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$Li_7La_3Zr_2O_{12}$ electrolyte for all-solid-state batteries

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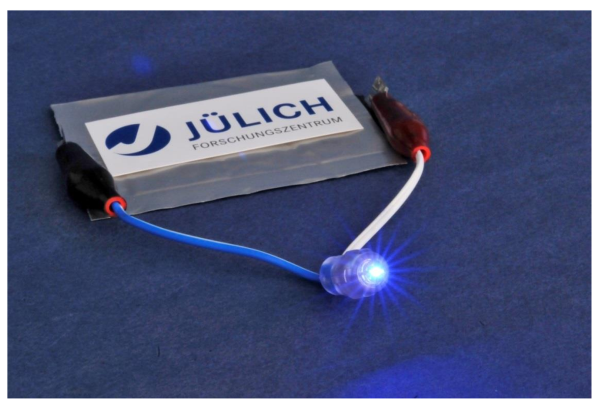
A major drawback of conventional Li-ion batteries is the use of organic liquid electrolytes. As an alternative, all-solid-state batteries with one of the most promising oxide materials, $Li_7La_3Zr_2O_{12}$ (LLZ) are investigated. LLZ is an ionic conductor with a good thermal and electrochemical stability (up to 1250°C and 8V vs. Li/Li⁺) and a chemical compatibility to metallic lithium. The ion conductivity can be further improved by partial substitution of Al, Ta or Y into the LLZ.

At IEK-1, we investigate two main approaches for all-solid-state battery fabrication. Very thin layers are processed by PVD aiming to achieve a thin film battery in the range of a few micrometers. For large scale fabrication of functional layers, tape casting of LLZ is investigated.

CONCLUSION & OUTLOOK

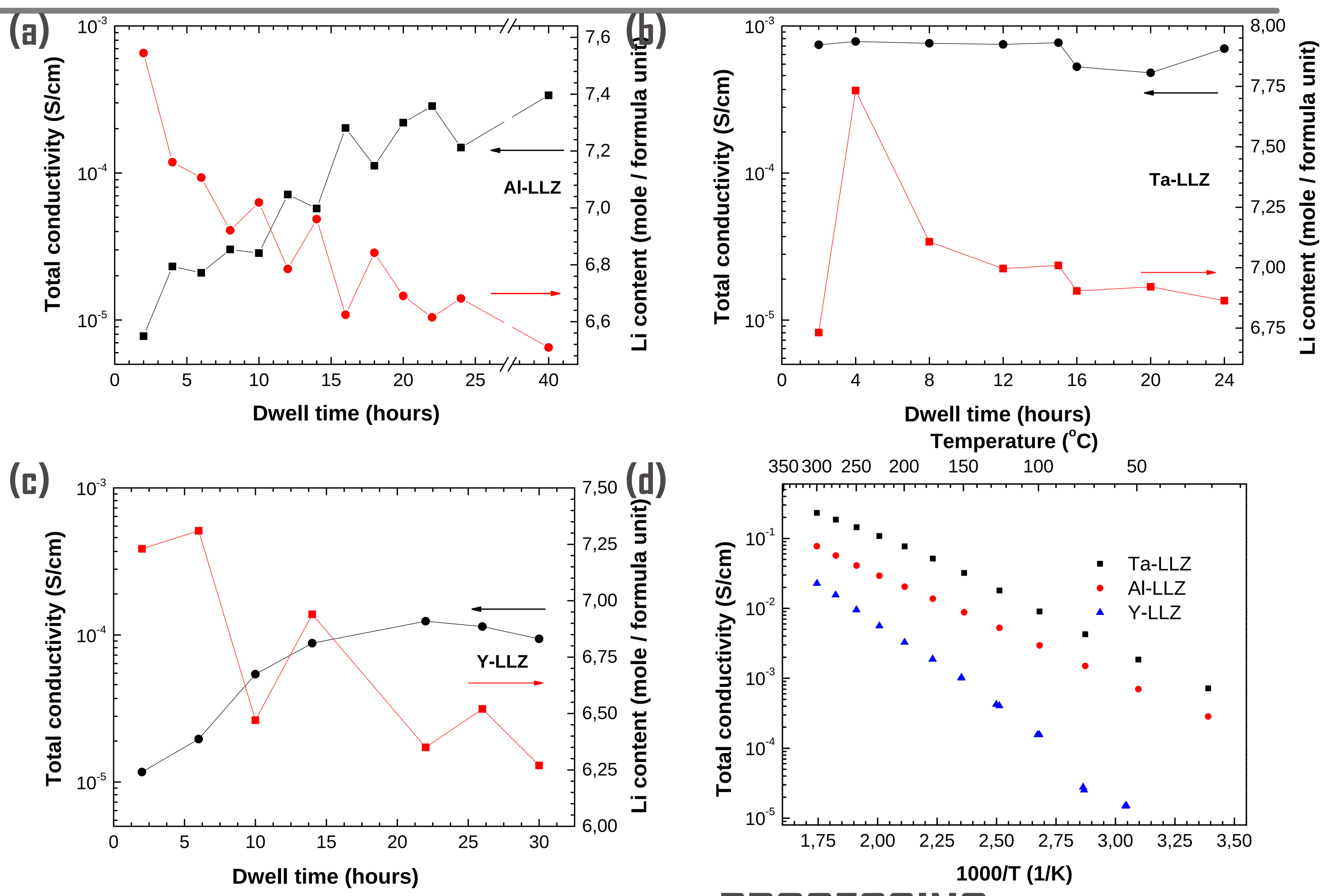
- Al-, Ta- and Y-substituted LLZ were investigated for different Li concentrations with respect to the materials conductivity. Ta:LLZ shows the highest total conductivity ($\sigma_{ion,RT} \approx 10^{-3}$ S/cm) and almost no dependence on Li concentration.
- LCO|Ta:LLZ|Li all-solid-state cell was fabricated from bulk sample and drove a LED at 22°C **(ASB works!)**
- LLZ can be fabricated by tape casting and PVD processing.

→ By combining tape casting and PVD processing a thin all-solid-state battery with higher energy density should be developed.

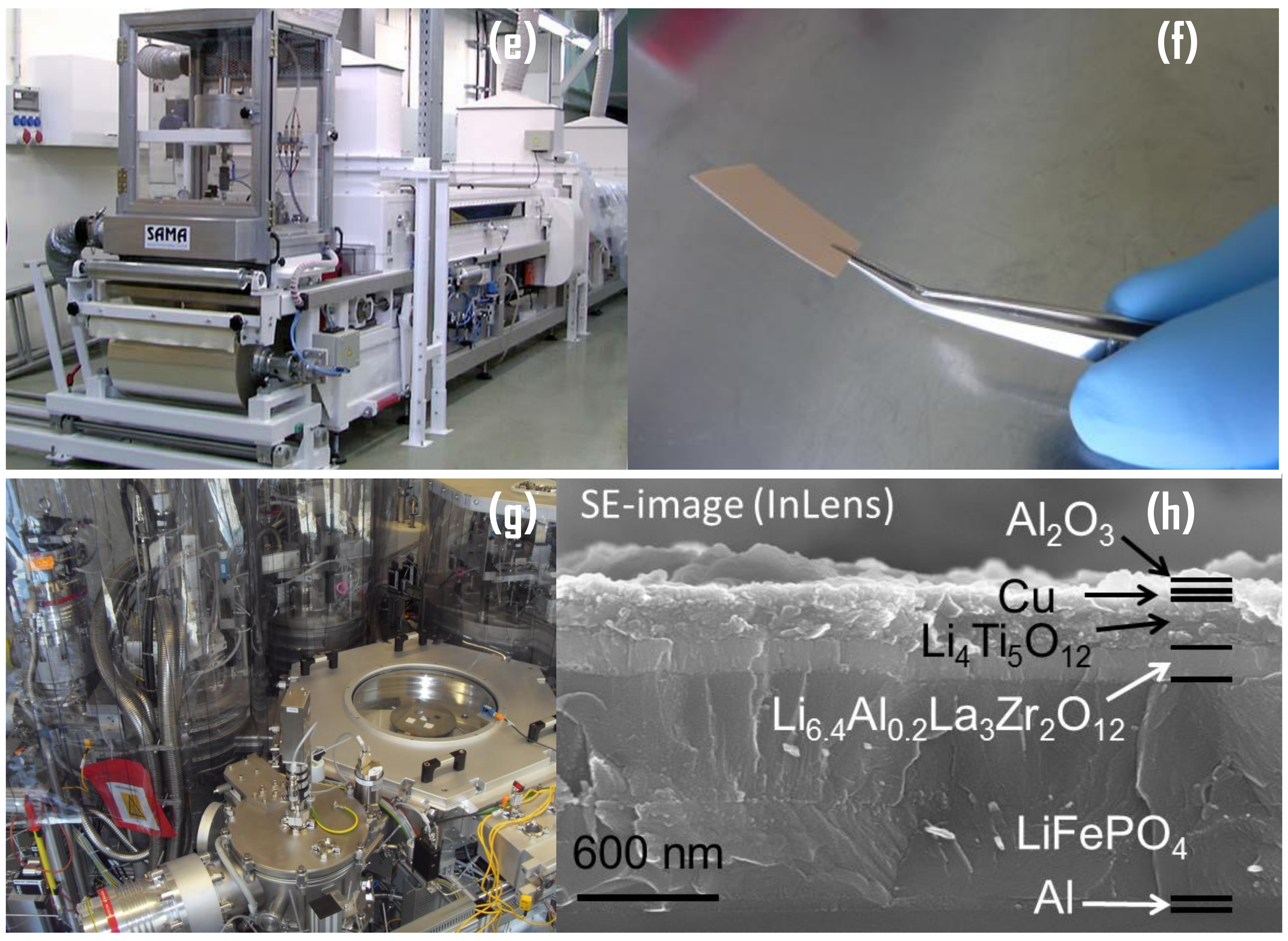


MATERIAL RESEARCH

- Inductively Coupled Plasma (ICP) measurement of Li concentration in **(a)** Al-substituted LLZ, **(b)** Ta-substituted LLZ and **(c)** Y-substituted LLZ. There is a high correlation between Li concentration in Al- and Y-substituted LLZ and the total conductivity but not for Ta-substituted LLZ. The molar numbers were normalized to Zr concentration.
- **(d)** Temperature dependence of the ionic conductivity of Al-, Ta- and Y-substituted LLZ. Ta-substituted LLZ shows the highest total ionic conductivity among the three materials. The total conductivity was derived from impedance spectra (1MHz-1Hz, amplitude: 20 mV/mm, temperature range: 22-300°C).



PROCESSING



- Synthesis of several kilograms of Al-substituted LLZ by spray pyrolysis at once.
- Fabrication of LLZ by easily up-scalable **(e)** tape casting at IEK-1 to **(f)** 90 µm thick green-tape substrates. These tapes are used for sintering studies at different temperatures and atmospheres.
- Thin film electrodes or solid electrolytes like LLZ can be successfully deposited by **(g)** physical vapor deposition (PVD) at IEK-1. The thickness and adhesion of these thin layers can be visualized by **(h)** SEM. Between an aluminum (Al) and copper (Cu) conductor LFP ($LiFePO_4$) was used as cathode and LTO ($Li_4Ti_5O_{12}$) as anode material. The electrodes were separated by Al-substituted LLZ ($Li_{6.4}Al_{0.2}La_3Zr_2O_{12}$). On top an Al_2O_3-protection layer is deposited.

Acknowledgements

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