Reactive Polyacrylate Binders for Silicon Anodes in Lithium-Ion Batteries

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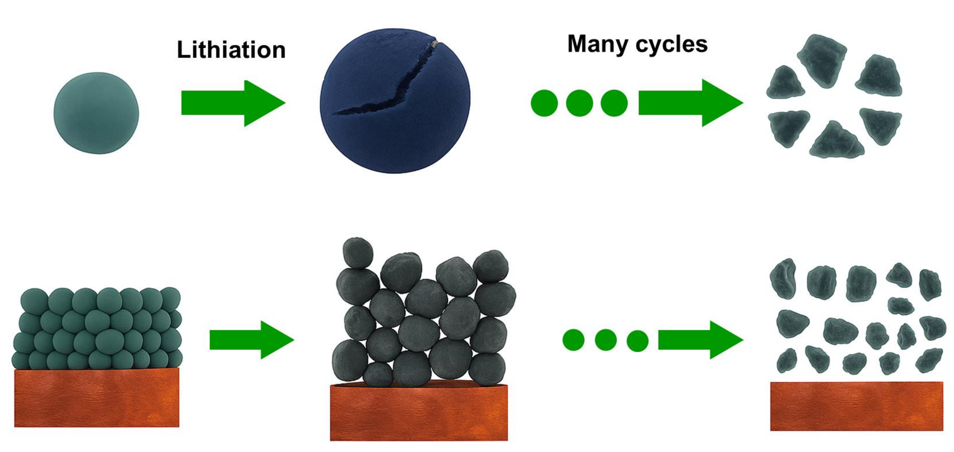
Abstract

Poly(acrylic acid) (PAA) is the most established binder for pure Si electrodes, primarily relying on abundant polar carboxylic groups that enable strong hydrogen bonding with the Si surface. Despite these advantageous properties, the cycling performance of the fabricated Si anodes still far from meeting the standards required for practical applications.

Herein, we propose a reactive polyacrylate binder that can accommodate the volumetric expansion of Si anodes. The binder consists of a uniform mixture of PAA and polyacrylate crosslinker, which forms a network structure upon curing at high temperatures. In this network, PAA effectively maintains the integrity of the electrode through strong hydrogen bonding with Si particles, while crosslinking with polyacrylate crosslinker chains simultaneously enhances its mechanical strength. The elasticity of the binder was carefully adjusted by controlling the ratio of monomers in the polyacrylate crosslinker to effectively accommodate Si volume changes. As a result, the binder demonstrates enhanced cycling stability and rate performance compared to conventional PAA-based binders.

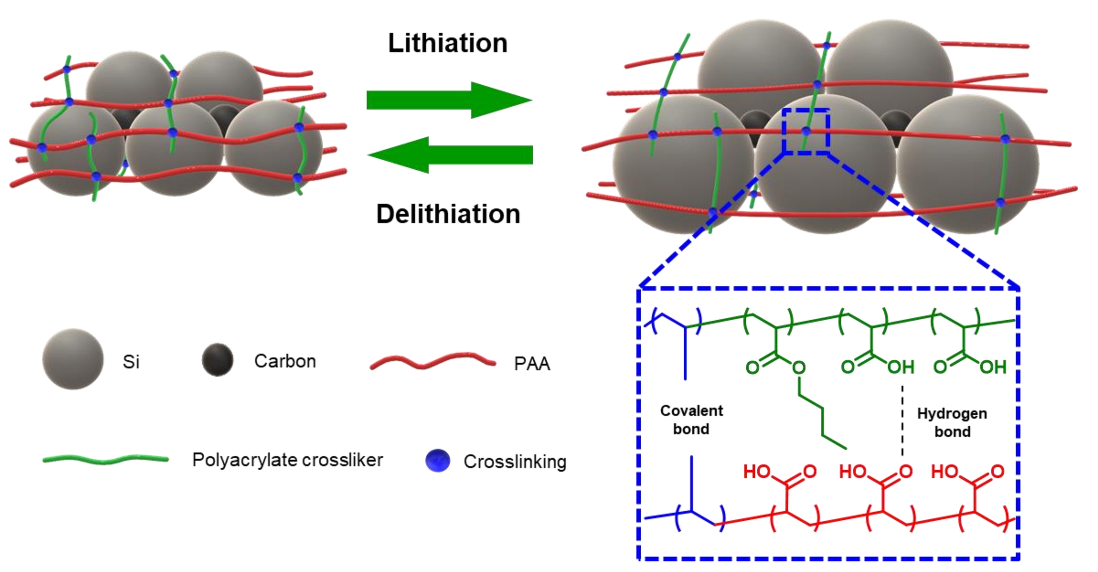
Introduction

- ***Silicon anode charging and discharging***
- *Over 300% volume change*
- *Si layer destruction and thickening*
- *Necessity of research on battery anode binder*



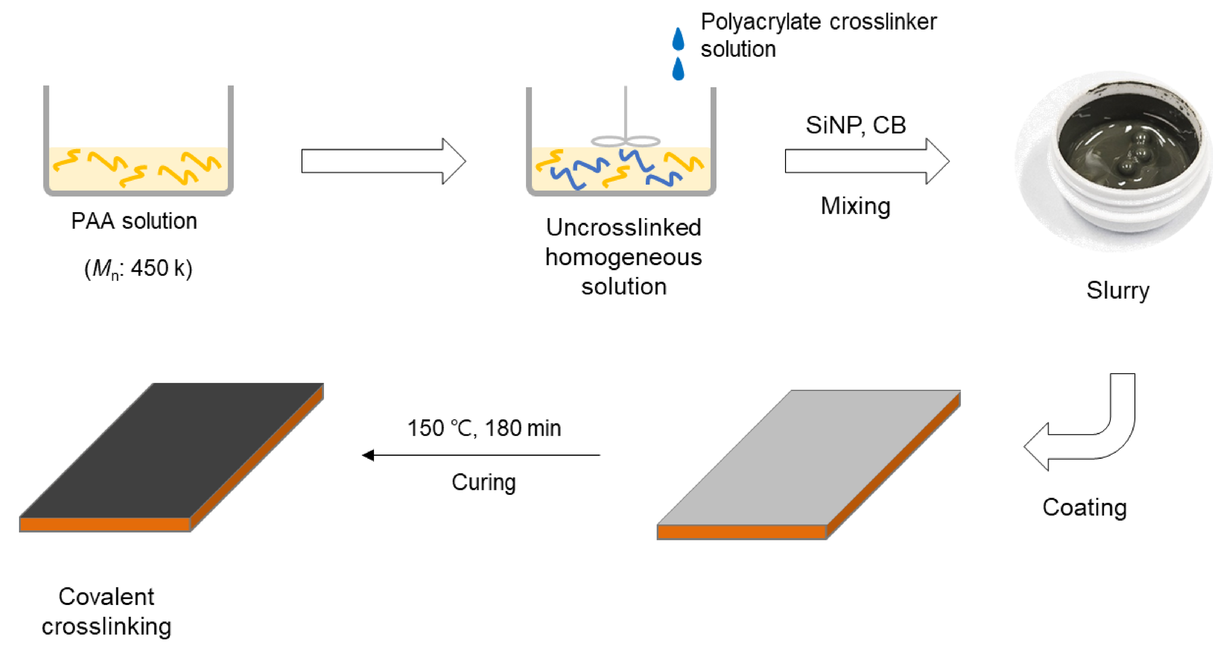
Research Goal

- ***Design of binders capable of accommodating volume changes of silicon during lithiation and delithiation***
- *PAA crosslinking with polyacrylate crosslinker chains*
- *Controlling ratio of monomers in the polyacrylate crosslinker to effectively accommodate Si volume changes*



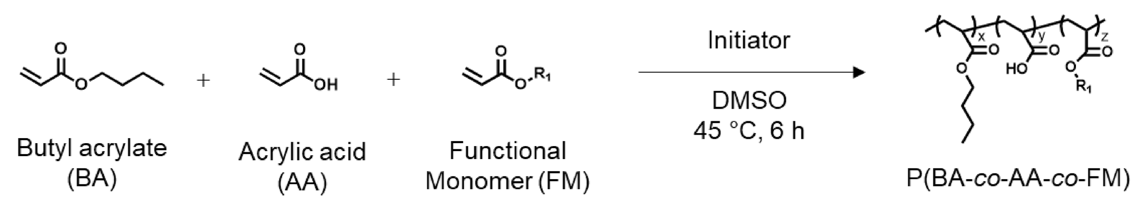
Procedure

- ***Electrode preparation***
- *SiNP:CB:Binder (weight ratio) = 70:15:15*
- *Solvent = DMSO*



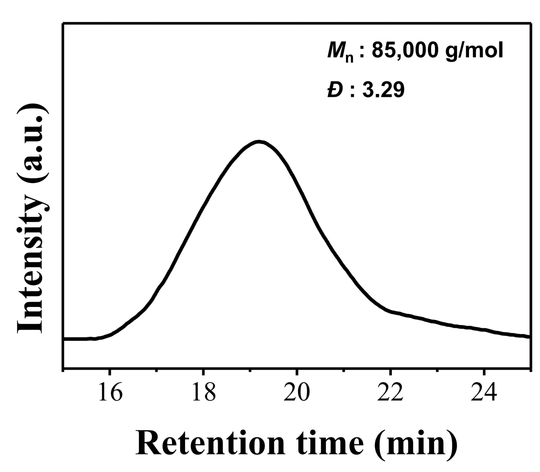
Result & Discussion

Synthesis of polyacrylate crosslinker

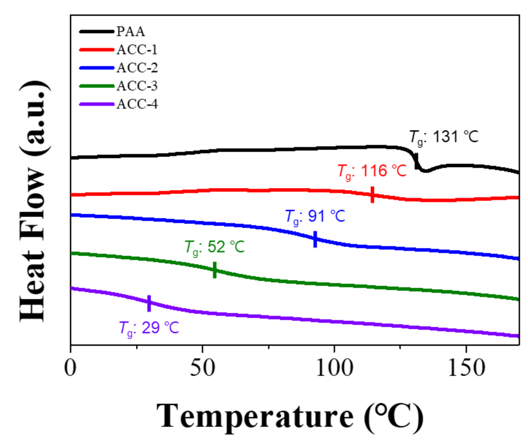


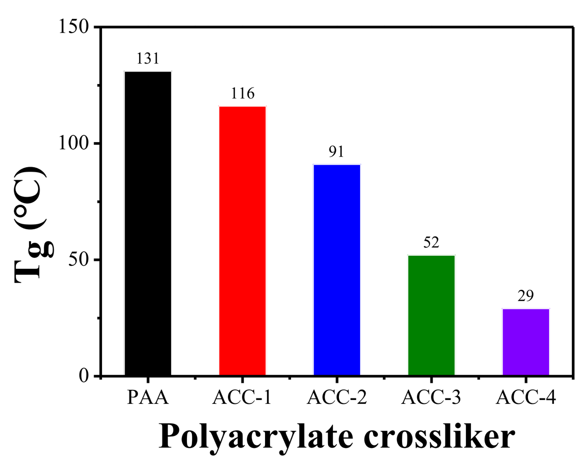
Sample name	Functional monomer (mol%)	BA (mol%)	AA (mol%)
ACC-1	5	15	80
ACC-2	5	25	70
ACC-3	5	35	60
ACC-4	5	45	50

THF GPC



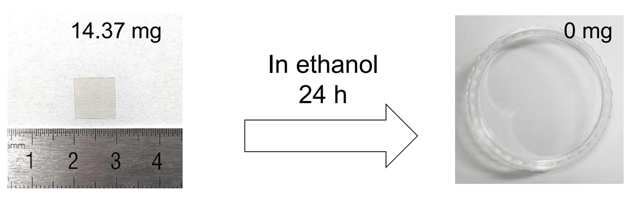
DSC



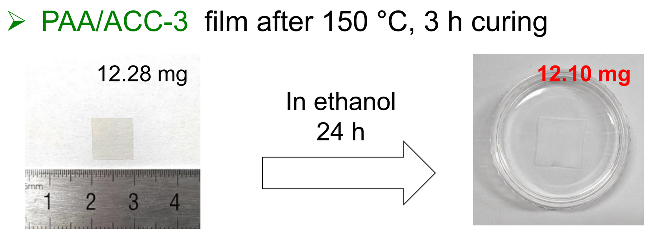


Gel contents test

- PAA film after 150 °C, 3 h



- PAA/ACC-3 film after 150 °C, 3 h curing



Mechanical properties of electrodes

Nanoindentaion

$H_{IT} = \frac{P_{max}}{A}$

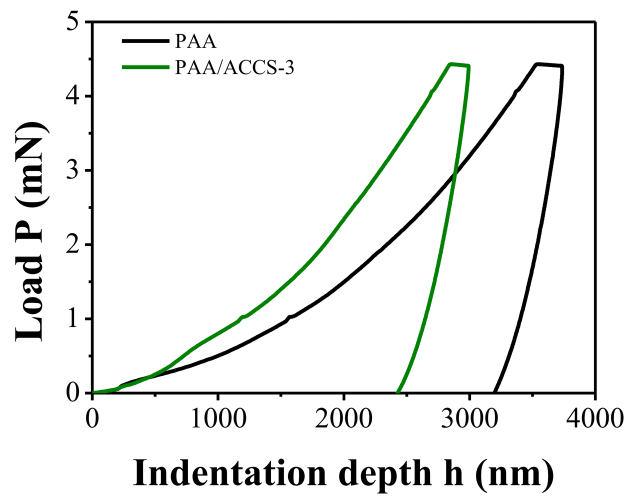
*P_{max} : Maximum load

*A : Residual indentation area

$nIT = \frac{W_{elastic}}{W_{total}} \times 100\%$

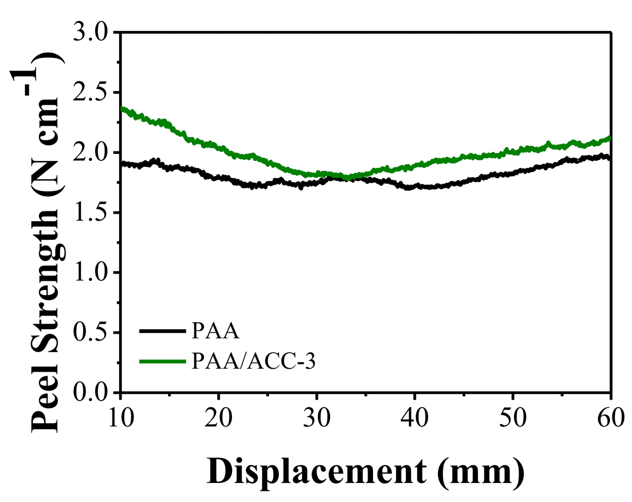
*$W_{elastic}$: Unloading curve area

*W_{total} : Loading curve area



- PAA electrode
  - HIT: 13.94 MPa
  - nIT: 14.04%
- PAA/ACC-3 electrode
  - HIT: 23.38 MPa
  - nIT: 19.02%

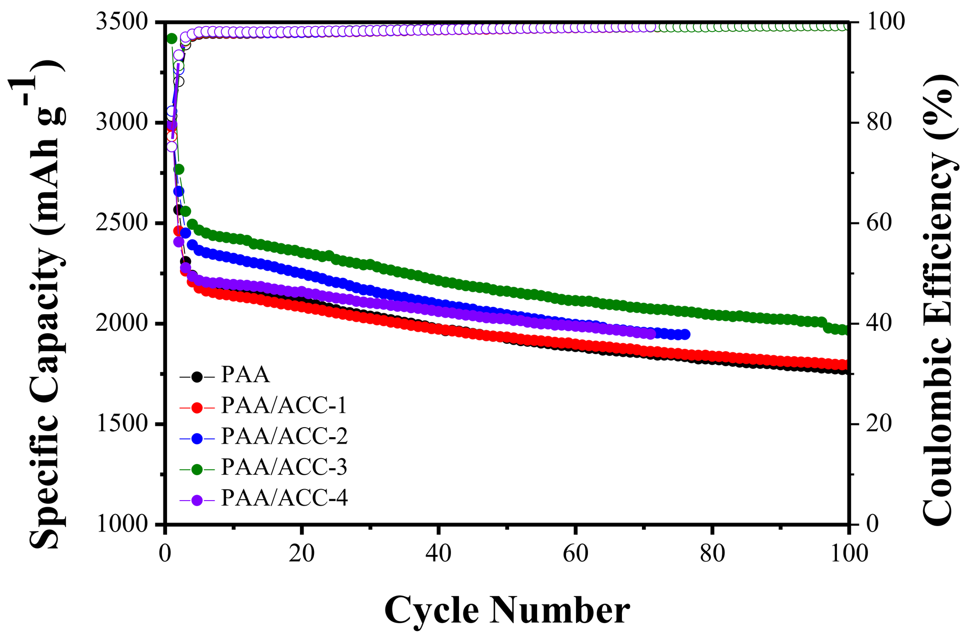
Peel test



- PAA electrode
  - Peel strength: 1.81 N/cm
- PAA/ACC-3 electrode
  - Peel strength: 1.98 N/cm

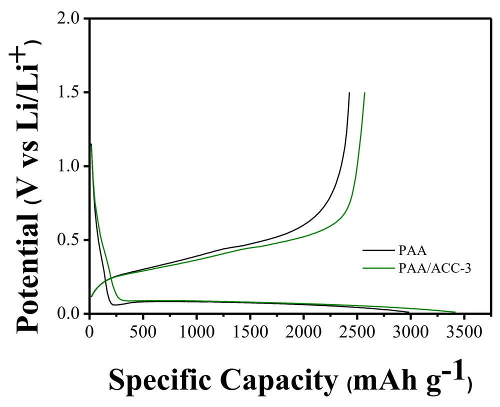
Electrical properties of electrodes

Cycling test

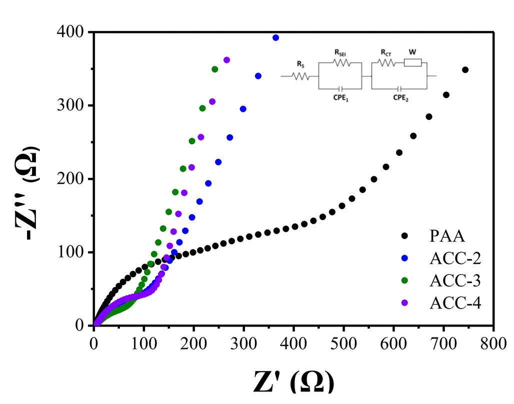


- PAA < PAA/ACC-1 < PAA/ACC-4 < PAA/ACC-2 < PAA/ACC-3
- Incomplete dissolution of ACCS-4 in DMSO due to high BA content

Discharge−charge curves



EIS test



- Higher reversible capacity (3418.5 mA h g^{-1}) compared to that of PAA electrode (2985.7 mA h g^{-1})
- As T_g decreases, charge transfer resistance decreases.

Conclusion

- ✓ Polyacrylate crosslinkers with different glass transition temperatures (T_g) were successfully synthesized by adjusting monomer ratio.
- ✓ The synthesized crosslinker was crosslinked with PAA at high temperature, showing a gel content of 98.05% with superior hardness and elasticity compared to PAA alone.
- ✓ Binder composed of PAA and polyacrylate crosslinker exhibited improved cycling performance and lower charge transfer resistance as T_g decreased.

Acknowledgements

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