

Air Stabilization of $\text{Li}_7\text{P}_3\text{S}_{11}$ Solid State Electrolytes through Laser-Based Processing

Yannick Eatmon^a, Joseph W. Stiles^b, Shuichiro Hayashi^c, Marco Rupp^d, Craig Arnold^{d,e*}

^aDepartment of Chemical and Biological Engineering, Princeton University, Princeton, NJ 08544, USA

^bDepartment of Chemistry, Princeton University, Princeton, NJ 08544, USA

^cSchool of Integrated Design Engineering, Keio University, Yokohama, Kanagawa, Japan

^dDepartment of Mechanical & Aerospace Engineering, Princeton University, Princeton, NJ 08544, USA

^ePrinceton Materials Institute, Princeton University, Princeton, NJ 08544, USA

Supplementary Information

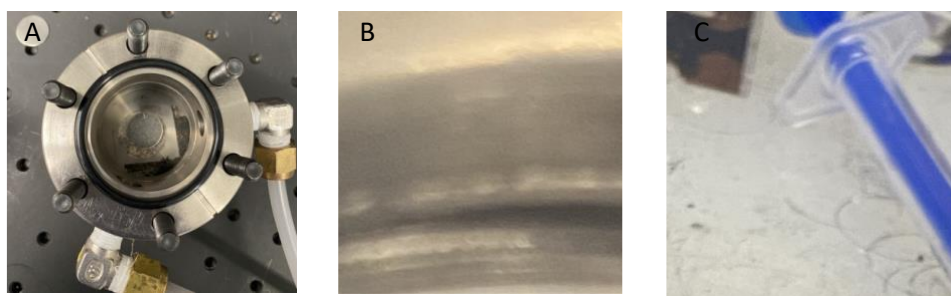


Figure S1. (A) Image of portable chamber used for laser processing electrolyte pellets while under Argon atmosphere. (B) Image of a pellet after being laser processed in two perpendicular directions. (C) Image of a laser processed pellet being used to make a symmetric (coin) cell.

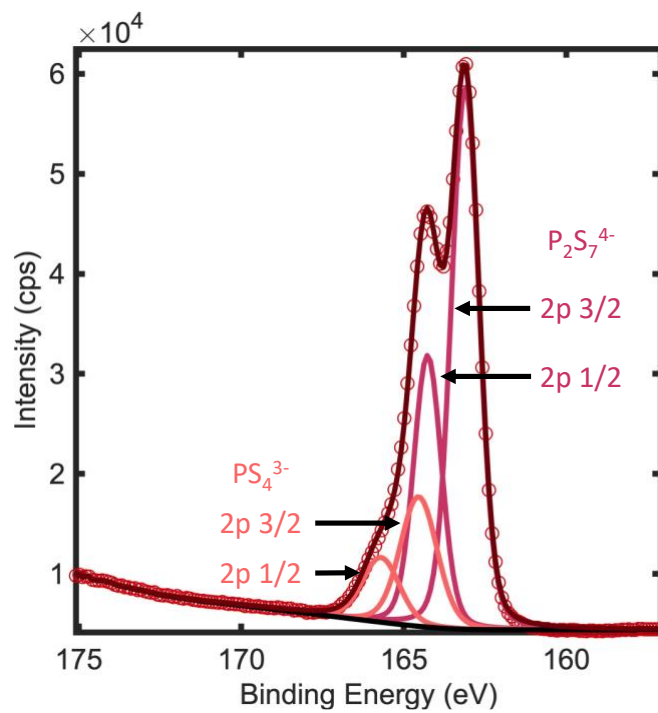


Figure S2. Deconvoluted X-ray Photoelectron Spectroscopy spectra of the Sulfur 2p region of $\text{Li}_7\text{P}_3\text{S}_{11}$ SSE samples. Figure shows the Sulfur $2p_{3/2}$ and $1/2$ orbitals.

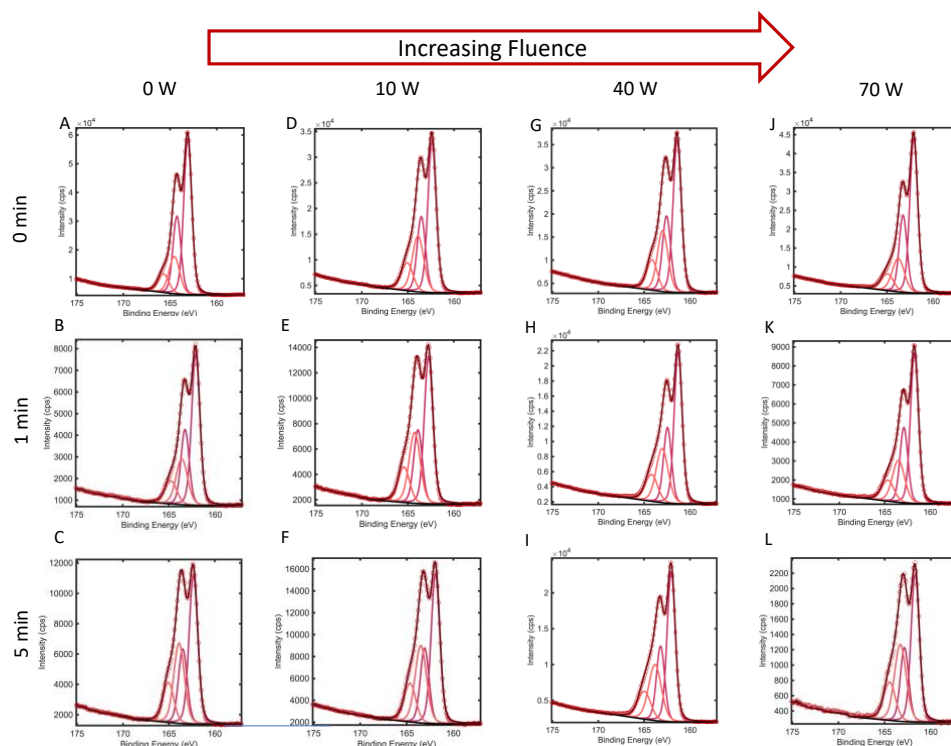


Figure S3. Deconvoluted X-ray Photoelectron Spectroscopy spectra of the Sulfur 2p region of $\text{Li}_7\text{P}_3\text{S}_{11}$ SSE samples. Spectra show the evolution of binding energies as a function of laser processing power and exposure time in air. (A-C) Unprocessed at (A) 0 min, (B) 1 min and (C) 5 min. (D-F) 10W at (D) 0 min, (E) 1 min and (F) 5 min. (G-I) 40W at (G) 0 min, (H) 1 min and (I) 5 min. (J-L) 70W at (J) 0 min, (K) 1 min and (L) 5 min.

	0 W	10 W	40 W	70 W
0 min	24.1%	33.8%	34.0%	24.8%
1 min	29.9%	38.6%	35.2%	29.9%
5 min	39.6%	40.4%	36.8%	42.1%

Table S1. Relative percentage of PS_4^{3-} in laser-processed samples as a function of effective laser power and exposure time in air. Relative percentages were calculated based on area under the curve calculations from deconvoluted S 2p XPS spectra (Figure S3). *Seemingly high initial concentration of $\text{P}_2\text{S}_7^{4-}$ is attributed to $\text{P}_2\text{S}_6^{4-}$ production from high power laser treatment and the overlapping peaks for $\text{P}_2\text{S}_7^{4-}$ and $\text{P}_2\text{S}_6^{4-}$.

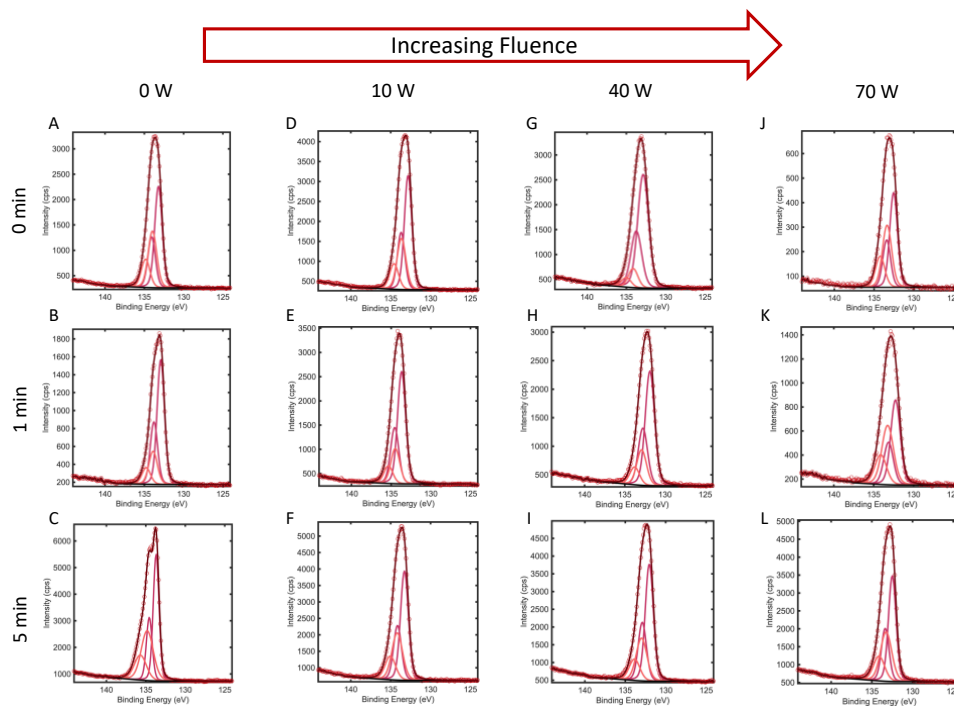


Figure S4. Deconvoluted X-ray Photoelectron Spectroscopy spectra of the Phosphorous 2p region of $\text{Li}_7\text{P}_3\text{S}_{11}$ SSE samples. Spectra show the evolution of binding energies as a function of laser processing power and exposure time in air. (A-C) Unprocessed at (A) 0 min, (B) 1 min and (C) 5 min. (D-F) 10W at (D) 0 min, (E) 1 min and (F) 5 min. (G-I) 40W at (G) 0 min, (H) 1 min and (I) 5 min. (J-L) 70W at (J) 0 min, (K) 1 min and (L) 5 min.

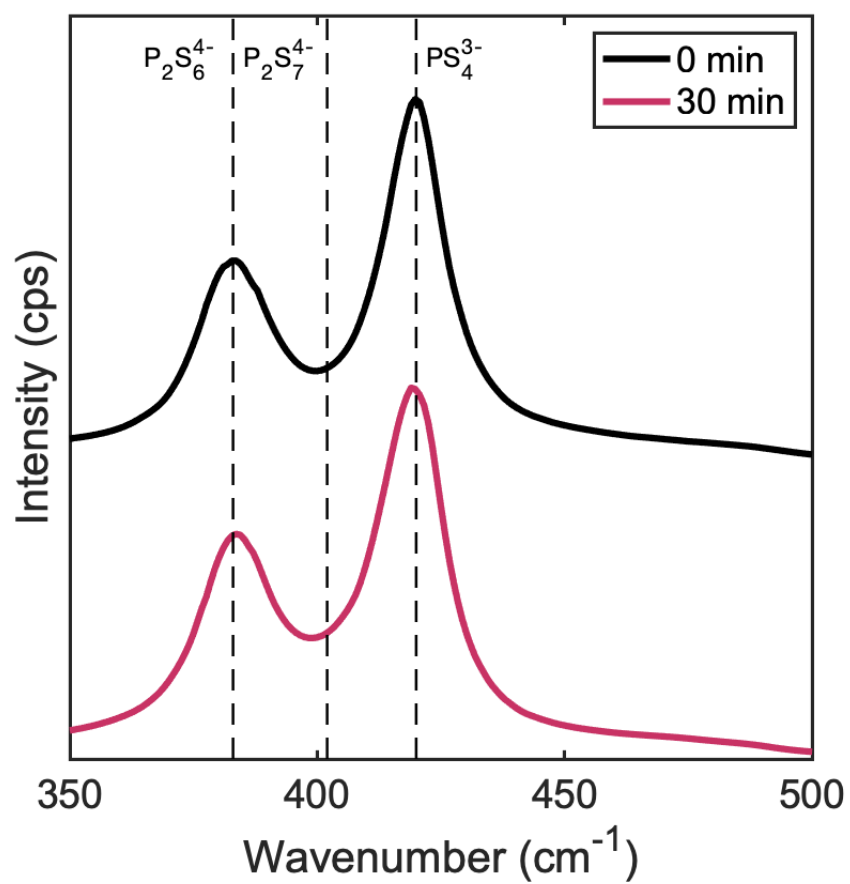


Figure S5. Raman Spectra of $\text{Li}_7\text{P}_3\text{S}_{11}$ pellets as a function of exposure time in air. Samples were processed at a laser power of 25W with a scanning speed of 200mm/s and a 200 μm hatch spacing.