Fabrication of 100 mAh-class Organic Radical Battery

K. Nakahara, J. Iriyama, S. Iwasa,
Y. Morioka, M. Suguro and M. Satoh

Functional Materials Research Laboratories,
NEC corporation

Miyazaki, Miyamae-ku, Kawasaki, Kanagawa,
216-8555, Japan.

Stable polyradicals have been attractive as a new type of organic cathode active materials. The use of organic materials as an active material in cathodes appears promising to design environmentally friendly, functional and high energy-density rechargeable batteries. We have been reported that the electrochemical properties of a stable nitroxyl polyradical, poly (2,2,6,6-tetramethylpiperidinyloxy methacrylate) (PTMA) (Fig.1), and the performance of the prototype coin cells using this design[1][2]. Here we describe a development of 100 mAh-class organic radical batteries and their battery performances.

The 100 mA-class organic radical batteries were fabricated as follows (Fig. 2). First, we synthesized an insoluble and stable nitroxyl radical polymer (PTMA) by using a radical polymerization of 2,2,6,6-tetramethylpiperidine methacrylate monomer (ASAHI denka, LA-87) with 2,2'-azobisisobutyronitrile, followed by oxidation with 3-chloroperoxybenzoic acid. A polymer pale orange in color was purified by reprecipitation with methanol. Then, 30 wt% of PTMA powder (specific density 1.15 g cm^{-3}, 600 mg) and 60 wt% of carbon black powder (1200 mg) were mixed and tempered with 10wt% of polytetrafluoroethylene powder (DAIKIN industries, F-104, 200 mg). The resulting clay was spread into the form of a thin board, and cut into the size of 52 × 70 mm. This composite cathode was dried under a high vacuum at 80 ^{o}C and impregnated with an electrolyte solution of DEC (7.0 g) and EC (3.0 g) into which $LiPF_6$ (1.5 g) had been dissolved. We put the composite cathode on a Al current collector, and on top of that we put a porous polyethylene film separator and Li metal anode (30 μm) and another current collector. We piled 6 layer of cathodes and anodes and packed with Al laminated films.

The fabricated battery was charged at constant current of 100 mA until its voltage reached 4.2 V. In the charge-discharge curves, an obvious voltage plateau is evident. The found plateau voltage of nearly 3.5 V is comparable to the redox potential of PTMA. The initial discharge capacity was 134 mAh at a current of 100 mA. Since the battery contained 2.0 g of PTMA, their specific capacity was estimated to be 67 Ah/kg. At the current of 1000 mA, the discharge capacity retained 77% of that found for the 100 mA measurement.

It was shown that organic radical battery has an ability as a practical rechargeable batteries with environmental friendly, high power density and high energy density.

References

[1] K. Nakahara, S. Iwasa, M. Satoh, Y. Morioka, J. Iriyama, M.Suguro and E. Hasegawa, *Chem. Phys. Lett.*, **359**. 351-354 (2002).
[2] K. Nakahara, S. Iwasa, J. Iriyama, Y. Morioka, M. Suguro and M. Satoh, *201st ECS. Meeting*, Philadelphia, 89 (2002).

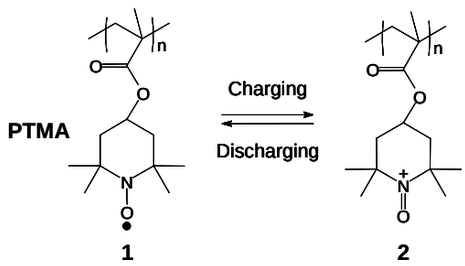

Figure 1 Electrochemical reaction of PTMA

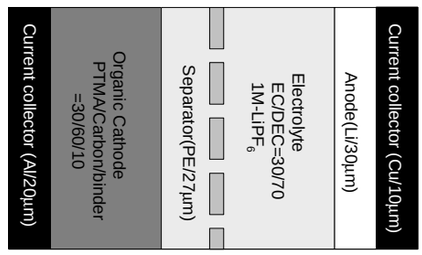

Figure 2 Schematic illustration of Organic radical batteries.

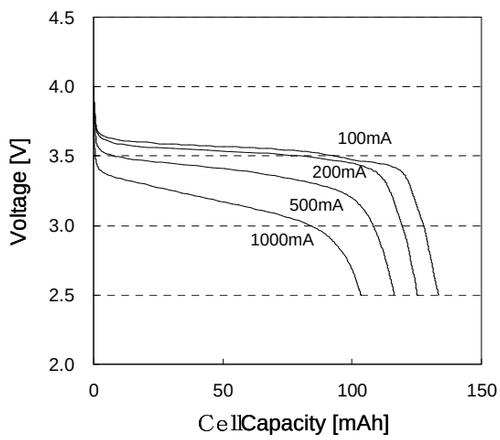

Figure 3 Discharging curves of Organic radical batteries at the discharge current of 100mA, 200mA, 500mA and 1000mA.(4.2V, constant current charging, 2.5V cutoff)