

a rewrite of M. Nakajima et al., The 65th Battery Symposium in Japan, 2F19(2024)

AC Impedance Measurements of Solid Electrolyte LLTO™(La_{0.57}Li_{0.29}TiO₃)

1. AC impedance measurement system

Fig.1 shows the AC impedance system consisting of NEISYS (Novocontrol), 22cm-coaxial cables and the heat controllable fixture (Qualtec, Japan). The fixture also provides us various gas atmosphere (Ar, N₂, vacuum etc.).

2. Evaluation of the AC impedance system

We used chip resistors(1Ω-1MΩ) as standard specimens and the 5% accuracy criteria for evaluation of this system. And we decided the 5% accuracy area by |Z| (absolute impedance) and θ (phase) measured. In the case of θ, within ±4.5° are acceptable because 4.5° are the 5% of the ideal maximum phase(±90°). The 5% accuracy map is the colored area of this system in Fig.2(left). Note the |Z| accuracy area also includes the θ accuracy. We finally achieved the 5% accuracy map of 5Ω-510Ω@100MHz, 1Ω-10kΩ@10MHz, 1Ω-100kΩ@1MHz and 1Ω-1MΩ@100kHz or less in this system.



Fig.1 The AC impedance measurement system (NEISYS, 22cm-coaxial cables and the fixture).

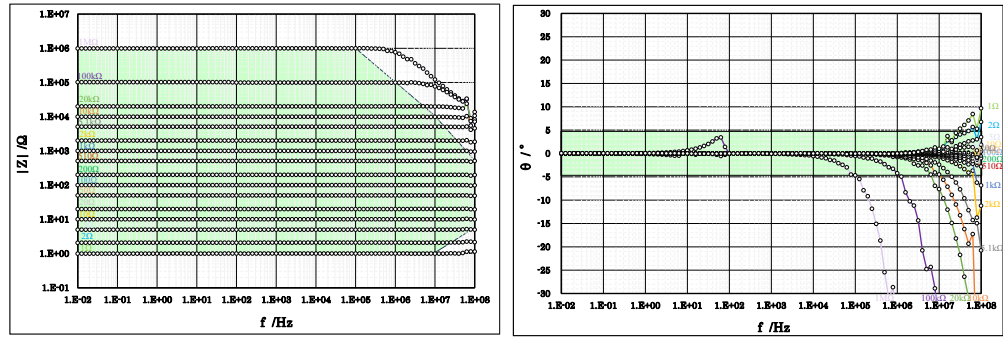


Fig.2 The colored 5% accuracy map of |Z| (left) and θ (right) in the AC impedance measurement system (NEISYS, 22cm-coaxial cables and the fixture). Note the |Z| accuracy includes the θ accuracy.

3. LLTO™ (La_{0.57}Li_{0.29}TiO₃) pellet sintered (approximately φ8mm and 1mm in thickness)

We measured AC impedance at -50°C, -25°C, 0°C, 25°C, 50°C, 100°C, 150°C, 200°C and 250°C under Ar gas atmosphere. Fig.3 is the Nyquist plots at 25°C showing clear separation of bulk, grain boundary and electrode components, respectively. Fig.4 is the overlapped data measured at all the temperatures showing that the data around 100MHz are also within the 5% accuracy map. In addition, all the data are in the 1st quadrant as expected before the measurement. Fig.5 is the Arrhenius plots showing that the linearity of grain boundary conductivity and total conductivity is kept all through the temperature range, while that of bulk conductivity isn't. The inflection of bulk conductivity plots around 150°C might show the existence of a second phase.

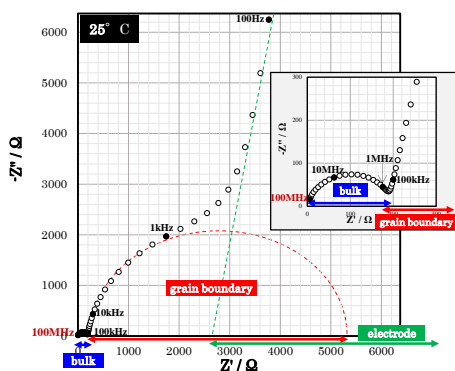


Fig.3 The Nyquist plots of LLTO™ pellet (25°C only). The inserted graph is enlarged data around 100MHz.

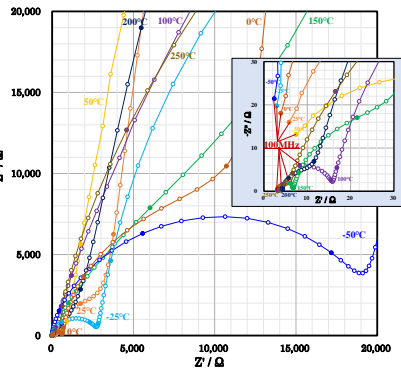


Fig.4 The Nyquist plots of LLTO™ pellet (-50°C-250°C). The inserted graph is enlarged data around 100MHz.

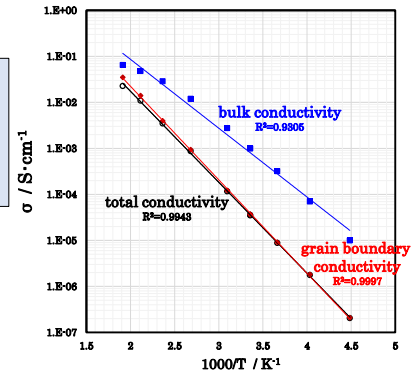


Fig.5 The Arrhenius plots of LLTO™ pellet show the clear separation of bulk(blue) and grain boundary(red) conductivity.