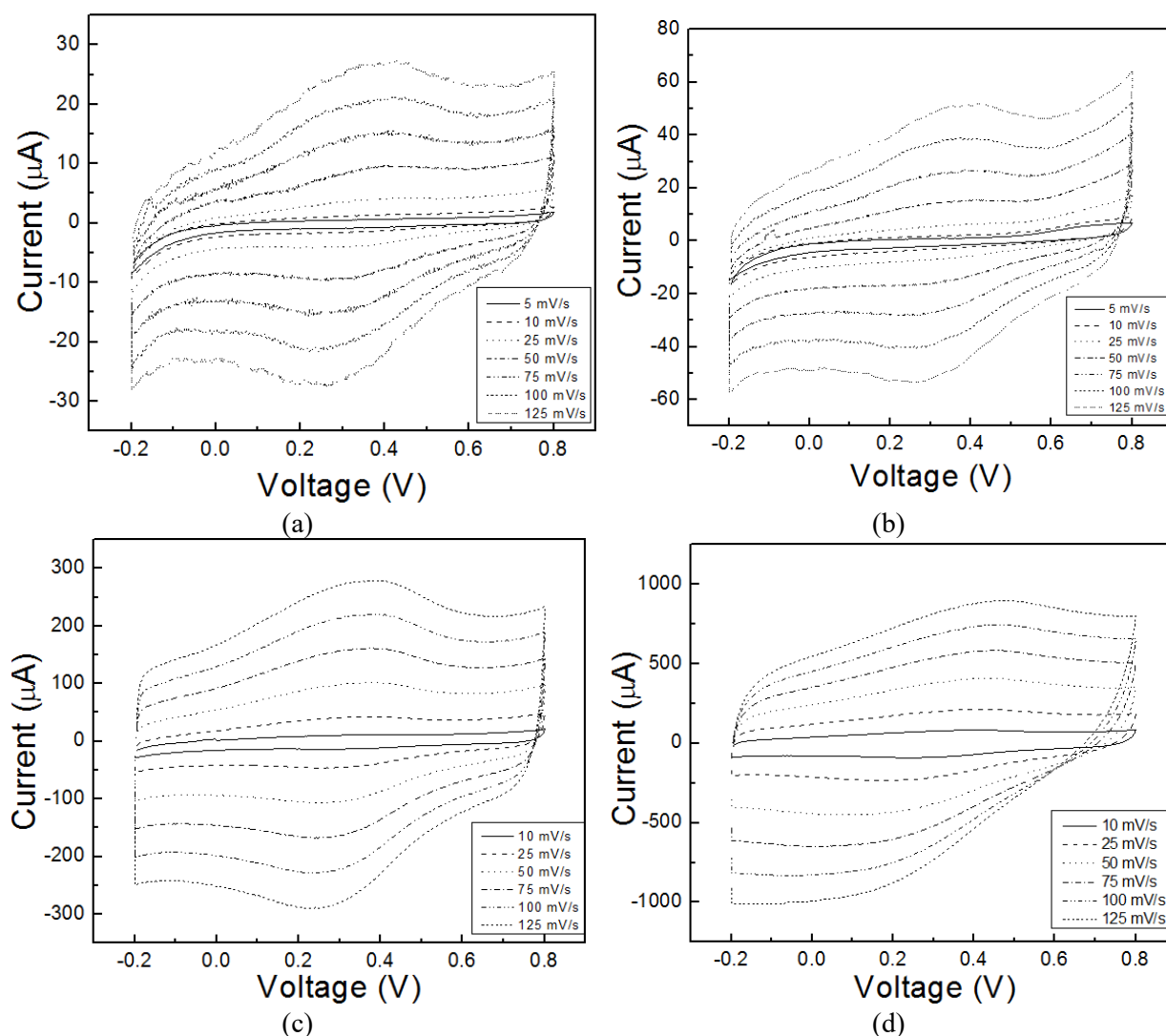


Figure 1. Cyclic Voltammetry curves of RGO electrodes on ITO glass substrate at various scan rates of 5, 10, 25, 50, 75, 100 and 125 mVs⁻¹ measured from -0.2 V to 0.8 V of samples that prepared using varied GO concentration, 1 mg ml⁻¹ (a), 2 mg ml⁻¹ (b), 3 mg ml⁻¹ (c), and 4 mg ml⁻¹ (d).

From the Figure 1, it is obvious that all CV curves of RGO electrode measured from -0.2 V to 0.8 V at various scan rates of 5, 10, 25, 50, 75, 100 and 125 mVs⁻¹, respectively show a capacitive behavior. The increasing of a scan rate is only slightly distorted rectangular-like shape of CV curves, that is similar as previously reported [8]. The rectangular-like shape of the CV curves of prepared RGO films clearly indicated electrochemical double-layer capacitance characteristics, similar as previously reported [8]. The figures also show that each similar rectangular shape of CV has one pair of redox peaks. It is assigned as a typical transition between quinone or hydroquinone groups of carbon materials [9]. Electrochemical behavior of RGO electrode in 1 M H₂SO₄ has similar shape as reported previously that interpreted as a hybrid type of both a pseudocapacitive and an electric double layer capacitive [10].

Effect of increasing a scan rates from 5 to 125 mVs⁻¹ on CV curves of RGO electrode at four different RGO concentration are also shown in Figure 1. The curves are not deformed due to increment of scan rate, but proportionally enlarged as scan rates increased up to 125 mVs⁻¹. We can see an obvious hysteresis of the CV curves.

The CV hysteresis area for these samples are increased due to increasing of the scan rates. In order to get a quantitative value of an increment of the hysteresis area, we applied equation (1) [11] on the data of Figure 1. The symbol P is a CV hysteresis area (in μ W), i is a current respond, and dV is step potential that was 0.00244 V in this measurement.

$$P = \left(\int_{-0.2}^{0.8} i dV \right)_{ox} + \left(\int_{0.8}^{-0.2} i dV \right)_{red} \quad (1)$$

The hysteresis area of each samples that calculated using equation (1) are shown in Figure 2. We can see that the CV hysteresis area of each samples are increased with the rise of scan rate. It is related with current response of RGO electrode, which at the higher scan rate the RGO electrode can give the higher current response. This characteristics indicate a stability of capacitance of the RGO electrode. In order to obtain a specific capacitance value of the RGO electrodes from CV curves, we employed equation (2) [11] and using previous calculation results of hysteresis area. The symbol C is specific capacitance of RGO (F g⁻¹), m is mass of electro-active materials (g), ΔV is potential window (V), s is scan rate (mV s⁻¹), and i is current response in a given potential V.

$$C = \frac{1}{2m \Delta V s} \left(\int_{-0.2}^{0.8} i dV \right)_{ox} + \left(\int_{0.8}^{-0.2} i dV \right)_{red} \quad (2)$$

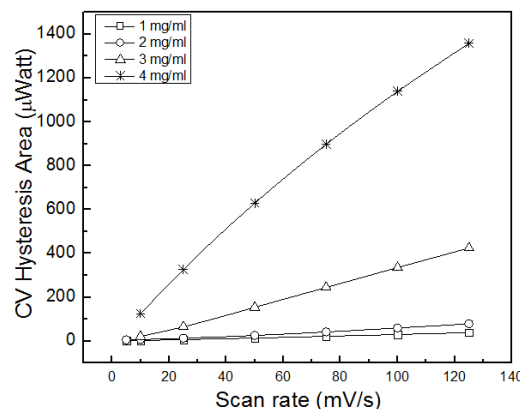


Figure 2. The CV hysteresis area of various RGO electrodes that prepared from varied concentration of GO dispersion that measured at various scan rates.

The calculation results of specific capacitance of RGO electrodes that prepared from varied concentration obtained from varied scan rates measurements are shown in Figure 3 (a) and (b). The specific capacitance of RGO electrode that prepared from certain concentration is remain the same

when measured at varied scan rates. Further for RGO electrodes prepared from 1, 2, and 4 mg ml⁻¹, increasing the scan rates can result a decreasing of the specific capacitance. It can happen because an increment of CV hysteresis area is inversely proportional with scan rates increment as shown by equation (2). The highest specific capacitance that can be reached in this experiment is 6.53 mF g⁻¹. It is obtained from RGO electrode which was prepared using 4 mg ml⁻¹ and scan rate 25 mV s⁻¹.

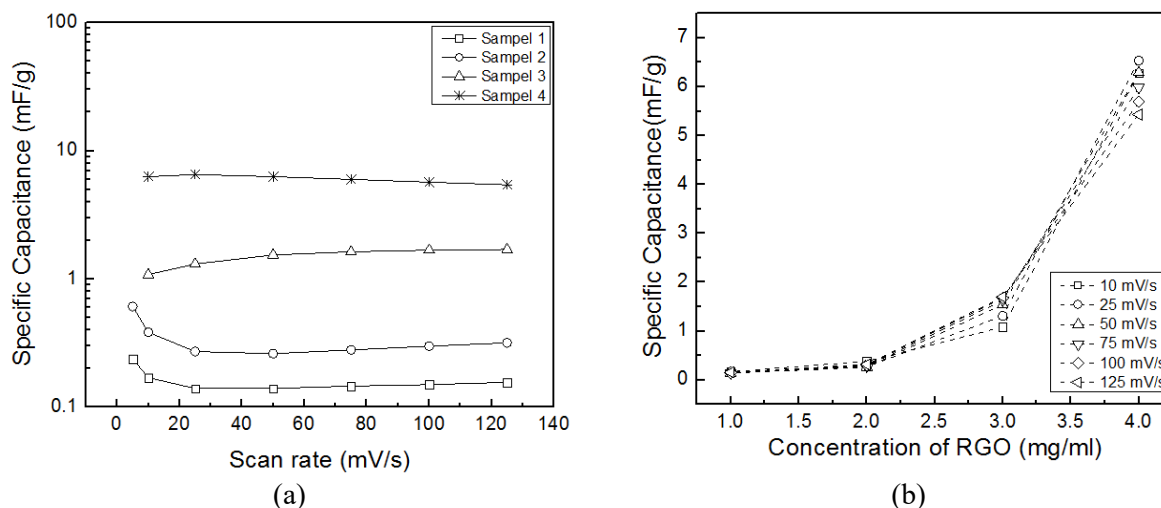


Figure 3. The calculated specific capacitance of RGO electrodes at various scan rates that prepared from varied concentration of GO dispersion (a), and at various concentration that measured at varied scan rates (b).

4. Conclusions

The thermally RGO film is potentially used as an electrode material for supercapacitor as confirmed by cyclic voltammetry measurement. The capacitance of RGO film will be increased by increasing film thickness through using higher concentration of GO dispersion and will be decreased by increment a scan rate. The highest specific capacitance obtained in this work is 6.53 mF g⁻¹ from RGO film that prepared using 4 mg ml⁻¹ GO dispersion and measured at scan rate of 25 mVs⁻¹.

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