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# On the role of binder in all-solid state battery using sulphide based electrolytes

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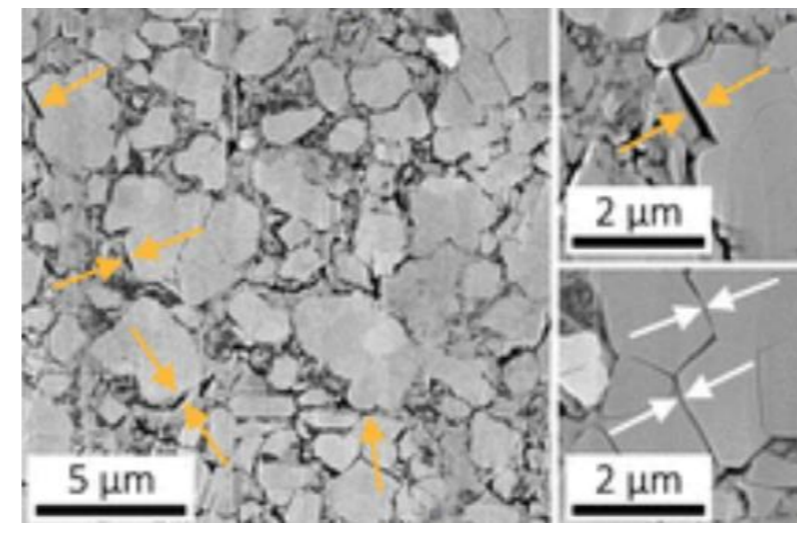
## Introduction

Sulphide solid electrolytes

- Very good ionic conductivity
- Easy processing (cold sintering)
- Poor electrochemical stability windows (**chemical decomposition**)
- Poor mechanical properties during cycling (**mechanical decomposition**)



Can the binder help improving the mechanical stability of the solid electrolytes ?



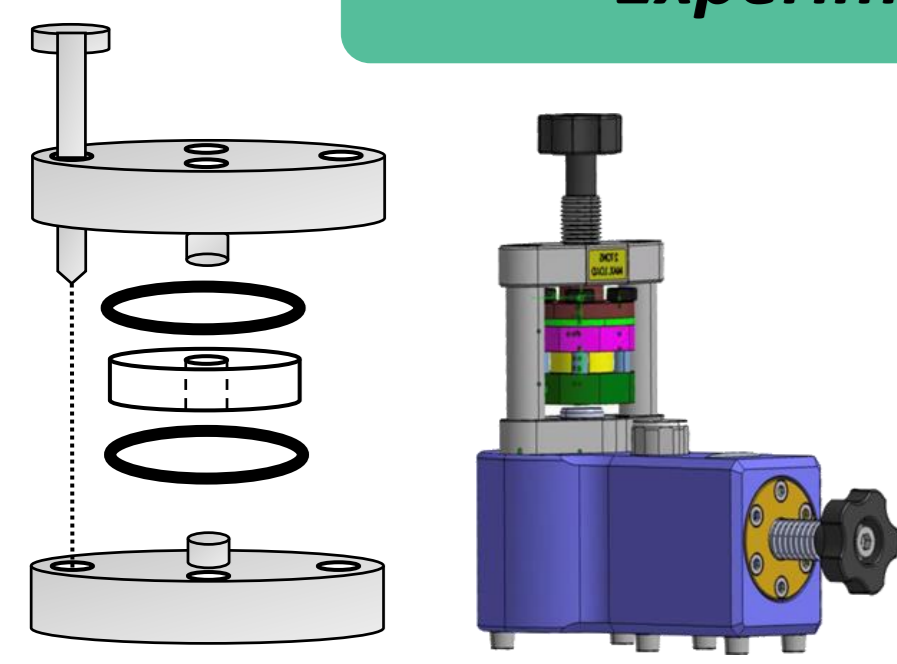
Doerr C, et al. ACS Appl Mater Interfaces. 2021;13(31):37809

## Strategy

Checking the impact of the binder on:

- The ionic conductivity
  - Impedance measurement
- The electrochemical stability window
  - Cyclic voltammetry
- The mechanical/chemical stability
  - X-ray tomography
  - Synchrotron XRD

## Experimental setup



Electrochemical cell closed under pressure in Ar-filled glovebox  
 Pressure up to 250MPa

## Chemicals

**Solid electrolyte**

Commercial LPSCI NEI

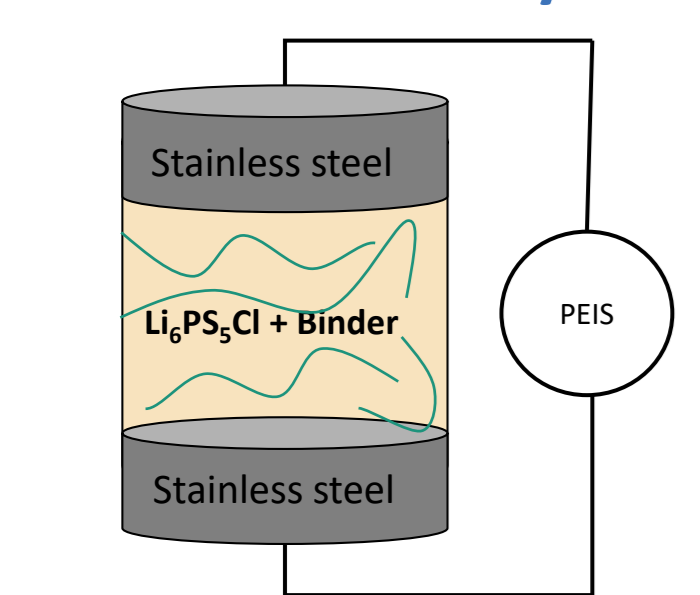
**Binder**

Polymer

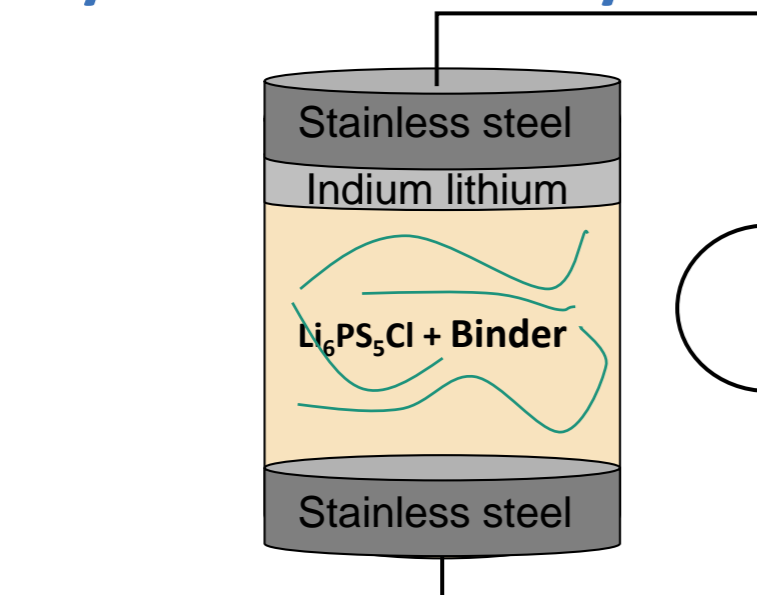
**Protocol**

Mixing LPSCI with X wt% of binder  
 Dry route  
 Hand mixing

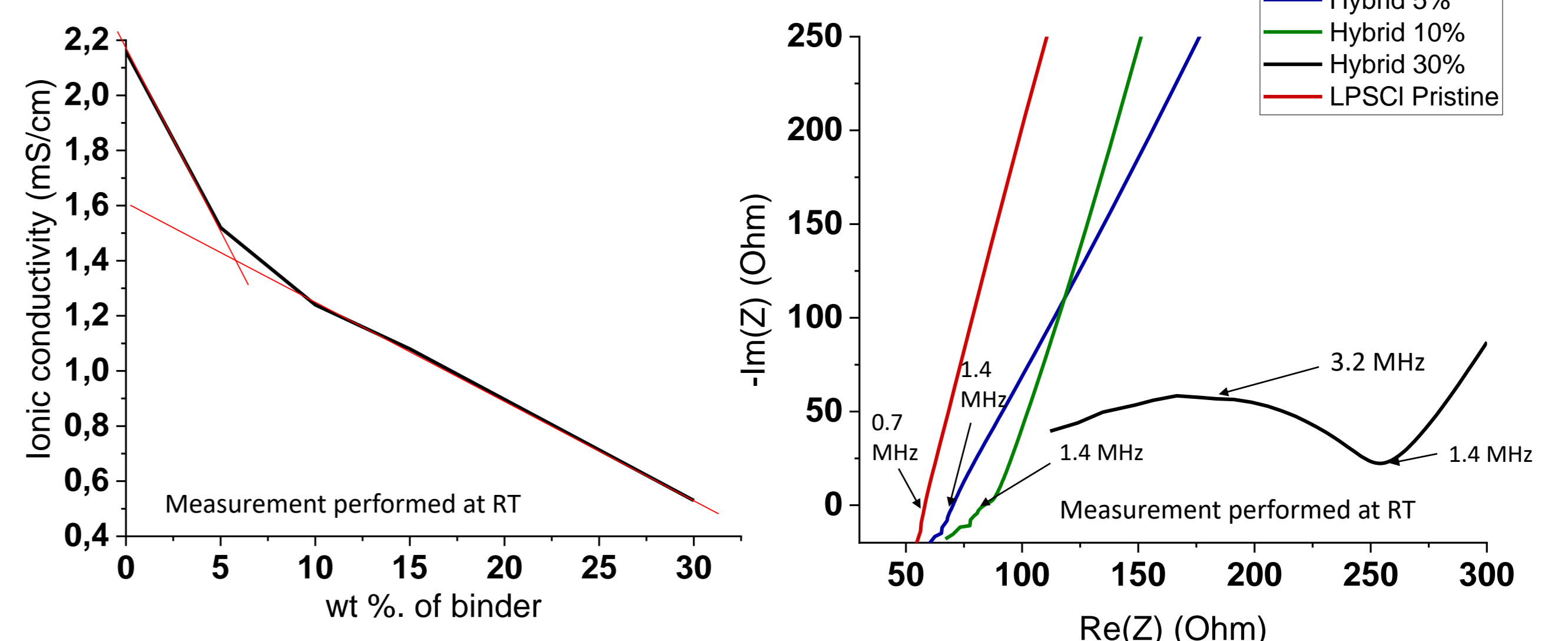
**Ionic conductivity cell**



**Cyclic voltammetry cell**



## Ionic conductivity



At low wt%, binder has a low impact of the ionic conductivity 😊  
 Ideal ratio content between 5-10 wt%.

## Structural evolution

### At RT

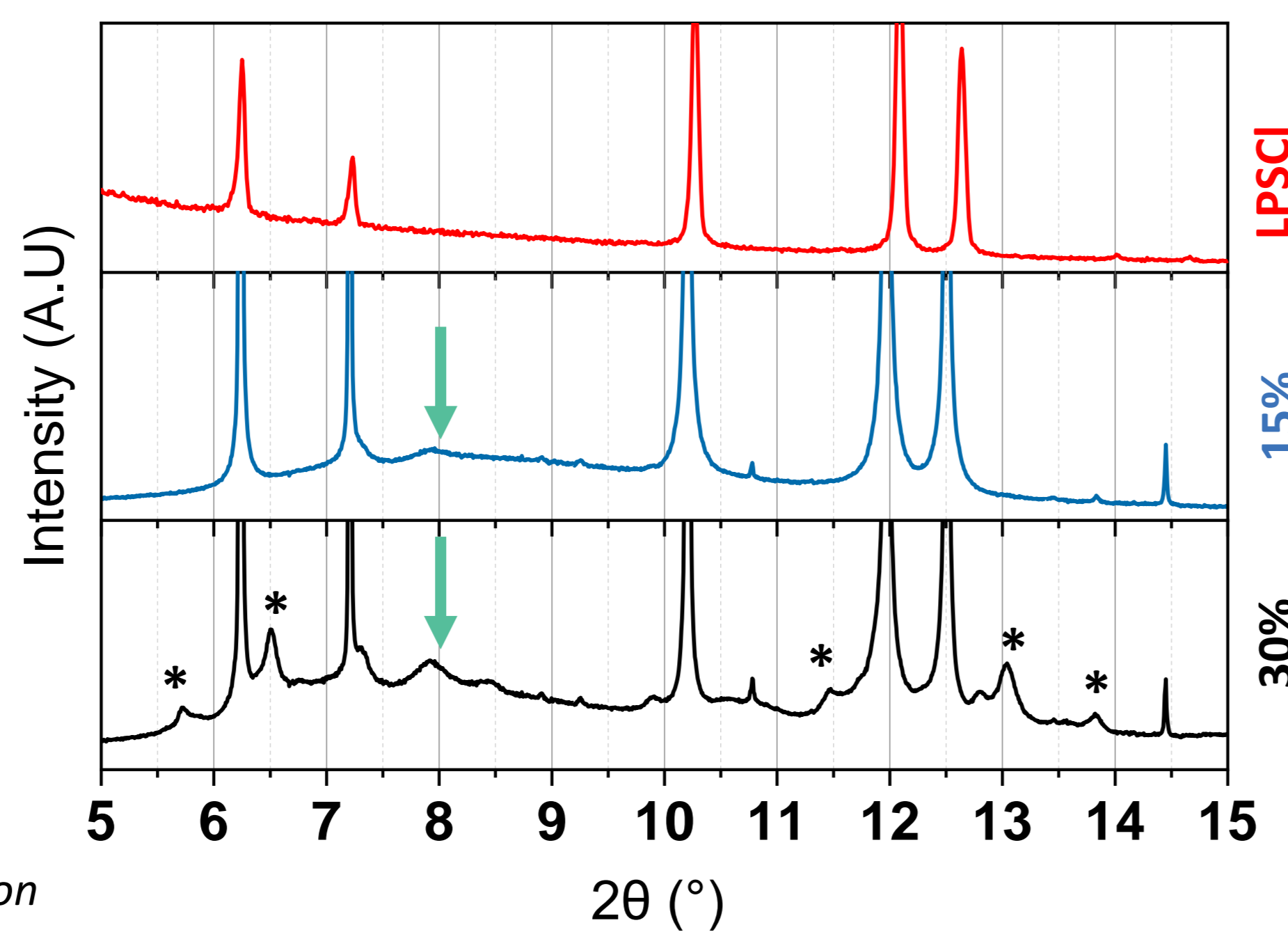
**LPSCI**

Well indexed as compared to the literature

**Hybrid**

Amorphous background coming from the binder

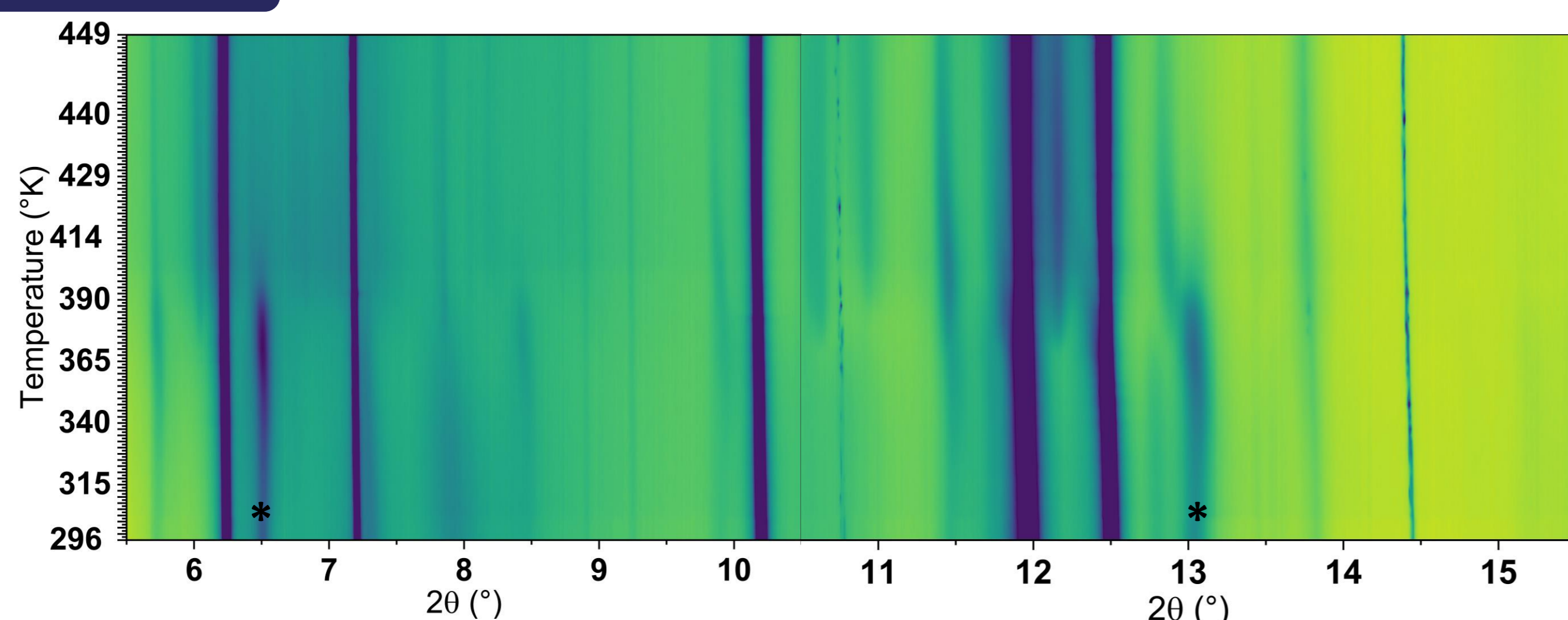
Little changes at 15 wt% (nothing at 10%)  
 Additional contribution at 30 wt% showing **chemical reaction** at RT



XRD patterns collected at ALBA synchrotron

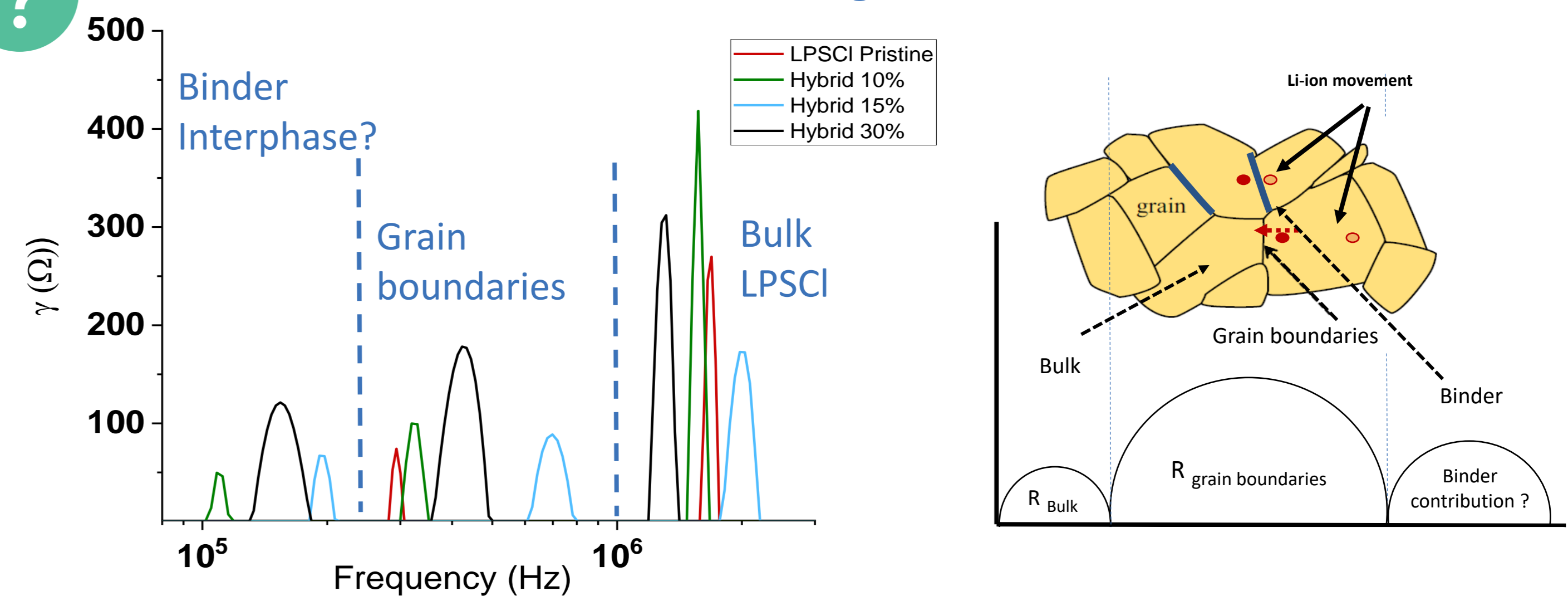
### During heating

Sample containing 30 wt% binder



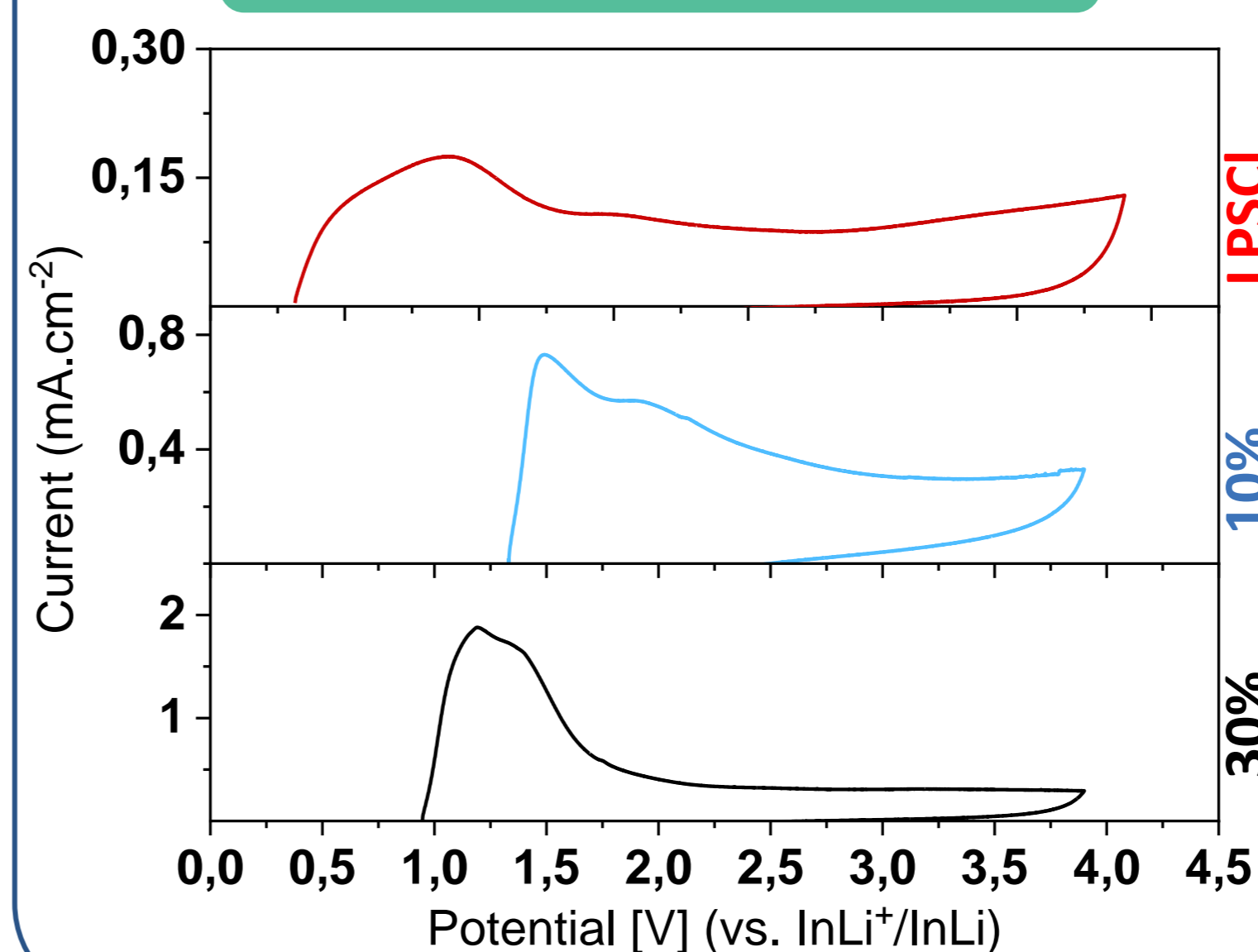
- Chemical decomposition visible at 25 °C with 30 wt% binder
- Impurities disappear with temperature treatment
- Other impurities appear with T°
- Irreversible structural transition from the binder under T°

## Does the binder influence the grain boundaries?



- At 10 wt% binder, the **hybrid electrode keeps LPSCI properties** with an additional contribution
- At higher loading, the frequency of R<sub>binder</sub> and R<sub>grain boundaries</sub> changed  
 → **chemical modification**

## Cyclic voltammetry



- Starting by oxidation
  - Decomposition visible at the beginning of the oxidation scan
- OCV shifts when binder is added : **chemistry evolved prior cycling (see XRD)**
- Current in oxidation is higher
  - Decomposition of the solid electrolyte **enhanced**

## Conclusion

- Impact of binder on hybrid ionic conductivity at room temperature :  
 - high ionic conductivity at 5 wt% :  $\sigma = 1.5 \text{ mS.cm}^{-1}$
- High percentage of binder is detrimental but help understanding the degradation process
- Optimal ratio to keep good LPSCI properties  
**Between 5-10 wt%**

## Outlook

- Degradation of hybrid electrolyte as a function of time
- Use different binder chemistry to tune the binder effects
- Study of morphology by tomography at ESRF : in progress

