Electronic Supplementary Material (ESI) for Materials Advances.

Supplementary Information

Integration of Carbon Nanotubes and Azo-Coupled Redox-Active Polymers into Core-Shell Structured Cathodes with Favorable Lithium Storage

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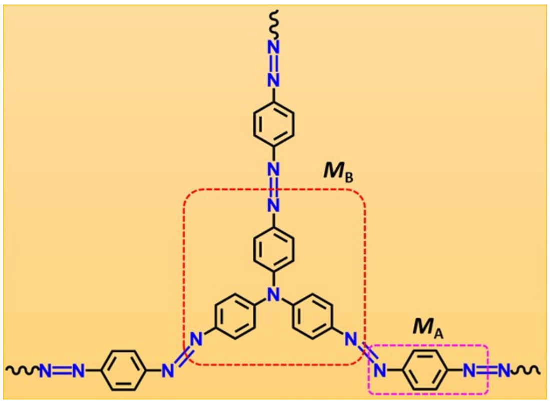
Theoretical capacity (C_{th}, mAh g^{-1}) of NHP moieties

The C_{th} was calculated on the basis of the literature method reported elsewhere [*Adv. Mater.* 2019, 31, 1901478]. The equation can be expressed as $C_{th} = F/(3.6 \times M_w)$, where F is the Faraday constant (*ca.* 96485 C mol^{-1}) which equals to $e \times N_A$ (e, the charge of electron, $1.60217662 \times 10^{-19}$ Coulombs; N_A, the Avogadro constant, 6.022141×10^{23} per mole). M_w is the equivalent molecular weight of organic active materials, and defined as molecular weights of the repeating unit cell divided with the number of electrons (n) involved. Aa shown in Scheme **S1**, the molecular weight of the repeating *p*-phenylenediamine (M_A) and tris(4-aminophenyl) amine (M_B) units in NHP are 104, and 284, respectively. Based on the equimolar reaction between amino groups from *p*-phenylenediamine and tris(4-aminophenyl) amine monomers, the number of M_A and M_B units is 3 and 2 in the repeating NHP unit cell, respectively. The molecular weight of a unit cell is thus calculated to be $M_{unit\ cell} = 3 \times M_A + 2 \times M_B = 3 \times 104 + 2 \times 284 = 880$. Furthermore, in such a unit cell, the number of electrons involved is 8 (including six azo groups and two tertiary amine sites). Accordingly, M_w is 110 by using the equation of $M_w = M_{unit\ cell}/n$ (880/8). As a result, the C_{th} is about 243.6 mAh g^{-1} using the above equation.



Scheme S1 Chemical structures of M_A and M_B units in the NHP moiety.

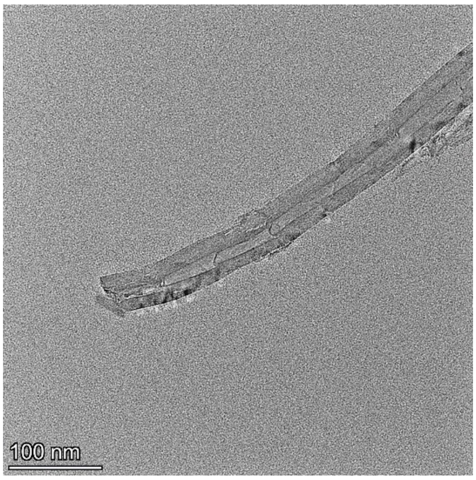


Fig. S1 Typical TEM image of bare CNTs

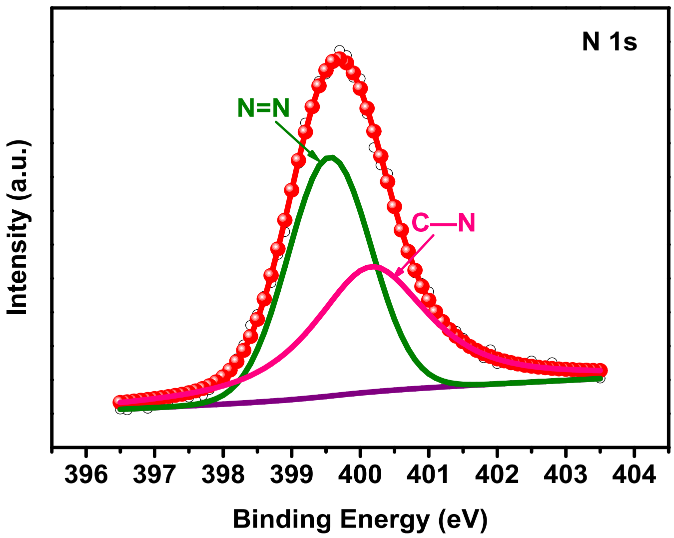


Fig. S2 High-resolution N1s XPS spectrum of NHP moieties.

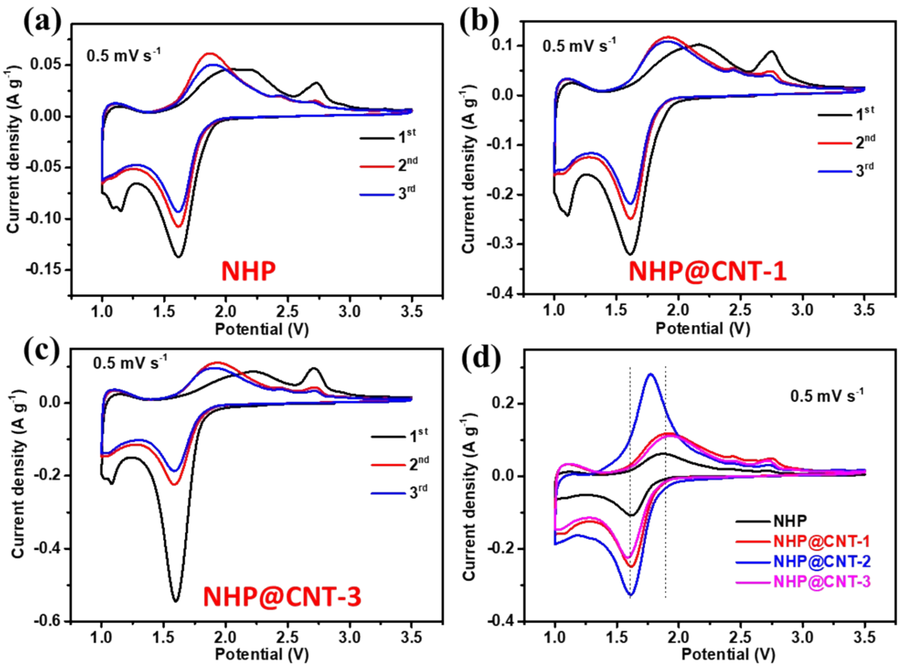


Fig. S3 CV cures of (a) NHP, (b) NHP@CNT-1, and (c) NHP@CNT-3 cathodes measured at 0.5 mV s^{-1} in the first three cycles; (d) stable CV profiles of NHP and NHP@CNTs cathodes at 0.5 mV s^{-1} in the third cycle.

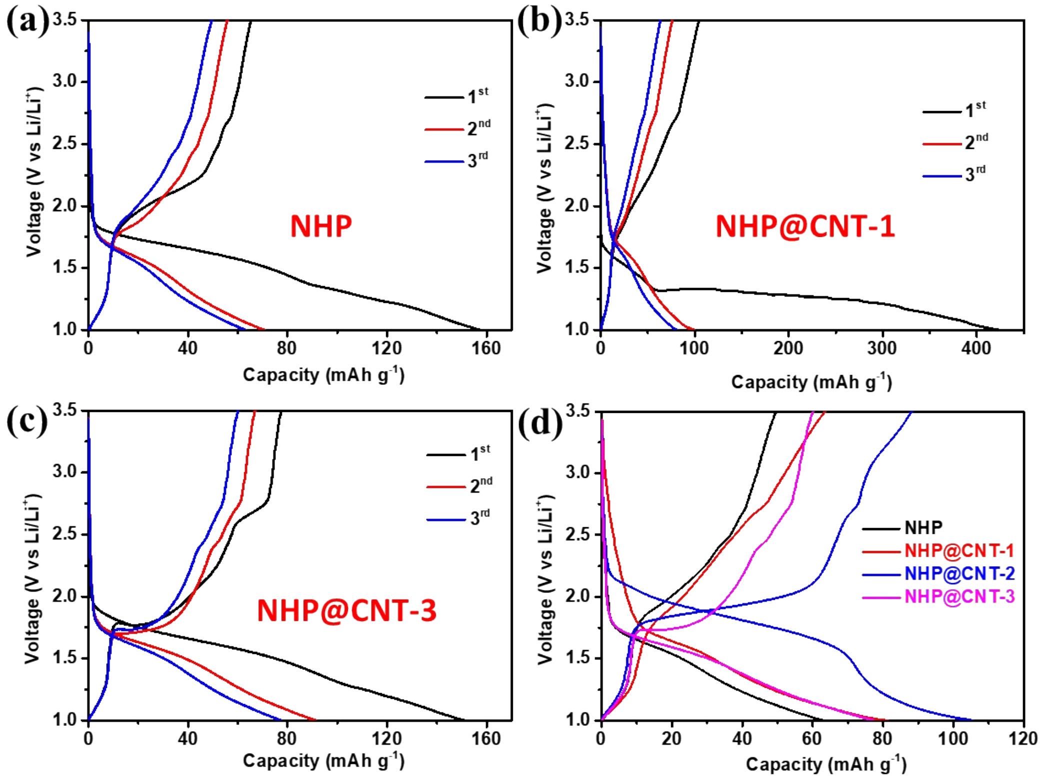


Fig. S4 Charge/discharge voltage profiles of (a) NHP, (b) NHP@CNT-1, and (c) NHP@CNT-3 cathodes measured at 0.05 A g^{-1} in the first three cycles; (d) stable charge/discharge voltage profiles of NHP and NHP@CNTs cathodes at 0.05 A g^{-1} in the third cycle.

Table S1. Elemental contents of NHP and NHP@CNTs obtained from elemental analysis

Sample	N (wt%)	C (wt%)	H (wt%)
Pure NHP	15.7	64.9	4.0
NHP@CNT-1	13.0	67.8	3.6
NHP@CNT-2	11.5	75.3	3.4
NHP@CNT-3	8.3	78.9	2.5
Pure CNTs	0.03	97.6	0.01