Supporting online information

Li-Air Rechargeable Battery based on Metal Free Graphene Nanosheets Catalysts

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Fig. S1(a) shows the discharge curves of heat-treated GNSs at a current density of 0.5 $mA \cdot cm^{-2}$ for 24 h at room temperature. The discharge voltage of the heat-treated GNSs remained at about 2.95 V. Fig. S1(b) shows the charge–discharge curves of heat-treated GNSs at a current density of 0.5 $mA \cdot cm^{-2}$ for 1 to 50 cycles. The voltage gap between the charge voltage and discharge voltage at the first cycle for heat-treated GNSs was approximately 0.95 V, which is higher than that of GNSs (0.56 V). However, after charge–discharge cycling, the differences in voltages between the first and the fiftieth cycle were 0.16 V for charge and 0.07 V for discharge of heat-treated GNSs, which are lower than the corresponding values for GNSs (0.4 V for charge and 0.2 V for discharge).

Fig. S2 shows the TG and DTA curves of GNSs, heat-treated GNSs, graphite oxide (GO), and AB. The temperature was increased from room temperature to 800 °C at a 5 $°C \cdot min^{-1}$ under an atmosphere of air. The TG curve for GNSs showed significant weight loss (20%) below 100 °C, probably as a result of removal of water molecules present in the GNSs [Fig. S2(a)]. The large weight loss of 15% observed in the range 80–450 °C was attributed to the removal of the functional group from the GNSs surface. These results show that although there may be functional groups present on the GNSs surface, the number of these functional groups is much less than that on GO [Fig. S2 (c)]. Moreover, Fig. S2(a) shows that GNSs is burned out at 450–630 °C with a peak of heat production at around 530 °C, corresponding to combustion of the carbon framework. The relative content of the carbon framework was estimated to be 62% by integration of the TG curve. In contrast, as shown in Fig. S2(b), the TG curve of heat-treated GNSs showed that the sample lost little mass (3%) below 50 °C and then lost significant amounts of mass (85%) from 500 to 700 °C as a result of combustion of the carbon framework. This suggested that almost all the water molecules and functional groups on GNSs were removed by heating at 950 °C. From these results, heat-treatment of GNSs can be regarded as leading to restoration of the carbon framework. For AB [Fig. S2(d)], two characteristic DTA peaks were observed at about 430 and 700 °C, corresponding to the combustion of functional groups and the carbon framework, respectively.

Table 1 shows the content of functional groups on GNSs and heat-treated GNSs as calculated by integrating the XPS peaks. The content of sp^2 carbons for GNSs is reduced while that of oxygen-related sp^3 carbons increases. However, in the case of heat-treated GNSs, oxygen-related functional group were removed and, therefore, the sp^2 carbon content increased significantly compared with that of GNSs. Furthermore, the oxygen content of heat-treated GNSs is less than that of GNSs.

Fig. S3 shows the Raman spectra of GNSs and heat-treated GNSs. The two peaks at 1588 and 1330 cm^{-1} are called the G and D bands, respectively, and they can be assigned to the active E_{2g} vibration of graphite and disordered structures, respectively. It is known that the intensity ratio of the G and D peaks (I_G/I_D) provides information on the degree of graphitization of GNSs. We found that the intensity ratio of I_G/I_D increased on annealing of GNSs, indicating that graphitization of the GNSs occurs.

Table 1 the atomic concentration of C_{1s} and O_{1s} peak obtained from XPS analysis

	Graphene nanosheets		Heat-treated graphene nanosheets		
	Binding energy/ eV	%	Binding energy/ eV	%	
C_{1s}	284.6	54.1	284.6	70.3	C=C (sp^2)
	285.6	16.6	285.7	13.2	C-C(sp^3)
	286.8	6.9	286.9	5.6	C-O
	288.3	5.0	288.4	2.3	C=O
		31.0		19.0	sp^3/sp^2
O_{1s}	531.2	2.8	530.9	0.6	O-C=O
	532.4	3.7	532.3	0.4	C=O
	533.6	2.2	533.5	0.6	C-O

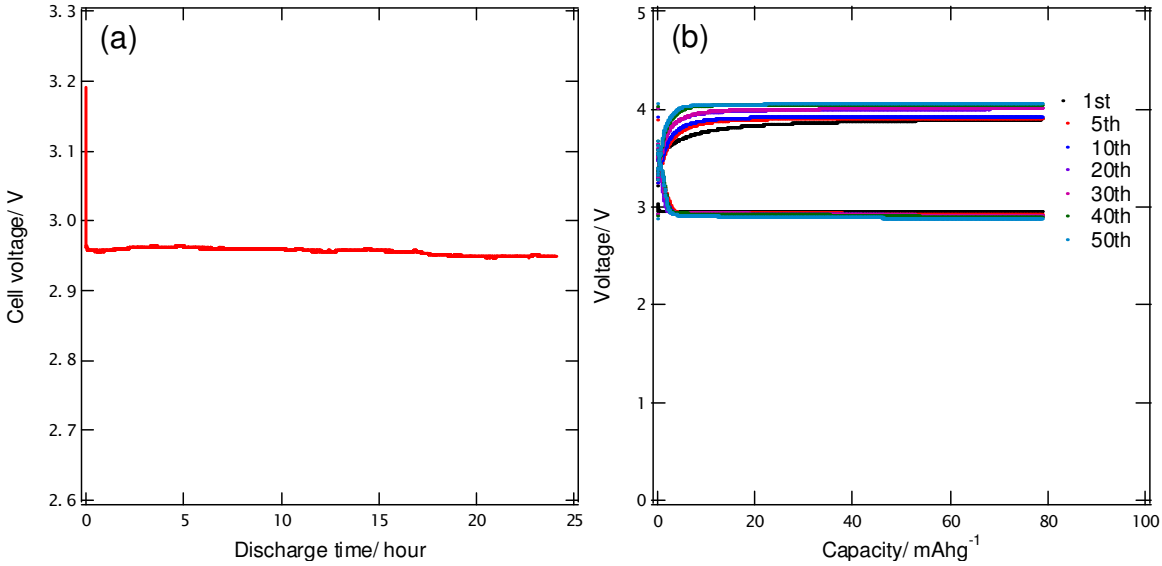

Fig. S1 Discharge voltage of the heat-treated GNSs at a current density of 0.5 mA·cm^{-2} for 24 h (a), the correlation between cycle number and capacity of heat-treated GNSs (b)

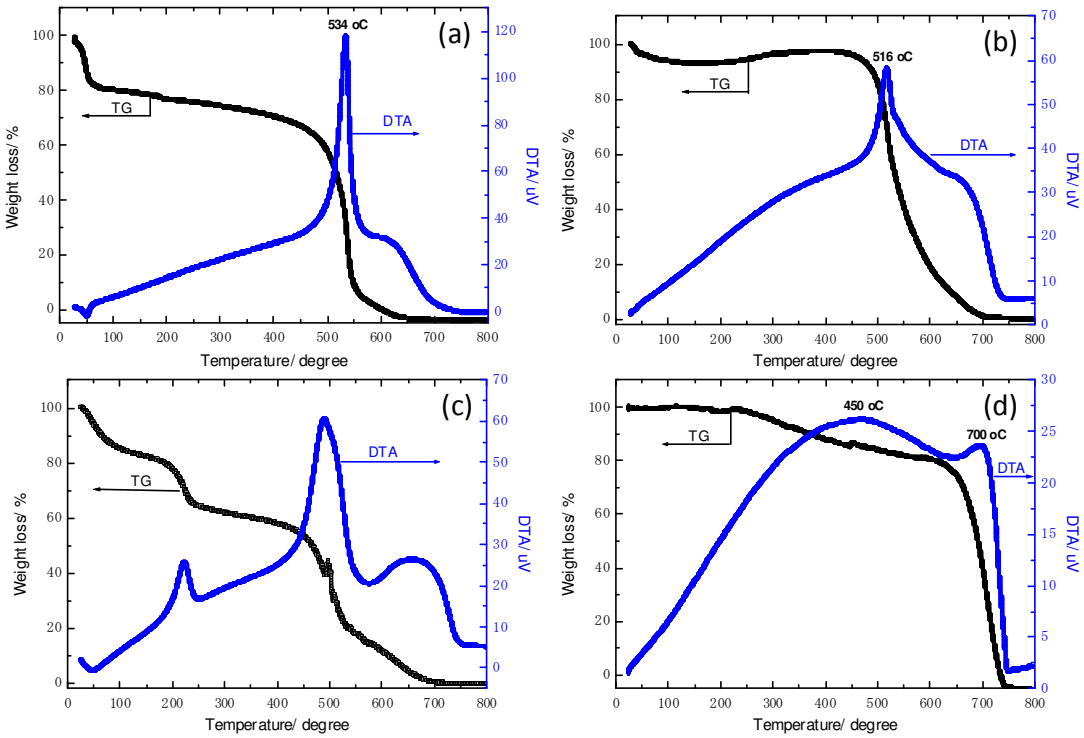

Fig. S2 TG and DTA result of (a) GNSs, (b) heat-treated GNSs, (c) GO and (d) AB

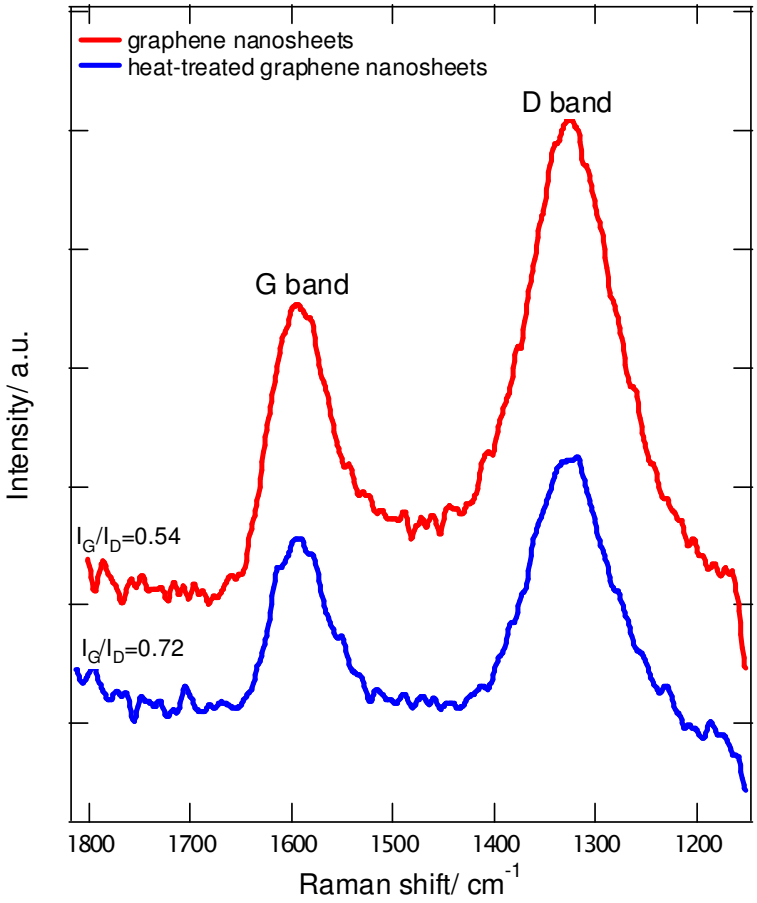

Fig. S3 Raman sepectra of GNSs and heat-treated GNSs