The authors could address my initial comments. However, upon re-reading this manuscript, I have additional comments, especially regarding the solid-state NMR analysis. I consulted an NMR specialist whom I trust. The paper requires additional revisions before JMCA can consider it.

Page 3: "However, it has been previously reported that Li_3InCl_6 can turn into $Li_3InCl_6.xH_2O$ upon exposure to moisture.[40,41] Thus, the formation of a similar compound such as $Li_{2.1}Gr_{0.1}Zr_{0.9}Cl_6.xH_2O$ could be expected."

Their interpretation that the shoulder at -0.9 ppm (for the LGZC electrolyte on its own) is due to the formation of a hydrated $Li_{2.1}Ga_{0.1}Zr_{0.9}Cl_6.xH_2O$ phase is hard to believe. While a hydrated phase is formed for Li_3InCl_6 ($Li_3InCl_6.xH_2O$) (Sun and coworkers, Energy Environ. Sci., 2019,12, 2665-2671, and Chem. Mater. 2020, 32, 16, 7019–7027) this is an exception in this family of compounds and is the reason why this is the only material that can be recovered by heating after moisture exposure.

Page 3: "To further validate this, the ^1H spectrum of the halide was recorded and analyzed. As seen in Figure 1.b, the ^1H spectrum reveals several broad signals. "

The authors attempt to prove that the protonated phase forms by collecting ^1H NMR data on the halide electrolyte, but their spectrum looks exactly like one would expect for the NMR probe ^1H background signal (the spectrum obtain if one were to collect a ^1H spectrum on an empty probe or empty rotor in the probe because some parts of the probe are made of plastic and this results in a background signal). Basically, there is no signal at all from the pristine LGZC sample. In Figure S4, they compare the 1H spectra obtained on fresh and aged halides, and the aged halide clearly shows extensive protonation (the probe background signal is no longer visible).

Page 4: "As shown in Figure S.5, this spectrum evolution was also accompanied by an increase in the ^7Li T_1 relaxation time. This indicates that the interphase species have slower local Li dynamics than the main argyrodite and halide phases. "

There are two significant issues with this claim:

1) There are several signals in the spectra shown in Figure 3: each signal has a specific T1 relaxation time, yet the authors only mention "the" T1 relaxation time. Which T1 are they talking about? Is this an average T1 value for all signals? This makes no sense at all.

2) One cannot infer slower Li dynamics by comparing the ^7Li T1 relaxation times *at a single temperature point*. Here, the authors compare "the T1" of the composite electrolyte pre and post-heating, presumably at one temperature because they do not mention any variable-temperature measurements, and then say that because "the T1" increases in the sample post-heating, this means that the Li dynamics are slowing down. To do this properly, one would need to measure the T1 relaxation time for each signal in the sample at various temperatures and then fit the T1 data (for each signal separately) using, for example, the Bloembergen-Purcell-Pound (BPP) equation:

$$\frac{1}{T_1} = \frac{2(\delta_\omega)^2\tau_c}{1 + (\omega_0)^2{\tau_c}^2}$$

where ω_0 is the Larmor frequency and $\delta\omega$ the average difference in the local field as seen by the nuclear spin during each molecular jump of which the average jumping time is τ_c. An Arrhenius behavior can generally be assumed for the Li-ion residence time $\tau_c = \tau_0 \exp(\frac{E_a}{k_b T})$, and from this one can obtain an activation energy E_a for diffusion.

Page 5: The authors then go on to inferring that the slower Li-ion conductivity of the interphase formed between the two electrolytes (as indicated from the change in “the T1” at a single temperature of the powder) explains why the EIS data does not evolve after 9 hours → they seem to suggest a passivating interface but they have no evidence for this. They are incorrectly using their NMR data to support this.

Page 5: Were the EXSY spectra shown in Figure 5 presumably obtained on the composite electrolyte after heating? This is not mentioned in the figure caption nor the main text.

When explaining the EXSY results on page 6, the authors refer to “the two halide signals at -0.85 and -1.1 ppm,”, i.e. they are assigning the -1.1 ppm signal that appeared upon mixing the two electrolytes to a halide phase but they do but they do not have any proof and should support their assignment better.

Later on, the authors discussed decomposed sulfide (1.1 ppm) and decomposed halide (-1.1 ppm). Still, again, they do not support their assignments (the decomposition phases come from *both* the sulfide and the halide, so why attribute the origin to only one of the electrolytes?)