OPERANDO EXPERIMENTS TO CHARACTERISE BRITTLE FRACTURE-LIKE EVENTS IN CERAMIC ELECTROLYTES *VIA* PHOTOELASTICITY

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Abstract
Solid electrolytes at current densities that are relevant to real battery operating conditions are prove to the penetration of lithium metal protrusions, also known as “dendritic” events, that are formed during battery charging. In this work, we show *via* operando photoelasticity experiments that the dendritic events at high current densities can be understood by the classic Griffith - Irwin fracture theory.

1. Introduction
The measurement of stress fields around lithium metal dendrites in solid electrolytes in operating conditions is critical for the design of next-generation, dendrite-resistant solid electrolytes. Prior work, both experimental and theoretical, indicates that the developed stresses in the in the electrolyte can be significantly high. However, direct stress measurements are still missing as they entail inherent experimental difficulties associated with probing stress fields at small scale brittle specimens’ operando.

2. Results
By employing the principle of photoelasticity combined with electrochemical cycling in a plan-view cell the aforementioned challenges are bypassed, allowing not only to track the stress field as the dendrite events progress, but also to obtain whole-field stress information on a propagating dendrite in an LLZTO electrolyte. This new experimental methodology allows for direct stress measurements around the dendrite tip. The experimental data demonstrate that the dendritic events at such current densities can be understood by the classic Griffith fracture theory. For an idealized straight Griffith crack where a constant internal pressure, p_o, exerts a normal force the crack faces, the following result has been obtained:

$$\sqrt{\left(\frac{\sigma_{xx} - \sigma_{yy}}{2}\right)^2 + \tau_{xy}^2} \cong p_o \sqrt{\frac{c}{8\,r}} \sin\psi \left(1 - \frac{3\,r}{4\,c}\cos\psi\right) \quad (1)$$

where r is the radial distance from the crack tip while ψ is the transformed angular variable.

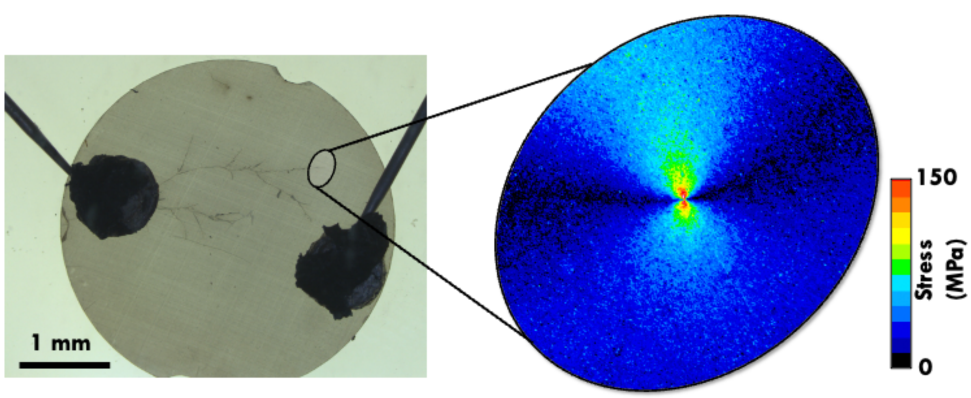

Figure 1: (left) Dendritic events on a ceramic electrolyte pellet (right) Operando photoelasticity measurements at the tip of the dendritic events provide critical information on the stress field.

3. Conclusions

The results of this work provide fundamental understanding of dendritic growth phenomena at high current densities in solid electrolytes and can be used to design next-generation dendrite-resistant electrolytes though mechanical design. The contactless characterization platform developed here can be further utilized for in operando studies of complex electro-chemo-mechanical phenomena at multiple length and time scales.

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