**SUPPORTING INFORMATION**

Optimization of the Mixed Proton-Electronic Conducting Materials based on the
\((Nd_{5/6}Ln_{1/6})_5_3WO_{11.25-δ}\)

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Figure S1: Structural characterization of NLnW compounds: XRD patterns as a function of the final sintering temperature.
Figure S2: Shrinkage evolution of pressed pellets made of NLnW as a function of the sintering temperature. For this purpose, pellets with a diameter 13 mm and uniaxially pressed at 301 MPa for 1 minute from the powder calcined at 900 ºC were sintered at different temperatures. From the resulting sample diameter, shrinkage curves were obtained. At 1480 ºC, the doped compounds suffer higher shrinkage than the observed in the parent compound.
Figure S3: Total conductivity of NWO and Gd, Sm and Yb doped compound in wet H₂ and Ar. No improvement in the total conductivity is obtained by partial substitution of Nd using Gd, Sm and Yb in wet Ar. Nevertheless, only NYbW presents higher total conductivity than undoped compound in wet H₂.
Figure S4: Hydrogen production scheme: hydrogen production by proton transport through the membrane on the left hand and hydrogen generation from water splitting reaction due to the oxygen ion transport from the sweep side to the feed side on the right hand.
Figure S5: XRD patterns of NLnW after stability treatments: materials sintered at 1350 °C exposed to a continuous gas flow composed of 10% (vol.) CO$_2$ and 90% CH$_4$ (2.5% water) at 800 °C for 72 hours (a); NWO sintered at 1350 °C and 1550 °C and NCeW sintered at 900 °C exposed to a continuous gas flow composed of 115 ppm H$_2$S, 4.43% CO$_2$, 2.12% CO and 92.09% H$_2$ at 500 °C and 30 bars for 40 h (b).