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The specific capacitance was measured bv chronopotentiometry and calculated by $C = I/[(dE/dt) \times m] \approx$ $I/[(\Delta E/\Delta t) \times m]$, where I is the constant discharging current, dE/dt indicates the slope of the discharge plot of the $_5$ discharging curves, and *m* is the mass of the corresponding electrode materials measured.

Shown in Fig. 6, the sample weight decreases slowly between 100 and 240 °C, this is expected to be due mainly to the removal of chemically adsorbed H₂O; A very obvious 10 weight loss of 10.65% is present between 240 and 290 °C on

the TG curve, it is believed that the reaction of

 $Ni(OH)_2 \rightarrow NiO + H_2O$ (4)

occurs in this region; With the increase of temperature over 290 °C, the TG curves become flat, which indicates that no

15 phase transformation occurs. With the hydroxyl groups removed from Ni(OH)₂, the NiO crystal phase is expected to be formed, and further heating could only make the structure of the products more crystalline, which is confirmed by the following XRD results. This is the reason why the sample was 20 calcined at 250, 300, and 350 °C.

The electrochemical impedance measurements were carried out on the NiO electrodes at 0.229 V (vs. SCE). The typical results are shown in Fig. 7. The complex plane impedance plots for each sample can be divided into the high-frequency

- 25 component and the low-frequency component. A distinct knee in the frequency can be observed in curves of Fig. 7. From the point intersecting with the real axis in the range of high frequency, the internal resistances (which equal to R_b) of the NiO electrodes are 1.0, 1.1 and 1.4 Ω , corresponding to the
- 30 NiO samples obtained at 250, 300 and 350°C, respectively. It includes the total resistances of the ionic resistance of electrolyte, intrinsic resistance of active materials and contact resistance at the active material/current collector interface. The semicircle in the high frequency range associates with the
- 35 surface properties of porous electrode corresponds to the faradic charge-transfer resistance (R_{ct}). It can be seen that the combination resistance of ionic resistance and charge-transfer resistance of the NiO materials is in the order of 250 $^{\circ}C < 300$ $^{\circ}C < 350 ~^{\circ}C$ prepared materials. This can be explained that
- 40 with the increase of the heat-treatment temperature, some NiO nano-sheets grew to a stacking particles, the decrease in the specific surface area can weaken the diffusivity of the electrolyte irons in the NiO electrode. At the lower frequencies, a straight sloping line represents the diffusive
- 45 resistance (Warburg impendence) of the electrolyte in electrode pores and the proton diffusion in host materials. The phase angles for impedance plots of all the NiO electrodes were observed to be higher than 45° in the low frequencies clearly. These findings suggest that all the electrodes are not
- 50 controlled by diffusion process, implying the good accessibility of the ions and/or the possible contributions of supercapacitors.¹⁵



Fig. 6 TG plots of Ni(OH)2 precursor.



Fig. 7 Complex-plane impedance plots of the NiO electrodes prepared at different heat treatment temperatures (0.229 V vs. SCE; electrolyte: 2 M KOH). The inset is the equivalent 60 circuit.









Microsphere growing

Growing on microsphere surface

(B) Surface growth

Fig. 8 schematic formation mechanism of the as-prepared Ni(OH)₂ precursor.

¹⁵ M. W. Xu, D. D. Zhao, S. J. Bao, H. L. Li, J. Solid State Electrochem. 2007, 11, 1101; S. S. Zhang, K. Xu, T. R. Jow, Electrochim. Acta 2004, 49, 1057.