Water based synthesis of highly conductive $Ga_xLi_{7-3x}La_3Hf_2O_{12}$ garnets with comparable critical current density to analogous $Ga_xLi_{7-3x}La_3Zr_2O_{12}$ systems.

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Figure S1. XRD of Ga-LLHO and LLHO during synthesis, all heating steps were 700°C, with a 20% wt. excess of lithium added between steps.



Figure S2. Al-LLHO synthesis steps, with a 20% wt. excess of lithium added between steps, except for the final 950°C heat treatment. A prior synthesis demonstrated no gain in reheating to 700°C, hence the step was removed for this synthesis approach.



Figure S3. XRD data for $Ga_{0.15}Li_{6.55}La_3Hf_2O_{12}$ during attempted synthesis (showing the presence of a mixed cubic and tetragonal phase), an additional 20% wt. of Lithium was added until the 3rd heating cycle, with no excess added for the heat treatment at 800°C.



Figure S4. XRD data $Li_7La_3Zr_2O_{12}$ synthesised by the dissolution method, illustrating the expected tetragonal symmetry with space group I4₁/acd (no. 142)



Figure S5. XRD data for attempted co-doping at the Li site with the formula $Ga_{0.2}Li_{6.4}La_3Hf_{1.8}Ce_{0.2}O_{12}$. The system demonstrated similar impurities to the Ga-LLHO synthesis with an addition of Cerium oxide based impurities which were unable to be removed, despite adding 10-20 % wt. lithium and/or addition of 3-5% HfO_2 excess (due to hydration levels of starting Hf salt) during the heating steps.



Figure S6. XRD data for attempted Pr doping at the Hf site with the formula $Li_7La_3Hf_{1.5}Pr_{0.5}O_{12}$. The system demonstrated similar impurities to the LLHO synthesis with an addition of Pr_2O_3 impurities which were unable to be removed, despite adding 5-10 % excess Li_2CO_3 and/or addition of 3-5% Hf/La excess (due to hydration levels of starting salts) during the heating steps.



Figure S7. XRD data for attempted La replacement with Nd, Eu, Gd and Er, with only Nd-LHO demonstrating the formation of a garnet type phase. All intended formulas correspond to $Li_7M_3Hf_2O_{12}$ (M = Nd, Eu, Gd, Er).



Figure S8. Example Rietveld refinement data for Ga-LLHO.



Figure S9. Impedance spectrum of Ga-LLHO at 25°C when densified under O_2 . Although the spectrum was fit to two R/CPE components in parallel (where R1/CPE1 corresponds to the bulk and R2/CPE2 to the grain boundary contributions), the errors for the fit were quite large and, considering the similarities in conductivity between O_2 and N_2 densification, this sample was not cooled any further for analysis.

	Ga-LLHO (25°C)
σ_{total} (S cm ⁻¹)	3.7 x10 ⁻⁴
C _{bulk} (F / cm)	7.08 x10 ⁻¹²
ε _r	80
ρ _{rel} (%)	88

Table S1. Impedance data for Ga-LLHO when densified under O_2





10µm



Figure S10. a) t-LLHO SEM and EDX map and b) SEM and EDX map of ground powder from densified LLHO pellet. The LLHO pellet was unable to be sanded to examine the pellet surface due to the low relative density.

