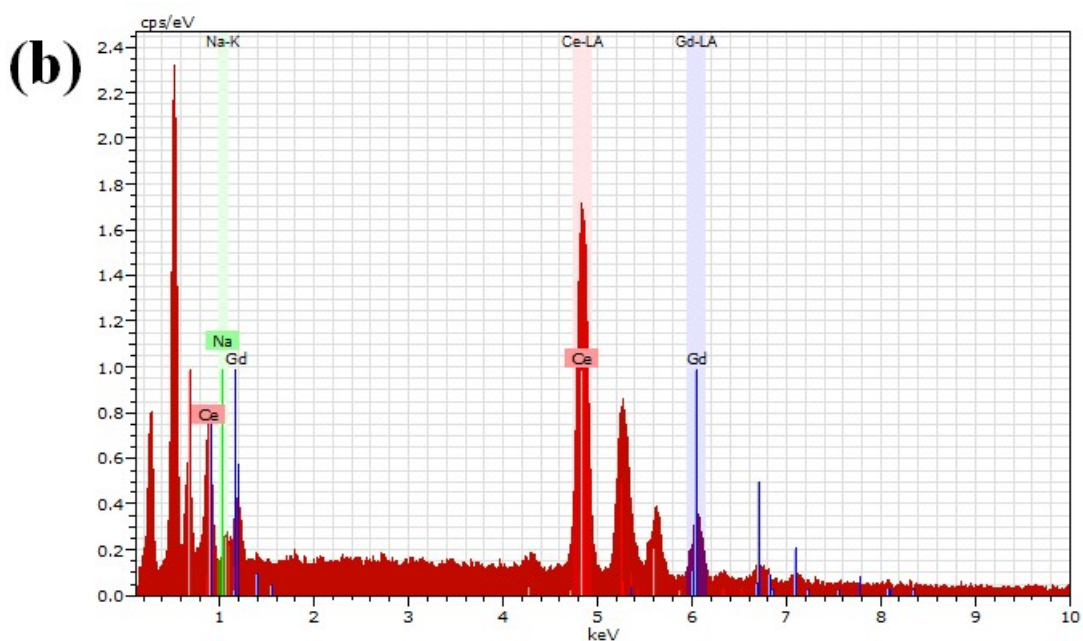
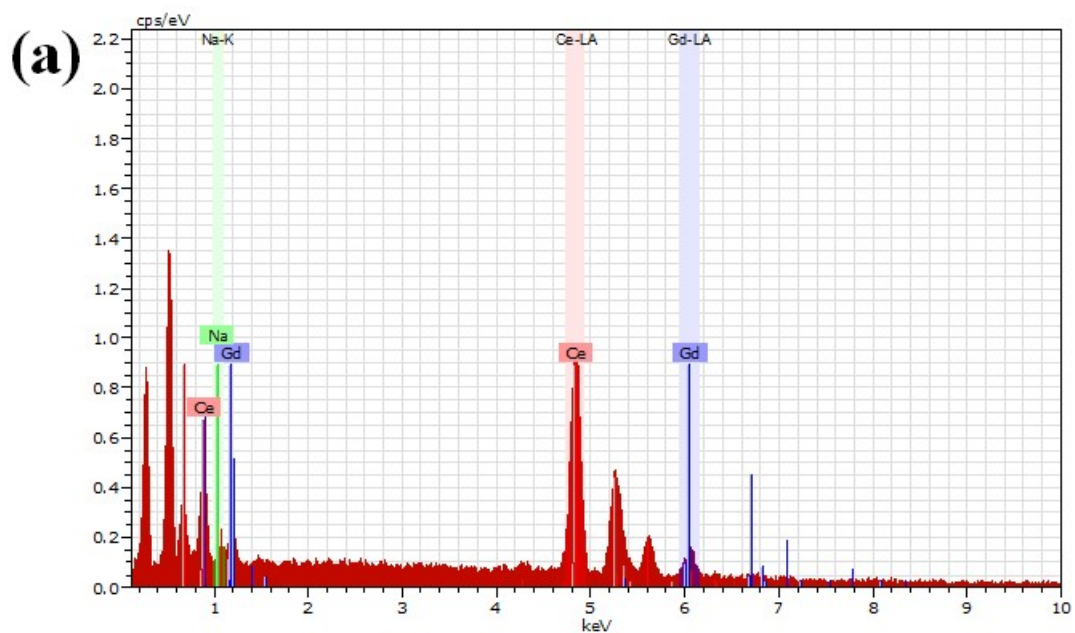
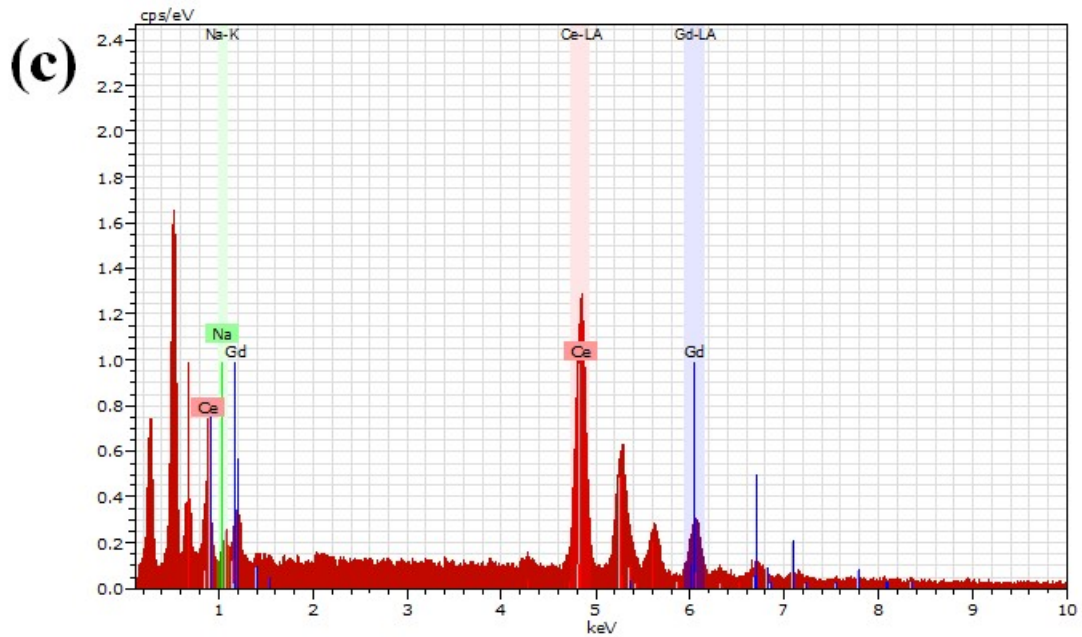


## Optimization of Safe Doping Level for Enhanced CO<sub>2</sub> Flux in Composite Membrane

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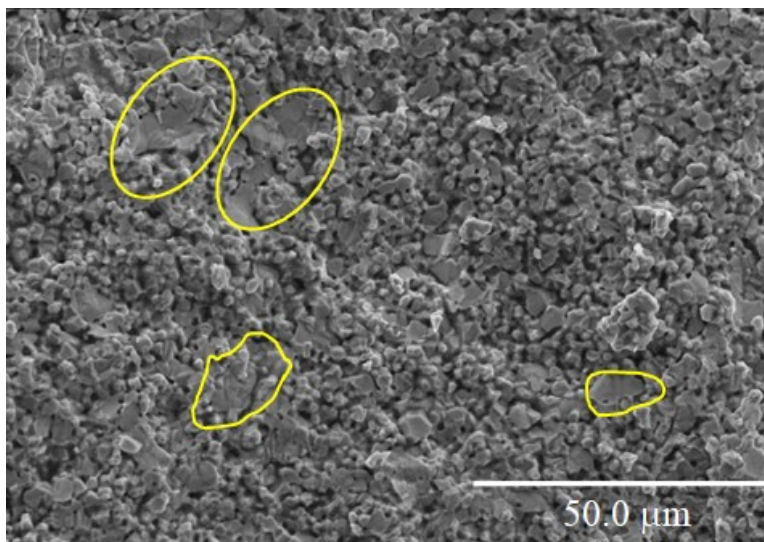
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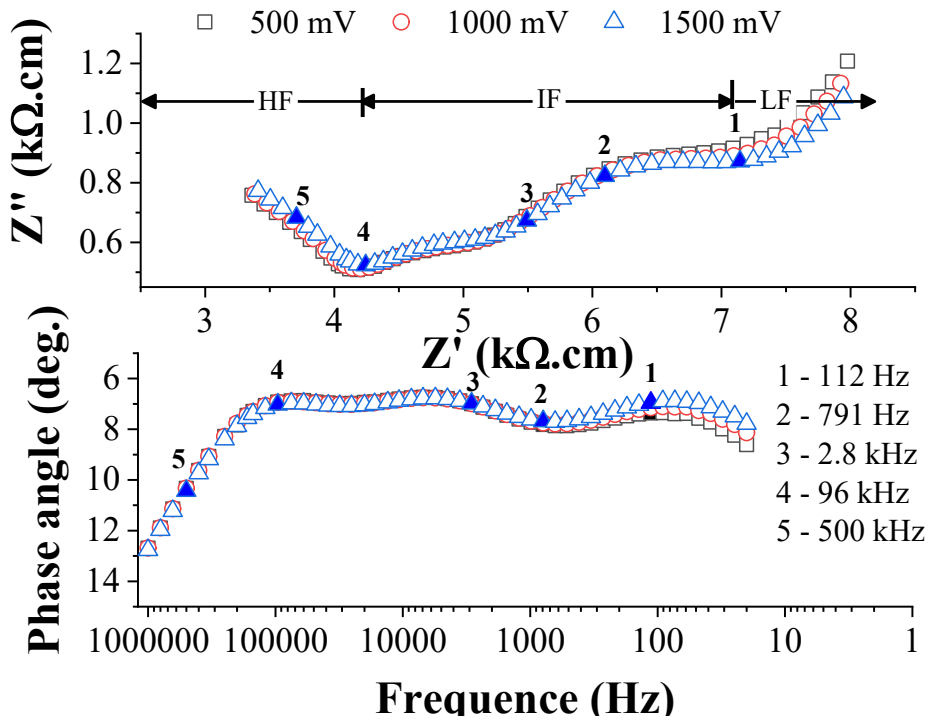
**Fig.S1:** EDS spectra of a) CGO10, b) CGO15, and C) CGO20 matrices

EDS analysis of CGO10, CGO15, and CGO20 matrices was carried out at SEM magnification of 10K and for larger areas. The quantitative analysis was obtained from Ce-La and Gd-La lines of spectra. The increasing intensity of Gd-La peaks with respect of Ce-La of CGO10, CGO15, and CGO20 samples indicates the growing amount of Gd% in those samples. Gd-La to Ce-La intensity ratios calculated for respective samples also exhibit increasing trend as, CGO10(0.18)<CGO15(0.22) <CGO20(0.25).



**Fig.S2:** SEM of CGO15 matrix at low magnification

As it can clearly see from figure S2 some large and hard agglomerates or particles remain intact alongside of smaller sizes. The consequent agate mortar and ball milling grindings of powders retain particle both sizes in ceramic matrices and composites. Thus, sample's uniformity can be well judged from low magnified SEM (typically below 1K), while high magnified SEM (above 5K) gives the local microstructural identity of ceramics and composites.



**Fig.S3:** Impedance spectra of CGO20C composite at 350 °C

The Nyquist plots and phase angle dependence on a logarithmic scale of frequency are shown in **Fig. S3**. Nyquist plots at RMS amplitudes of 500 mV, 1000 mV, and 1500 mV depict three curves in high and intermediate frequency domains, and open arc in low frequency domain. The dispersive nature of low frequency arcs, recorded at different RMS voltages, gives indication of electrode contribution. Further, according to relaxation frequency, the ceramic grains and grain-boundary responds at high and intermediate frequencies, respectively, and electrode at low frequency. The co-existence of two consecutive curves in the intermediate frequency region designates the involvement of additional parallel conduction mechanism. As diffusion through pure  $\text{LiNaCO}_3$  activates only above the melting point (500 °C), the conduction in low temperature region can be a result of extrinsic and highly mobile ionic species. It is also well known that the carbonates are susceptible and easily adsorb water from air. Thus, changed surface chemical composition of carbonate due to water molecules may partly induce the conductivity resulting from fast diffusion of hydroxide ions in a low temperature region.