

Challenge to overcome carbon corrosion in supercapacitors

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Abstract:

Supercapacitors are rechargeable devices with the advantages of high power density and long-term stability despite their disadvantage of low energy density, compared with secondary batteries. Conventionally, activated carbons (ACs) are used as electrode materials, but their stability is not sufficient for use in objectives such as automobiles, as they require a high voltage and high temperature operation. There are two major methods to expand the working voltage of symmetric supercapacitors: (1) the development of stable electrolytes such as ionic liquids and (2) the development of stable carbon materials. The former has been intensively investigated, whereas very stable electrolytes are too expensive for practical applications. On the other hand, the latter including the study on the mechanism of corrosion reactions has been less studied because of difficulty in precise control of complex porous carbon structures.

In order to develop highly durable carbon materials, it is first necessary to understand the mechanism of the carbon corrosion reactions. However, there have been only a limited number of studies because of the very complex structures of porous carbons including ACs. Thus, on the first topic, our recent work on the carbon corrosion sites^[1] is presented. A variety of carbon materials are analyzed with many characterization techniques such as X-ray diffraction, Raman spectroscopy, N₂ adsorption, magnetic susceptibility measurement, and temperature programmed desorption up to 1800 °C, to find out the origin of corrosion reactions in an organic electrolyte. While carbon crystallinity and porosity are not directly related to the positive-electrode corrosion, a good correlation is found between the corrosion charge and the number of carbon edge sites terminated by H and oxygen-functional groups which are decomposed and release CO. On the second topic, the development of super-stable carbon material is presented. The material is a seamless mesoporous carbon sheet consisting of continuous graphene walls.^[2] The sheet contains very few carbon edge sites (only 4% of the number present in conventional activated carbons) despite its high specific surface area (1500 m² g⁻¹), and it is possible to use it to assemble symmetric supercapacitors with excellent stability under 3.5 V@60 °C and 4.4 V@25 °C conditions, even using a conventional electrolyte (1 M Et₃MeNBF₄/propylene carbonate). Moreover, high-voltage operation at 4.4 V results in a 2.7 times higher energy density compared to that achieved using conventional activated carbons.

Key words: supercapacitors; carbon corrosion; graphene

References

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