SUPPORTING INFORMATIONS

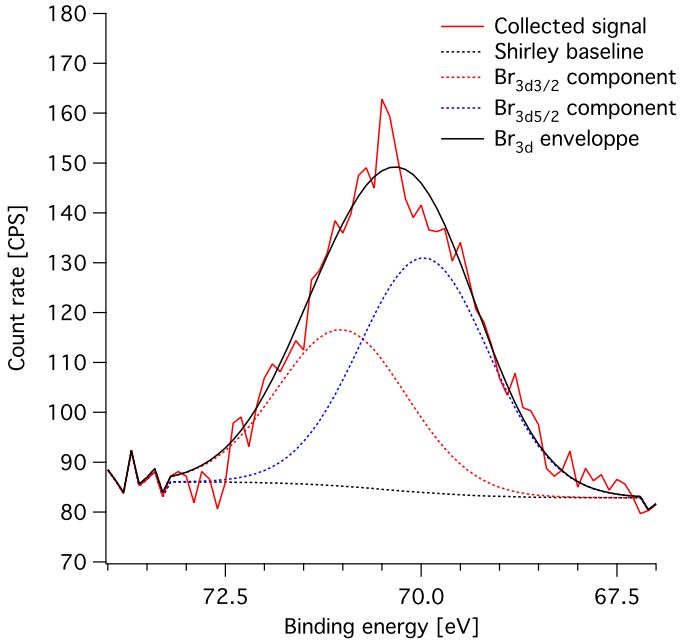

Figure S1. Br_{3d} narrow scan of the MWCNT-initiator.

Table S1. Experimental surface atomic concentrations for oxidized MWCNTs and MWCNT-initiator.

	Surface atomic concentrations (%)			
	C 1s	N 1s	O 1s	Br 3d
Oxidized MWCNTs	92.2	0.4	7.4	b.l.d.
MWCNT-initiator	90.1	0.6	8.9	0.2

b.l.d. =below limit of detection

TGA measurements after cycling

A cathode made of MWCNT-*g*-PTMA and pristine MWCNT (2 :1 ; m/m) was cycled until the capacity is the half of the one at the third cycle (500 cycles). At the end of the test, the battery is fully discharged. The cathode, soaked 3 times in AcCN during 1 week, is dried under reduced pressure. A TGA analysis is conducted.

The weight loss for the cathode is lower than the one for MWCNT-*g*-PTMA due to the presence of pristine MWCNT, introduced as the first layer during the fabrication process of the electrode. The weight loss between 200 and 450°C after cycling is 21.0%. Taking into account the mass of the materials in the cathode and the weight loss for the starting material MWCNT-*g*-PTMA (30.5 wt.%), the weight loss between 200 and 450°C can be calculated. This weight loss should be 2/3 of the MWCNT-*g*-PTMA weight loss, i.e. 20.3 wt.% (Degradation of pristine MWCNT is considered as negligible). This value is close to the one experimentally found after cycling.

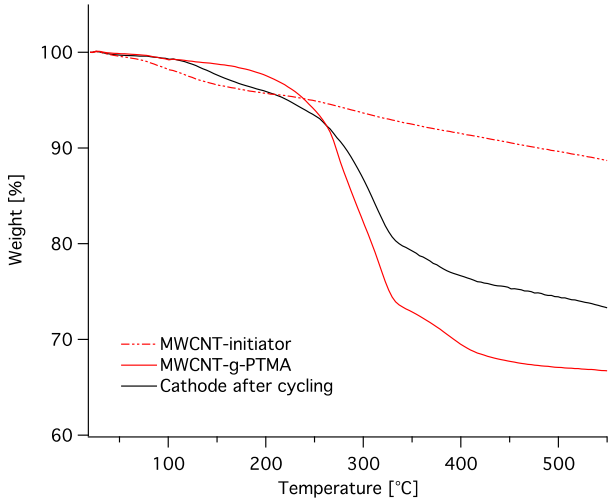

Figure S2. Thermograms of the MWCNT-initiator, MWCNT-*g*-PTMA and the cathode after cycling.

Untethered PTMA-based control electrodes

A physical blend of untethered PTMA and pristine MWCNTs has been prepared in order to conduct electrochemical measurements as control data. For this purpose, a linear PTMA homopolymer (degree of polymerization = 69) was synthetized through ATRP. The control composite measurements are compared to the results obtained for the MWCNT-*g*-PTMA electrodes.

The cathodes were prepared by blending the active material PTMA, MWCNTs (Nanocyl NC7000) and poly(vinylidene fluoride) (PVdF) binder. The blend has the following composition, 60 wt.% of PTMA, 30 wt.% of MWCNTs and 10 wt.% of PVdF binder. 5 mL of *N*-methylpyrorolidone were added per 1 g of solid materials. The ingredients were mixed uniformly at room temperature for 12 hours to yield the slurry. The slurry was cast on an aluminum foil using the doctor blade method (the thickness of the wet film was controlled to be 45 µm) and dried at 60 °C for 12 h. Circular discs of diameter 1 cm were cut and used as cathodes for evaluating the electrochemical properties. The electrodes were assembled following the same protocol as for the MWCNT-*g*-PTMA cathodes. The cyclic voltammogram of the control composite was recorded with a CHI1660B instrument. All others electrochemical measurements on the control composite were conducted with the same devices, apparatus and parameters as for the MWCNT-*g*-PTMA cathodes. 4 points conductivity measurements were obtained with the control composite coated on an insulate substrate. This coating was prepared with the same doctor blade protocol as for the control electrode but cast on a glass slide. The thickness of the coated material was found to be 30 µm (measured with a Draper Expert digital micrometer).

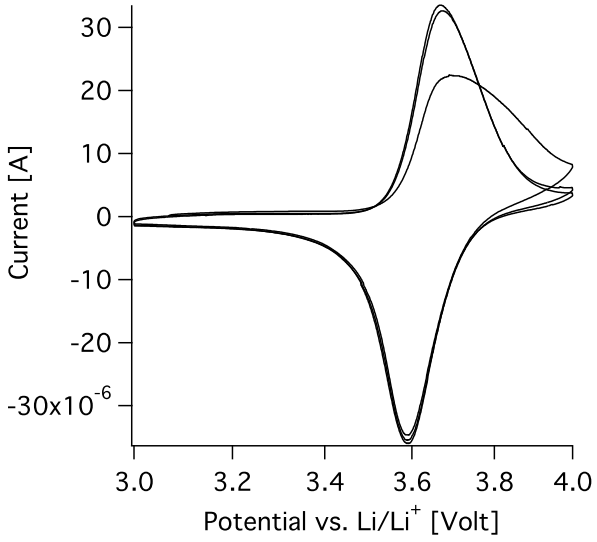

Figure S3. Cyclic voltammogram of the PTMA-based control electrode at 0.1 $mV.s^{-1}$ (EC/DEC/DMC 1:1:1 in v:v:v ; 1 M $LiPF_6$). The 3 first cycles are depicted.

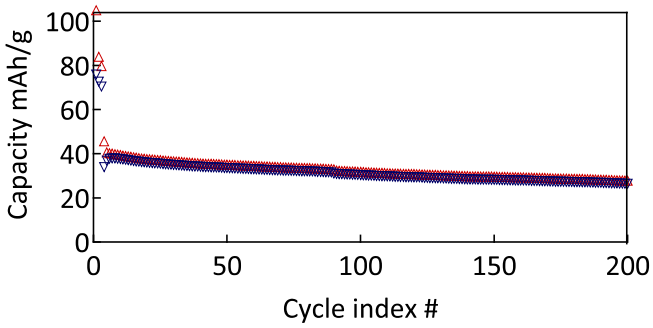

Figure S4. Cycle stability of the PTMA-based control electrode at 2C rate over 200 cycles (The 3 first cycles were recorded at C/6 rate).

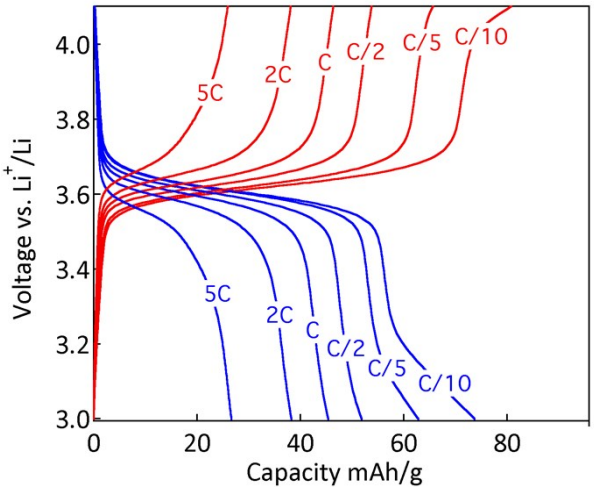

Figure S5. Charge-discharge curves of PTMA-based control cathode (60 wt.% PTMA) at various C rates (EC/DEC/DMC 1:1:1 in volume ; 1 M $LiPF_6$).