The Study of $Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$ (LLZTO) Thin Films Using Pulse Laser Deposition (PLD)

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1 Introduction

Lithium lanthanum zirconium oxide ($Li_7La_3Zr_2O_{12}$) (LLZO) is a ceramic material, specifically, garnet-type. LLZO has lithium-ion conductivity that can reach to 10^{-4} S/cm, or higher at room temperature. Also, it is considered to has high thermal and chemical stability interface with Li metal. It can be one of the promising electrodes due to its energy density, electrochemical stability, high temperature stability, and being safe [1]. LLZO has cubic crystal structure, but tetragonal crystal structure can also occur which is not quite helpful, as it has lower conductivity (lower by two orders of magnitude) compared to the cubic structure. The formation of the tetragonal structure can be avoided using high sintering temperatures and durations. However, due to the lithium volatility (at high temperatures), pores may develop, which will decrease the conductivity. In this case, an applicable solution can be substituting amounts of Tantalum (Ta), which then will be ($Li_{7-x}La_3Zr_{2-x}Ta_xO_{12}$) (LLZTO), can help with obtaining a cubic structure [2].

Synthesizing LLZTO can be a bit challenging, as using a normal solid-state reaction requires high sintering temperature which lead to the same problem faced in LLZO (results in a porous material, that may have lower conductivity). Recently, various studies have shown that the use of Sol-gel method can be a good synthesis method due to its various advantages, like high purity, high stoichiometry controllability, and the low processing temperature [2].

Using a large solid sample will not be useful and applicable for the real-world application, as the world and industries are now moving to use small objects, like phones, laptops, solar panels, etc. Therefore, a large LLZTO will not add much to this field. This research will focus on depositing a LLZTO thin film that then can be used in Lithium-ion batteries for the different applications. Also, thin films are considered to have better properties compared to the bulk, this can be due to the differences in morphology, structure (heterostructure will develop by using this technique), and composition. The synthesis of this thin film will be done using a Pulsed Laser Deposition (PLD) machine. PLD is based on physical vapor deposition (PVD) technique, inside an ultra-high vacuum chamber, a beam of laser will strike the sample, where LLZTO will be vaporized and deposited as a thin film (in micro-meter thickness) on a substrate. Further, X-ray Diffraction (XRD) analysis will be used to make sure that it is the desired material and structure. Such to ensure that it has a cubic structure, determine phases present, and check for any impurities that may exist.

In general, it is expected that the deposition will work well. However, there are some possible errors/challenges that may appear, including, being unable to have a good sample after dry pressing (this will need to be done multiple times to get a perfect sample), the substrate that the LLZTO film will be deposited on will not be appropriate (different substrates need to be used and examined), and the laser in the LLZTO deposition may need be adjusted multiple times to ensure perfect alignment.

2 Procedure

In this research, for better use of time, the Sol-gel method will not be used to produce a new LLZTO sample. As the main aim of this research is to successfully deposit a LLZTO thin film and characterize it. Instead, an old broken LLZTO sample will be re-used for this study, where the sample will be re-grinded to form a powder. Then, it will be dry pressed followed by sintering to allow more and even elements diffusion, and make the sample/target denser. After sintering, the sample will go through X-ray Diffraction (XRD) for further analysis.

After confirming that it is the right material and structure, using XRD, the sample/target will be placed in a Pulsed Laser Deposition (PLD) machine where the thin film will be deposited. Again, after successfully getting the deposited film, which is the main challenge and the purpose of this research, the film will go through XRD for analysis and make sure that the LLZTO was deposited right and as desired. Finally, after ensuring we have the right LLZTO film, it can go through further characterization, like measuring conductivity and other properties.

References

[1] Chengwei Wang et al. "Garnet-type solid-state electrolytes: materials, interfaces, and batteries". In: *Chemical reviews* 120.10 (2020), pp. 4257–4300.

[2] Sang A Yoon et al. "Preparation and Characterization of Ta-substituted Li 7 La 3 Zr 2-x O 12 Garnet Solid Electrolyte by Sol-Gel Processing". In: *Journal of the Korean Ceramic Society* 54.4 (2017), pp. 278–284.