



Title: Molecularly engineered siloxane binders: Elevating LFP cathode efficiency under

high active mass loading, Name: Celik-Kucuk Asuman*, Takeshi Abe

University/Organization: Kyoto University, JAPAN

Abstract: Previously, we demonstrated that siloxane structures (Sx@04 and Sx#O@06) mitigate the corrosive effects of LiTFSI by forming a protective layer on aluminum current collectors, enhancing lithium stability and battery performance (Journal of Power Sources 556 (2023) 232520). Our recent research revealed that siloxane-based polymers (Sx@04) used as binders in LiFePO₄ (LFP) cathodes significantly improve rate capability and cycling stability compared to traditional binders like PVDF and PEO (Journal of Power Sources 581 (2023) 233478). Building on these findings, we focused on modifying siloxane-based binders (Sx#O@32) to further improve their performance in LFP cathode applications. Testing at 60°C showed that LFP cathodes with Sx#O@32 had superior cyclic stability at 0.5 C, outperforming both Sx@04 and PVDF. Even at high mass loadings, Sx#O@32 maintained better cycling stability than PVDF. Additionally, the Sx#O@32 binder reduced ionic diffusion resistance (Rp) and charge transfer resistance (Rct), facilitating smoother lithiation and delithiation during battery operation. This enhanced performance is attributed to the low internal resistance of the composite electrodes using Sx#O@32. The stronger adhesion observed in these electrodes is likely due to increased cohesion from network formation via anion solvation of low molecular weight siloxane oligomers, enhancing performance over Sx@04 and PVDF binders.

Biography: I hold dual PhDs in applied chemistry from Tohoku University (MEXT scholarship) and polymer chemistry from Gebze Technical University. With a strong background in organic and inorganic polymeric materials, I specialize in the design and application of advanced hybrid materials for electrochemical devices, including rechargeable batteries and fuel cells. My career includes work as an assistant professor at Marmara University and a visiting researcher at Kyoto University, contributing to significant projects. I have authored 37 journal articles, hold two patents, and received prestigious awards such as the L'Oréal–UNESCO National Fellowship and a Hirose Foundation fellowship.

Supplemental Document

Half-cell performance of LFP at 60 °C, 0.5C rate with different LFP loading for Sx@04, Sx#O@32, and PVDF binders. 1M LiTFSI/PC was used as electrolyte for all systems.

| Binder | Active mass loading | Retention@ cycles | 1 st charge/discharge |
|---------|--|-------------------|----------------------------------|
| Sx@04 | 3.9 mg/cm ² , 190 μA/cm ² | 85% @ 120 | 157/157 |
| Sx@04 | 6.7 mg/cm ² , 326 μA/cm ² | 76% @ 60 | 163/162 |
| Sx@04 | 8.8 mg/cm ² , 428 μA/cm ² | 79% @ 60 | 163/163 |
| PVDF | 4.1 mg/cm ² , 198 μA/cm ² | 80% @ 90 | 152/150 |
| PVDF | 7.5 mg/cm ² , 356 μA/cm ² | 77% @ 113 | 158/158 |
| PVDF | 11.1 mg/cm ² , 538 μA/cm ² | 69% @ 113 | 161/159 |
| Sx#O@32 | 3.2 mg/cm ² , 154μA/cm ² | 76% @ 100 | 151/146 |
| Sx#O@32 | 8 mg/cm ² , 386μA /cm ² | 75% @ 100 | 157/157 |
| Sx#O@32 | 9.9 mg/cm ² , 481µA /cm ² | 69% @ 100 | 161/158 |
| Sx#O@32 | 11.7 mg/cm ² , 566μA /cm ² | 71% @ 100 | 162/164 |

