Tape-casting of NASICON-based solid state battery

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Motivation

- High pressures needed for cycling a solid-state battery.
- Contact loss due to swelling of active cathode material during cycling.
- One approach is to create porous electrolyte scaffold which could be infiltritated with active electrode material.

Choice of electrolyte material

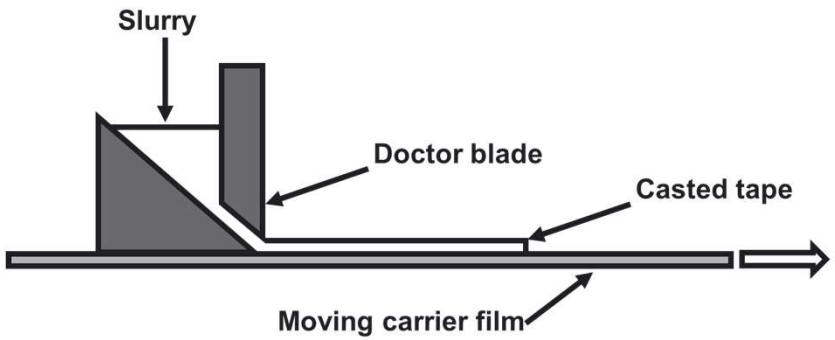
- Argyrodite (Li_6PS_5X) and LLZO ($Li_7La_3Zr_2O_{12}$) are two main model systems within SOLBAT project.

- NASICON composition $Na_{3+x}Zr_2Si_{2+x}P_{1-x}O_{12}$ (NZSP) was chosen as starting material due to:
  - Better processability for obtaining dense structures
  - Some experience from previous work done in St Andrews

- Optimized processing routes will be adapted for the target materials once the powders are available with sufficient quality and quantities.

Tape-casting technique

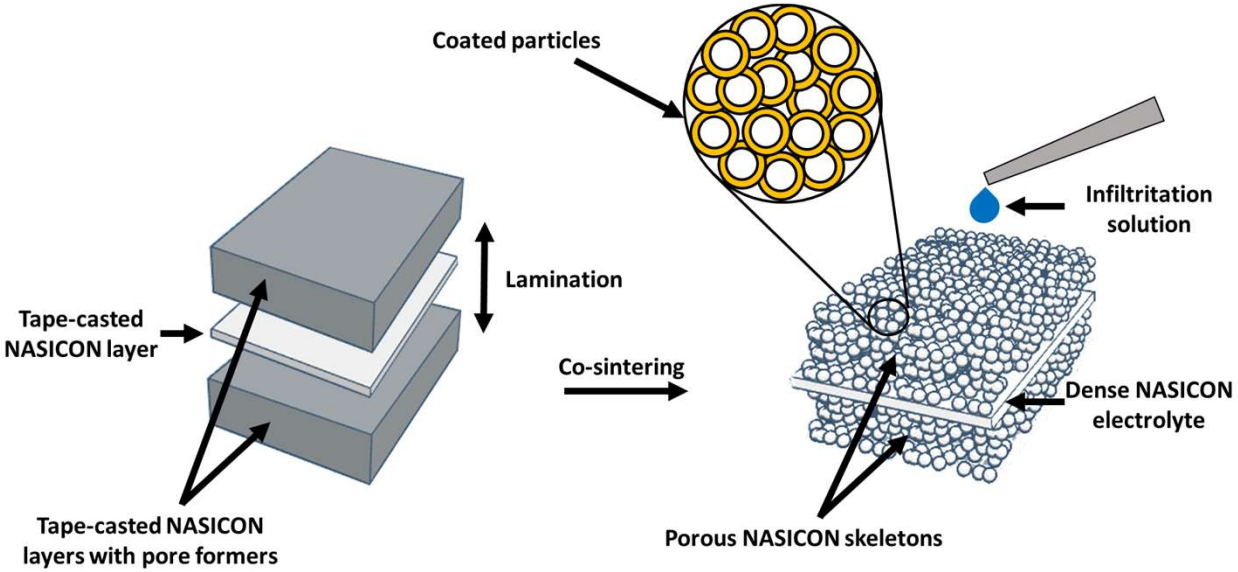
- Tape-casting technique can be easily scaled up.
- Well established cost-effective method for fabricating thin ceramic layers.
- Controlled layer thickness
- Good candidate method for providing thin electrolyte layers with sufficient conductance



Casting multilayer structures

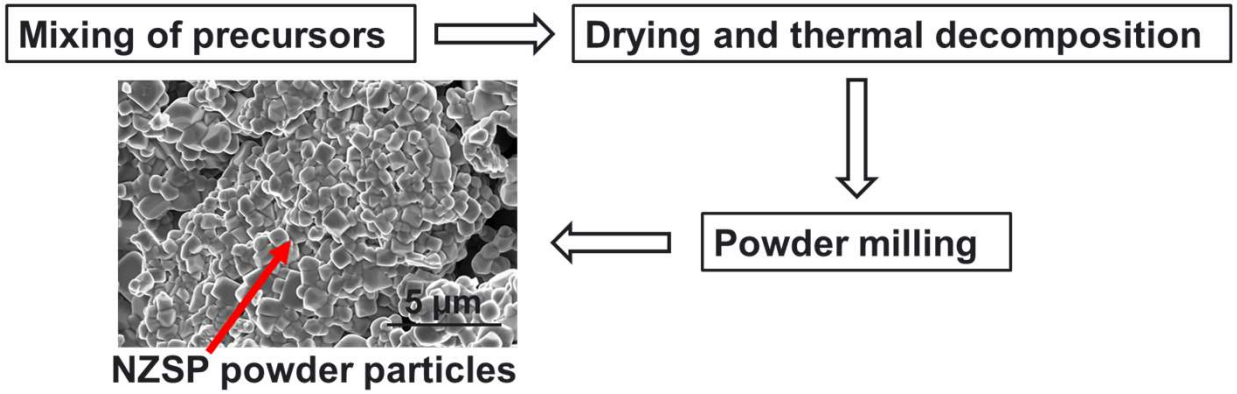
- Tape-casting technique is used to prepare multilayer NZSP structures.

- Porous NZSP scaffold will be impregnated with active material.



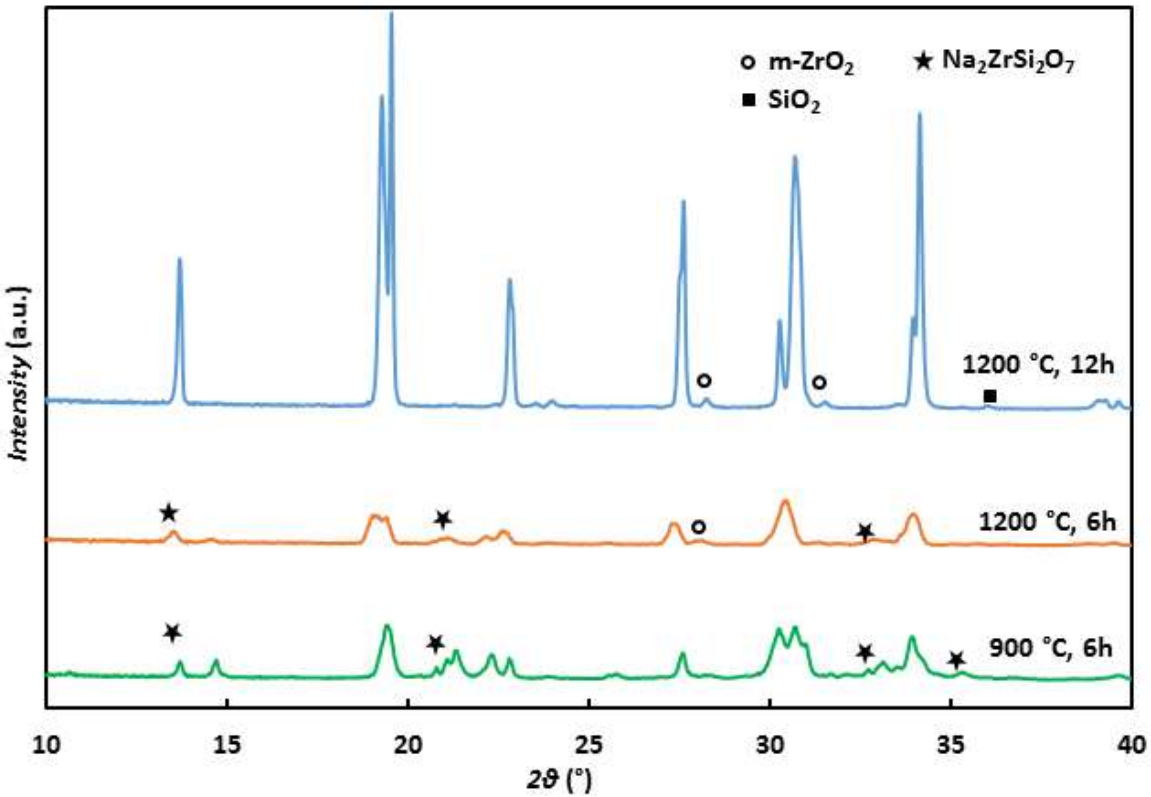
Powder synthesis

- Solid-state reaction procedure is used for synthesis of NASICON electrolyte material with composition of $Na_{3.5}Zr_2Si_{2.5}P_{0.5}O_{12}$ (NZSP0.5).
- Mixture of soluble precursor compounds provides homogeneous NZSP powder.

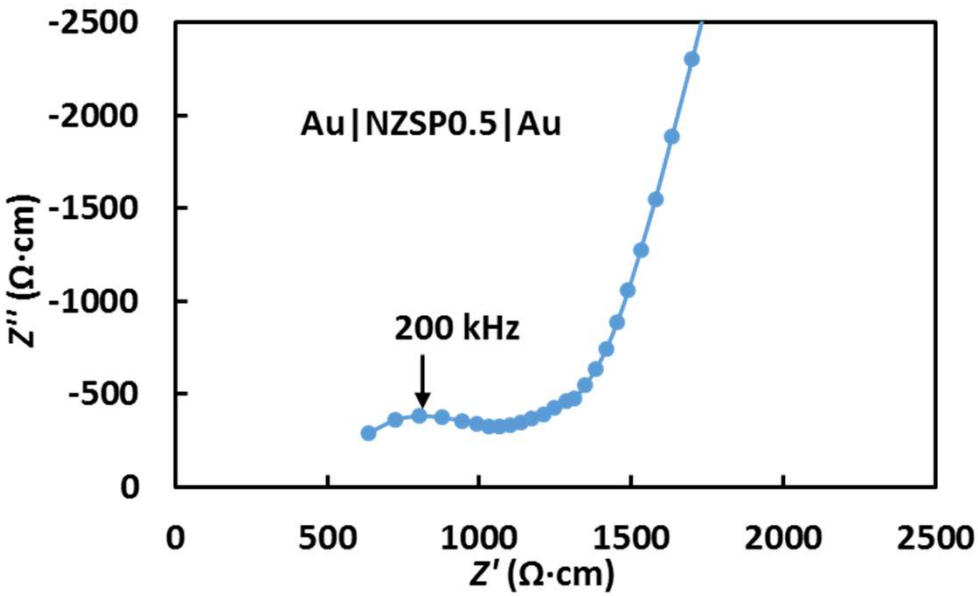


Phase purity

- Well crystallized sample was obtained after 1200 °C.
- Minor impurities
- Monoclinic zirconia is typical secondary phase for this material.



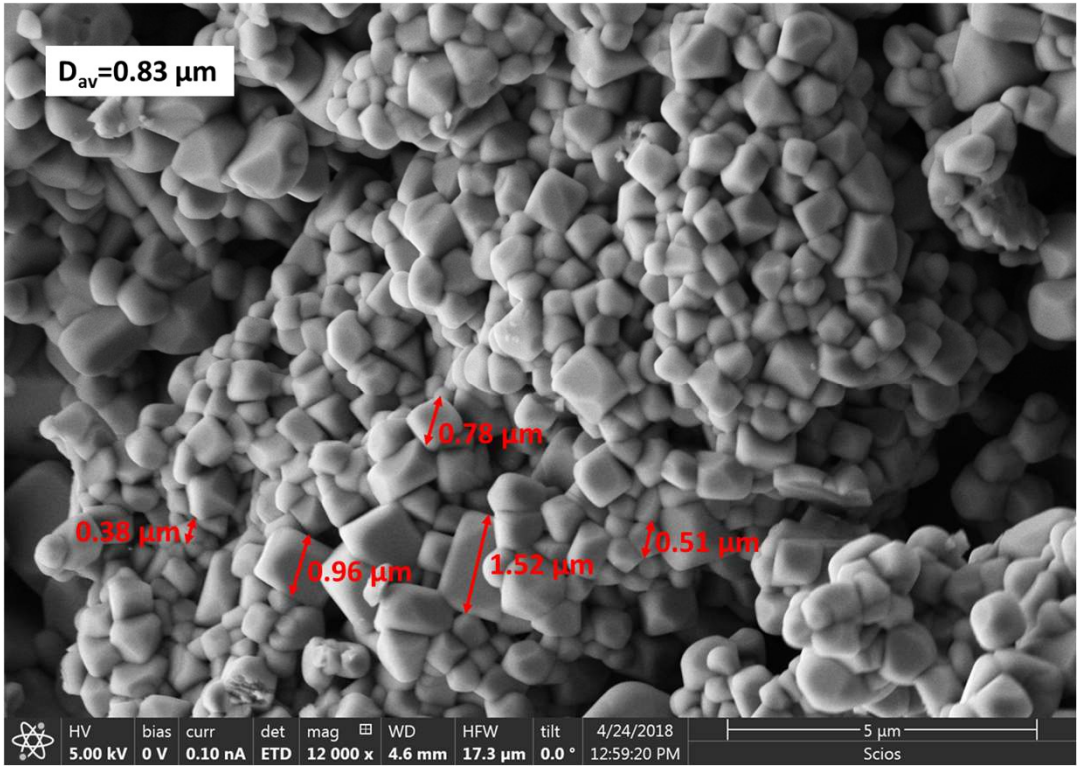
Conductivity



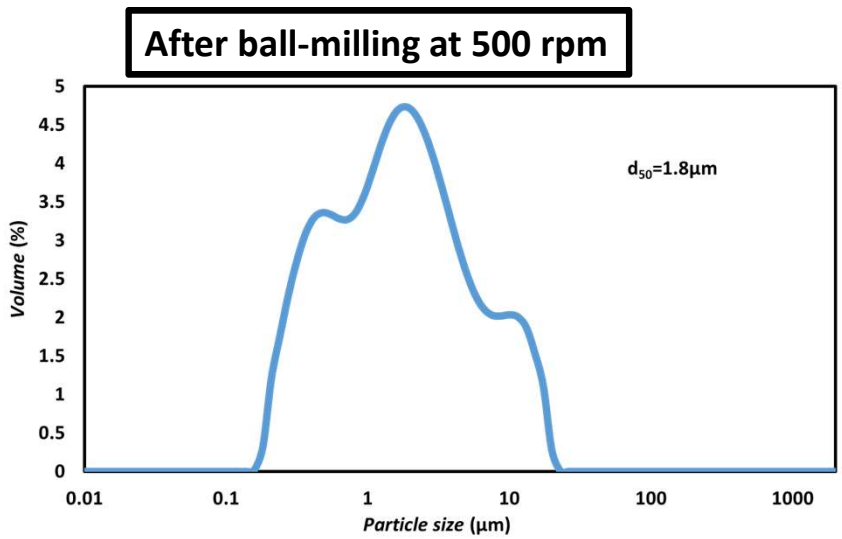
$\sigma_{20°C} = 0.83*10^{-3}$ S/cm

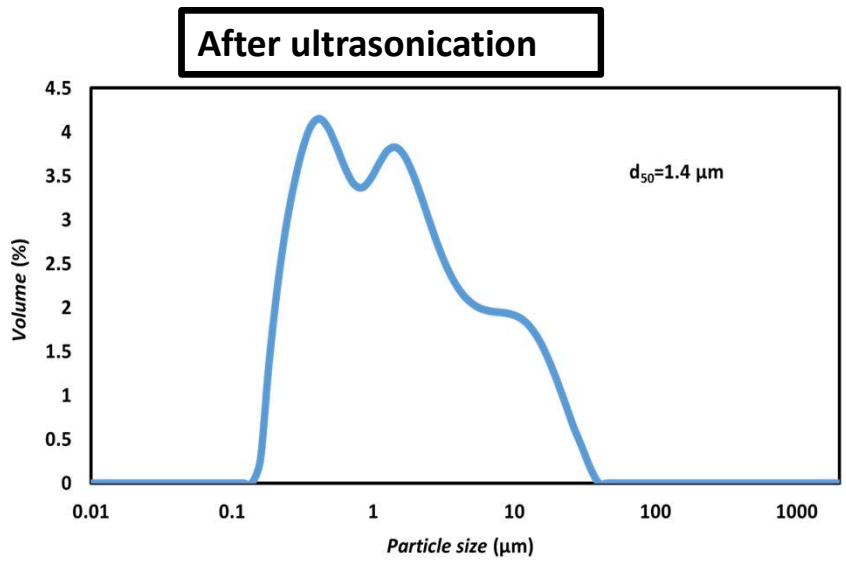
NZSP powder

- Average size of primary particle: 0.83 µm
- Agglomerates?



Powder processing





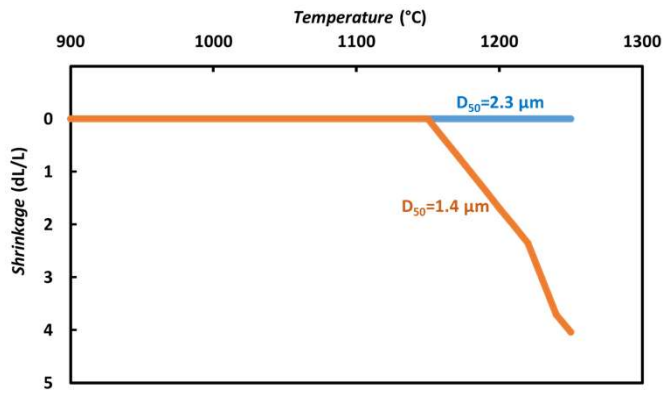
Removal of big agglomerates was carried out in two steps:

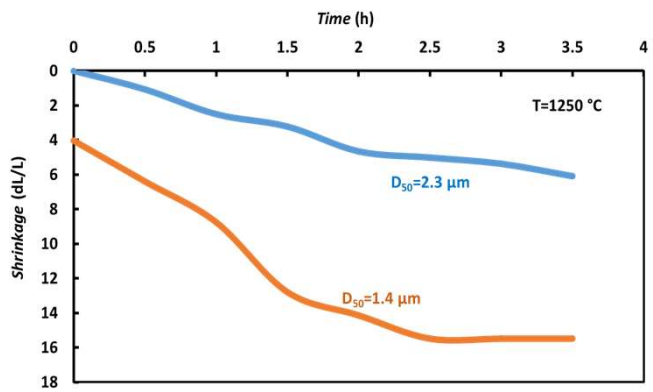
- Ball-milling
- Ultrasonication

Dilatometry for NZSP tapes

- NZSP tapes were prepared with two different particle sizes.

- NZSP powder with particle size D_{50}=1.4 µm resulted in:
  - lower sintering temperature
  - better sinterability

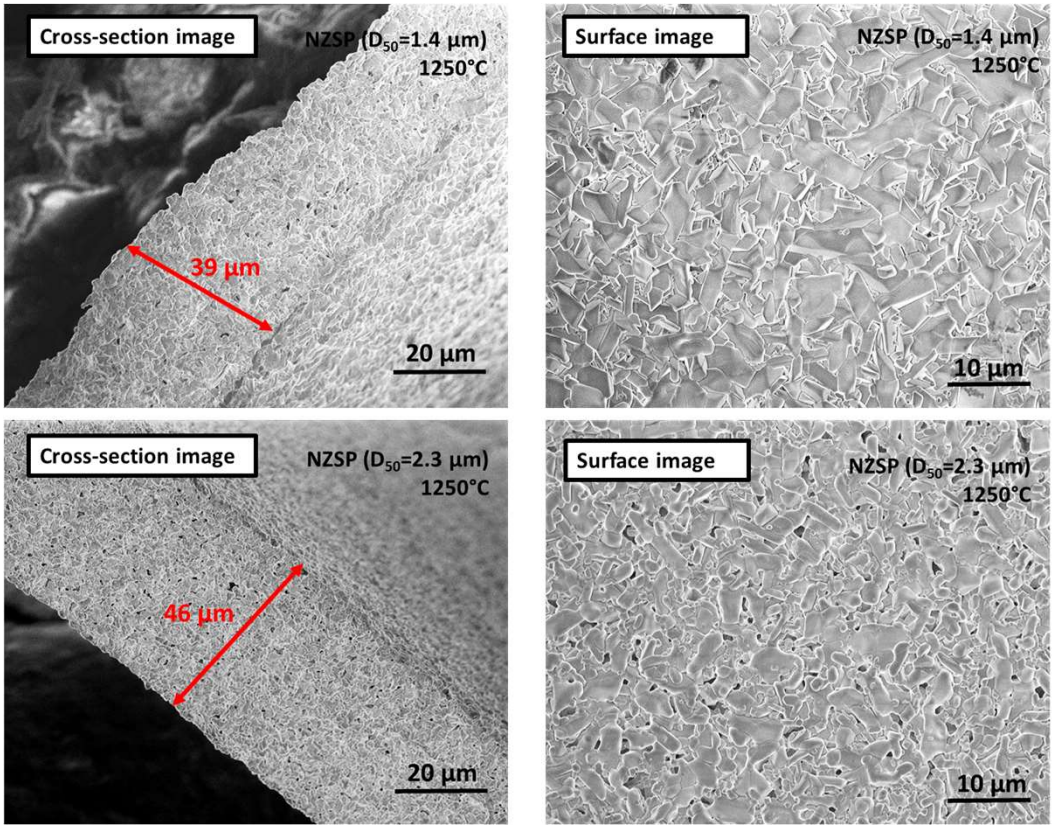




NZSP electrolyte layers

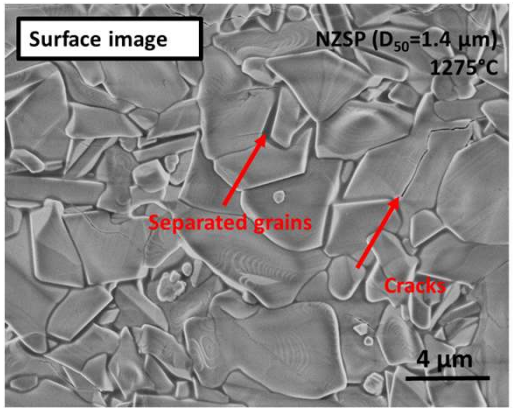
NZSP powder with better sinterability (D_{50}=1.4 µm)

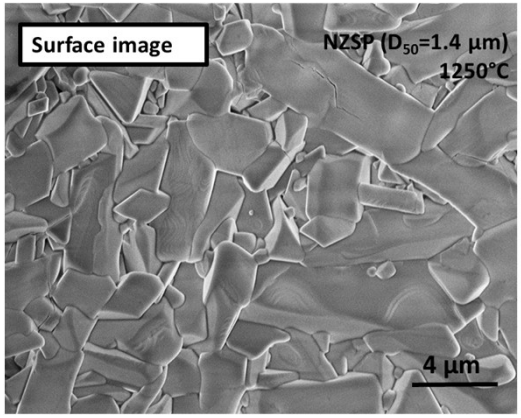
- denser microstructure
- smaller thickness



Effect of heat treatment

- Limits on sintering temperature
- Optimized heat treatment provides dense structure with good grain-to-grain connectivity.
- Higher temperatures cause formation of Zr-rich phase between the grains.
- Cracks are typical for this material.





Conclusion & Future work

- NZSP powder was synthesized and its particle size was tailored.
- NZSP electrolyte layers were fabricated by tape-casting.
- Optimized particle size and heat treatment ensured sufficient densification of NZSP material.

Next steps:

- Dense electrolyte will be sandwiched between porous scaffolds.
- Tape-casting technique will be adapted for LLZO and argyrodite materials.
- PhD student Cameron Bathgate will study wetting on LLZO material for infiltritation process.

Thank you for your attention!