## **Supplementary Information**

## Novel strategy for improving the oxygen permeability of the

## zirconia-based dual-phase membranes

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Fig. S1 Low magnification cross-sectional SEM image of NNO coated membrane.

The thickness of the coating layers was controlled by the number of printing, and low resolution SEM images of cross-section were shown **fig. S1**. The coating layer was uniformly formed on both sides of the membrane with a thickness of about 24 to 29  $\mu$ m, and it is confirmed that the thickness of coating layer was reproducible as well as homogeneous in this study.



**Fig. S2** Back-scattered electron image of 70 vol.% ScSZ- 30 vol.% LSM composite membrane (light grains: LSM, dark grains: ScSZ).



**Fig. S3** Oxygen permeation flux of NNO coated membrane as a function of the flow rate of sweeping gas at 900 °C (feed side: synthetic air, permeate side: He).



**Fig. S4** (a) XRD patterns of  $Nd_2NiO_4$  powder after aging test and  $Nd_2NiO_4$  coating layer after permeation test. (b-c) Surface images of NNO coating layer before and after permeation test (d-e) HR-TEM EDS mapping of NNO coated membrane before and after permeation test under air/CO<sub>2</sub> condition at 850 °C for 100 h.

**Fig. S4** shows the SEM/TEM/XRD analysis of post-tested samples exposed to  $CO_2$  at 850 °C for 100 hours. No differences in the results before and after permeation testing were observed in the post-test analysis of SEM, XRD, and TEM of NNO coating layer.



**Fig. S5** Impedance spectra of the NNO coated membrane measured at open circuit voltage and at cut-off potential of at 0 V, (inset) Time-dependence stability test of polarization resistance under oxygen permeation condition.

**Fig. S5** shows the time-dependence stability of the polarization resistance under real operating condition. The prepared NNO coated membranes [10 vol.% LSM- 90 vol.% ScSZ], which can be considered as the concentration cell, exhibit an open circuit voltage of 0.156 V under air/He gradient at 900 °C. Since the oxygen is not transported across the concentration cell in the OCV condition, the stability test was conducted at cut-off potential of at 0 volt which can implement the oxygen permeation. No deterioration of the polarization resistance was observed for 100 hours.