Laser-based processing enables high temperature sintering of LLZO on metallic current collectors.

Florian Ribbeck^{1*}, Nico Schmitt¹, Jonas Frühling¹, Samuel Fink¹, Nikolai Helth Gaukås², Per Martin Rørvik², Leif-Olav Jøsang³, Florian Harth⁴, Thomas Brand⁴

1 Fraunhofer-Institut für Lasertechnik ILT, Steinbachstr. 15 52074 Aachen Germany, North Rhine-Westfalia

2 SINTEF Industri, Forskningsveien 1 0373 Oslo, Norway

3 Ceramic Powder Technology AS, Kvenildmyra 6 7093 Tiller, Norway

4 Coherent-Dilas GmbH, Galileo-Galilei-Str. 10, 55129 Mainz, Germany

* Presenting author e-mail: <u>florian.ribbeck@ilt.fraunhofer.de</u>

As conventional lithium-ion batteries approach their maximum theoretical power densities, innovative cell concepts are under investigation. To achieve the highest possible gravimetric energy density a metallic lithium anode is desired. Dendrite growth through the separator is a safety concern and a major reason for decreasing battery performance. Lithium lanthanum zirconate (LLZO) is a garnetbased ceramic solid-state lithium-ion conductor with chemical stability against metallic lithium¹ and a sufficiently high shear modulus to withstand penetration of lithium-dendrites². The processing of this material and the implementation in an all-solid-state battery (ASSB) proved to be challenging³. Due to the formation of unwanted secondary phases during sintering and cofiring with cathode active materials (LCO, NMC) in conventional oven processes the development of novel sintering processes is required. Photonic sinter processes like rapid thermal processing (RTP)^{4,5}, blacklight sintering⁶ or laser-based sintering⁵ are promising technologies for sintering processes with short interaction times that allow a preservation of the desired crystal structure. In this work, the laser sintering of LLZO thin films is investigated. Therefore, LLZO-powder is mixed into a paste and applied to a titanium substrate by screen printing. The pure LLZO-layer shows an absorptance of around 20 % for blue laser radiation, compared to 5 % for 1 µm laser radiation. The absorptance of the layer is further adjusted using various amounts of copper oxide (CuO) as additive. The application of LLZO as a separator requires a high density and high ionic conductivity. Therefore, a laser sintering process with blue laser radiation is developed to form a dense layer on a steel substrate. Goal of this work is the investigation of the influence of process parameters on crystal structure, phase composition, porosity, adhesion, and electrochemical properties.

1 R. Murugan, V. Thangadurai, Angew. Chem. Int. Ed., 2007, 46, 7778-7781

2 C.-L. Tsai, V. Roddatis, ACS Appl. Mater. Interfaces, 2016, 8, 10617-10626

3 X. Huang, Y. Lu, Energy Storage Materials, 2019, 22, 207-217

4 A. T. Fiory, Journal of Electronic Materials, 2002, 31, 981-987

5 L. C. Hoff, W. S. Scheld, Proc. SPIE, 2022, 11989,

6 L. Porz, M. Scherer, Mater. Horiz., 2022, 9, 1717-1726