**Introduction**

Hard carbon (HC) has been extensively investigated as anode materials for high-energy lithium-ion batteries owing to its high capacity, long cycle life, good rate capability, and low cost of production. However, it suffers from a large irreversible capacity and thus low initial coulombic efficiency (ICE), which hinders its commercial use.

In this work, a prelithiation of the HC anode method was developed based on a chemical reaction using a lithium-containing reagent. The prelithiation extent can be easily controlled by tuning the reaction time.

With the prelithiated HC anode, the full cell exhibits a much improved ICE and cycling performance than those of the pristine full cell. This facile prelithiation method is proved to be a practical solution for the commercial application of hard carbon materials.

![Schematic illustration of the influence of prelithiation on the first cycle of an LIB.](image)

**Experiment**

**Anode:** A slurry of HC powder, super C65, and poly(vinyl difluoride) (PVDF) at a weight ratio of 94:3:3 was coated onto a carbon-coated copper foil and vacuum-dried at 120 °C overnight.

**Prelithiation Reagent:** A certain amount of biphenyl (Bp) was dissolved in tetrahydrofuran (THF) to make a 1 M Bp-THF solution. Then, an excess amount of Li metal was added in the solution and reacted overnight to prepare a 1M Li-Bp-THF solution.

**Prelithiation:** The HC anode was immersed in the prelithiation reagent for controlled times. This work includes 30 s, 2 min, 10 min, and 1 h. Then, the anode was washed by THF for three times.

**Cell:** The 2032 coin cells were made as half cell and full cell in an argon-filled glovebox. The full cell was assembled by layering base, spring, 0.5 mm spacer, prelithiated HC anode, electrolyte, separator, NCM111 cathode, 1 mm spacer and lid.

**Testing:** Charging/discharging tests were conducted at room temperature. The voltage window for full cell test was 2.5–4.1 V. After 5 cycles formation, the cell was tested for 200 cycles at 1C charge/discharge rate. The prelithiation mechanism was investigated by conducting X-ray photoelectron spectra (XPS) and electrochemical impedance spectroscopy (EIS) on half cells.

**Results**

![EIS Nyquist plots of (a) pristine HC; (b) PreLi-30s HC; (c) PreLi-2min HC electrodes.](image)

**Conclusions**

- Prelithiation of HC anode improves the ICE of the full cell at least 19.5% compared with the pristine full cell.
- Prelithiation improves the cycling performance of the full cell at least 26.2% compared with the pristine full cell.
- The prelithiation capacity increases as the prelithiation time increasing. However, 30 s prelithiation significantly improve the performance.
- The chemical prelithiation might be beneficial to the homogeneous SEI formation and thus cause the good cycling performance of the full cell.

**Future works**

- Practical prelithiation on pouch cell.
- Mechanism study on prelithiation SEI formation.
- Prelithiation on other anode materials such as silicon.
- Prelithiated anode matching high energy density cathode such as sulfur.

**References**