

Supplementary Information

Enhancing Surface Activity and Durability in Triple Conducting Electrode for Protonic Ceramic Electrochemical Cells

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This PDF file includes:

Equation S1

Figs. S1 to S11

Tables S1 to S19

Equation. S1.

Equation. S1. The relationship between the rate of a chemical reaction and the activation energy is described by the Arrhenius equation:

$$R_o = A_o \exp\left(-\frac{E_{a,o}}{\kappa_B T}\right) \quad (1)$$

$$R_p = A_p \exp\left(-\frac{E_{a,p}}{\kappa_B T}\right) \quad (2)$$

where A is the pre-exponent factor; E_a is the activation energy; the subscripts o and p denote the ohmic and polarization resistance, respectively; κ_B is the Boltzmann constant and T is the absolute temperature in Kelvin.

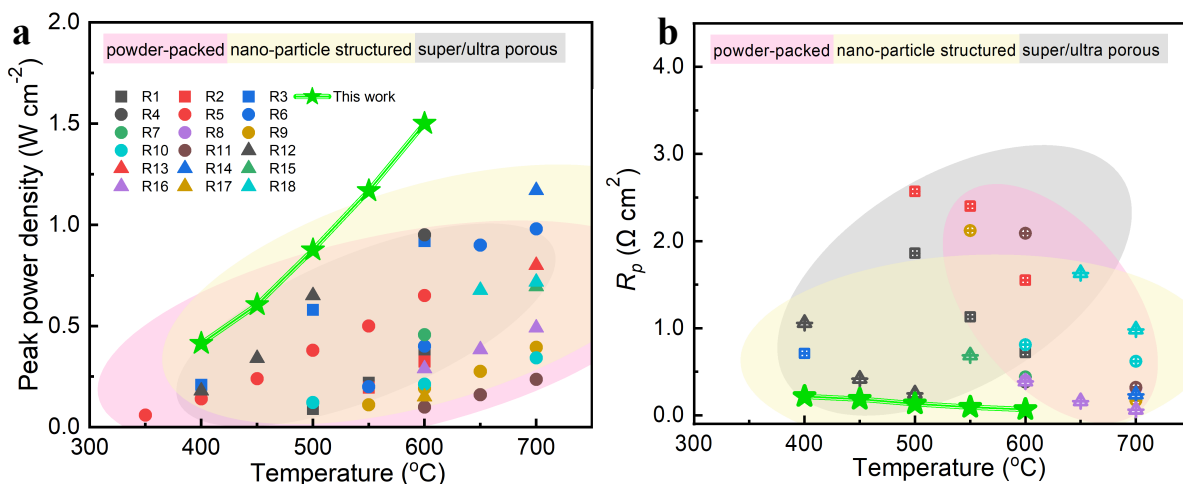


Fig. S1. Highly enhanced electrochemical performance of PCEC with nano-architecture ultra-porous (NAUP) PNC73 electrode. **a** Comparison on peak power density of NAUP-PNC73 (this work) in fuel cell mode with the literature results. The NAUP-PNC73 full cells are readily fabricated in this work using self-structured mesh to achieve ultra-porous configuration. Thus, another similar fabrication methods: powder-packed electrode; nano-particle electrode and super/ultra porous electrode, are used to provide a more distinct contrast. The temperature range of all conducted tests was 350-700 $^{\circ}C$ ¹⁻¹⁸. **b** Polarization resistance (R_p) of NAUP-PNC73 under OCV conditions in comparison with the representative results.

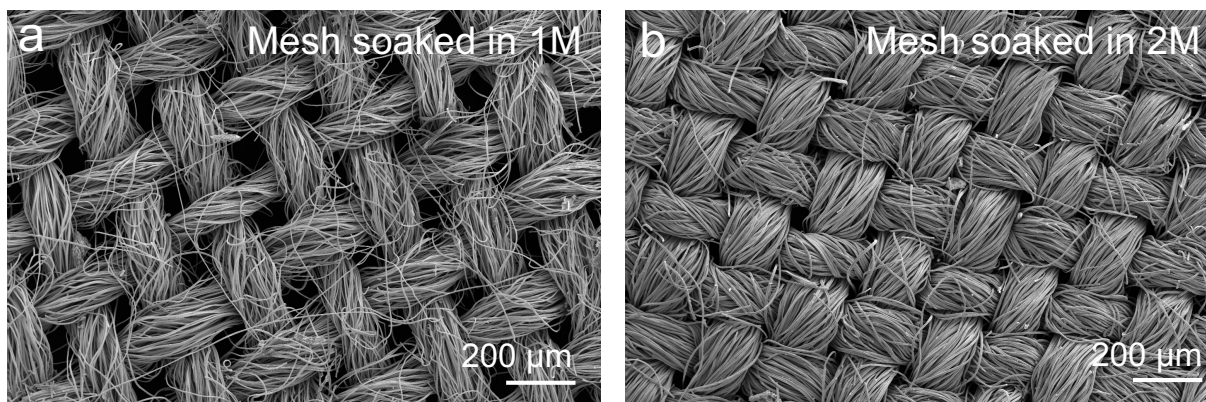


Fig. S2. SEM images of NAUP-PNC mesh-structured electrode. a Mesh soaked in 1M PNC73 solution. **b** Mesh soaked in 2M PNC73 solution.

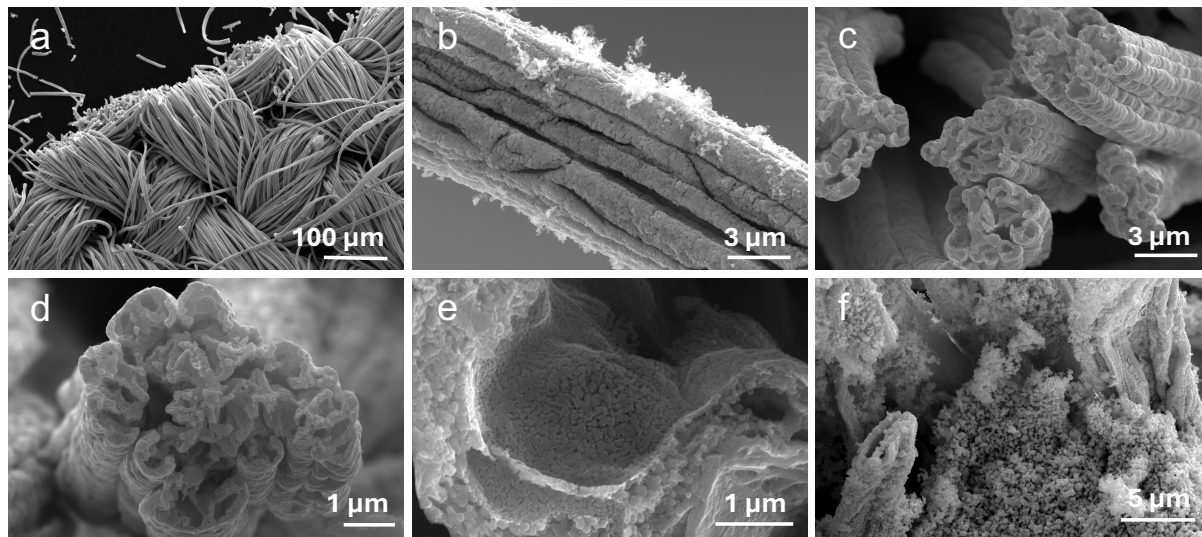


Fig. S3. SEM images of a NAUP-PNC mesh-structured electrode edges. **b** one NAUP-PNC mesh-structured electrode fiber. **c** cross sections of several NAUP-PNC mesh-structured electrode fibers. **d** the cross section of one NAUP-PNC mesh-structured electrode fiber. **e** the inner wall of one NAUP-PNC mesh-structured electrode fiber. **f** bonding relationship between NAUP-PNC mesh-structured electrode fibers and PNC73 ink.

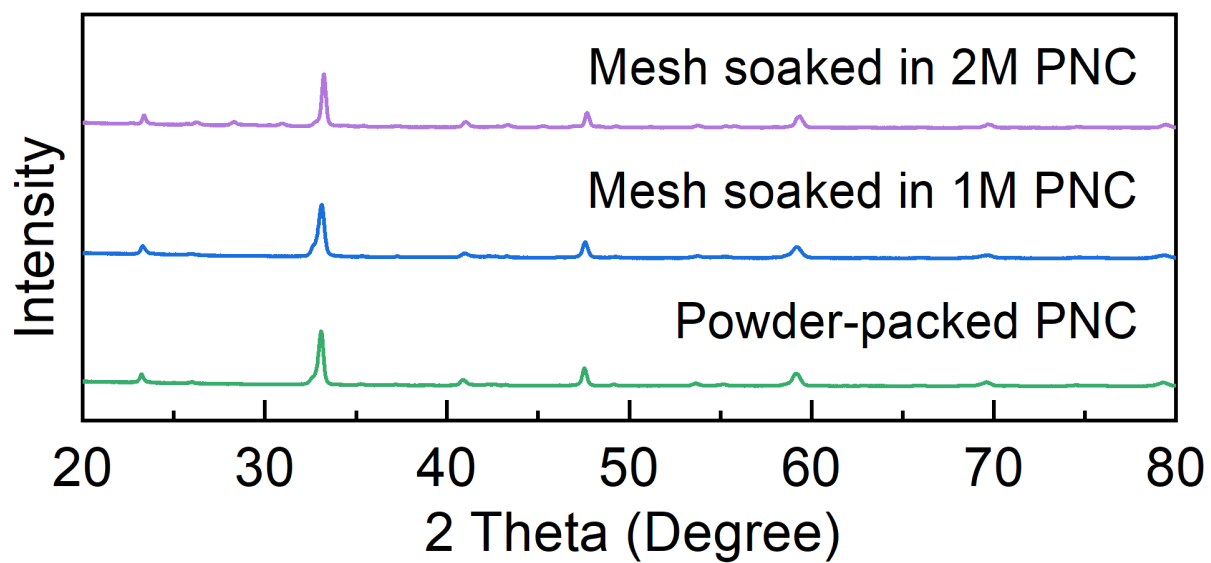


Fig. S4. XRD results of powder-packed PNC electrode. NAUP-PNC mesh-structured electrode soaked in 1M PNC73 solution and soaked in 2M PNC73 solution, respectively. All showed the pure phase.

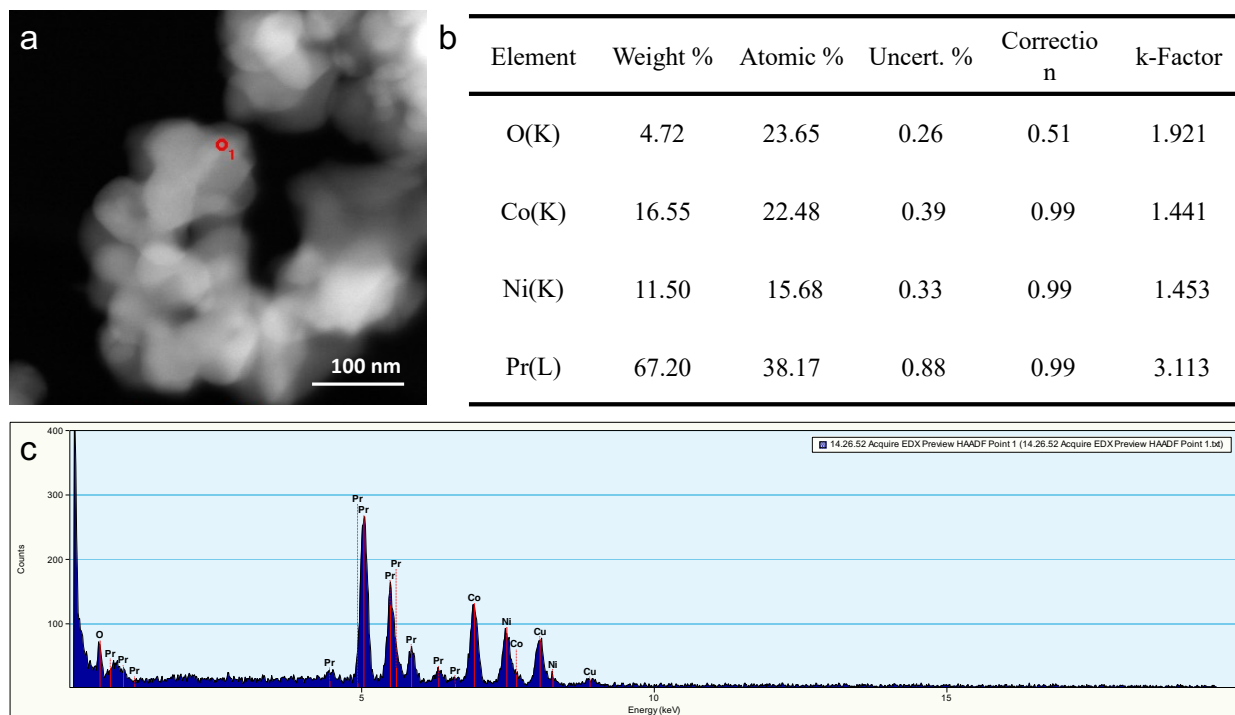
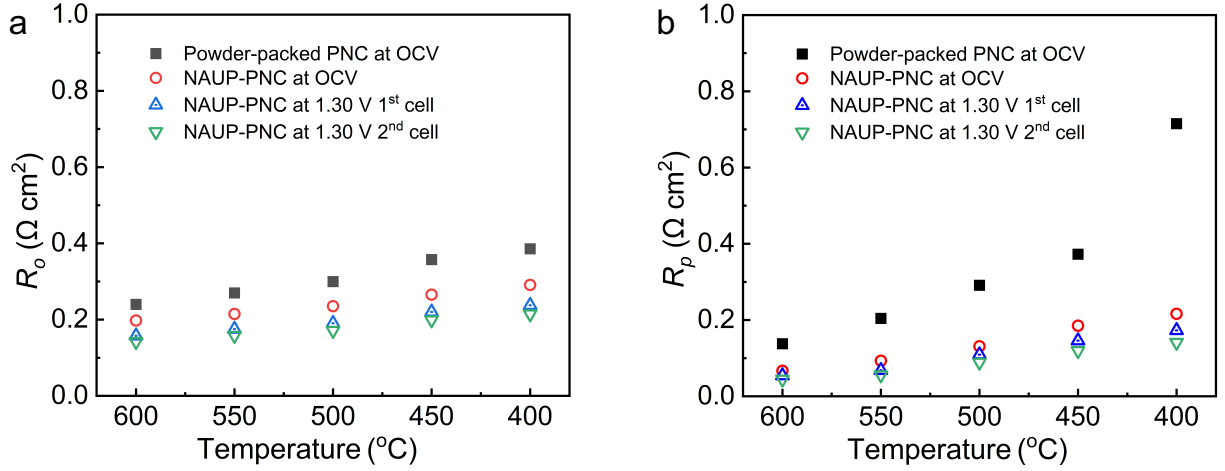


Fig. S5: TEM characterization of NAUP-PNC oxygen electrode. a HAADF image of NAUP-PNC nanoparticles. **b** Table of elemental amounts in percentages on site 1. **c** EDS spectra of HAADF site 1.



c

Powder-packed PNC at OCV	R_o (Ω cm ²)	R_p (Ω cm ²)	Standard deviation for R_p	NAUP-PNC at OCV	R_o (Ω cm ²)	R_p (Ω cm ²)	Standard deviation for R_p	NAUP-PNC at 1.30 V	R_o (Ω cm ²)	R_p (Ω cm ²)	Standard deviation for R_p
600 °C	0.2012	0.1767	0.00174	600 °C	0.1976	0.0668	0.00155	600 °C	0.1577	0.0530	0.00157
550 °C	0.2641	0.3277	0.00146	550 °C	0.2241	0.0840	0.00143	550 °C	0.1752	0.0682	0.00134
500 °C	0.3515	0.7146	0.00132	500 °C	0.2373	0.1278	0.00162	500 °C	0.1904	0.1091	0.00144
450 °C	0.4713	1.6939	0.00142	450 °C	0.2508	0.1997	0.00147	450 °C	0.2202	0.1460	0.00124
400 °C	0.4952	1.9105	0.00159	400 °C	0.2682	0.2387	0.00167	400 °C	0.2376	0.1729	0.00133

Fig. S6. Comparison of R_o (a) and R_p (b) between NAUP-PNC cells and powder-packed PNC cells under both SOFC and SOEC modes at 400-600 °C. c Resistance value with corresponding standard deviations.

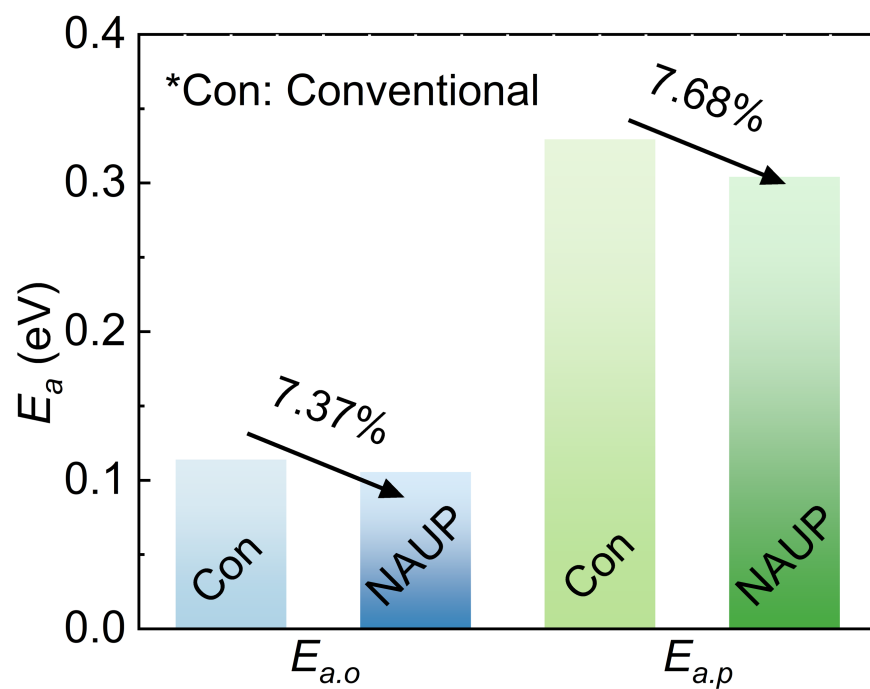


Fig. S7: Comparison between E_a from ohmic resistance and E_a from polarization resistance.

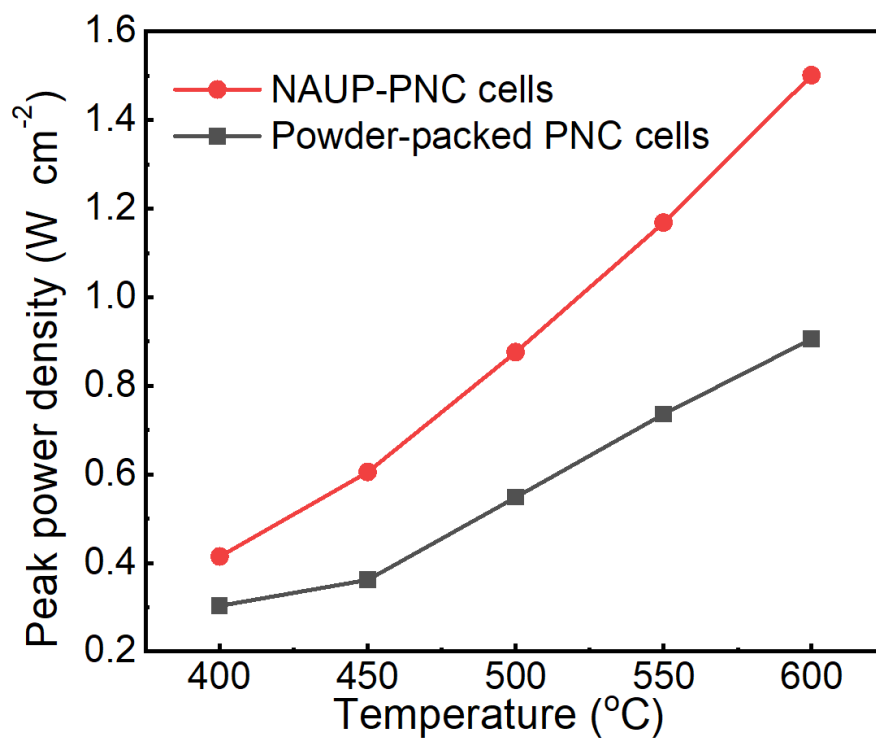


Fig. S8. Comparison of peak power density between NAUP-PNC cells and powder-packed PNC cells under SOFC mode at 400-600 °C.

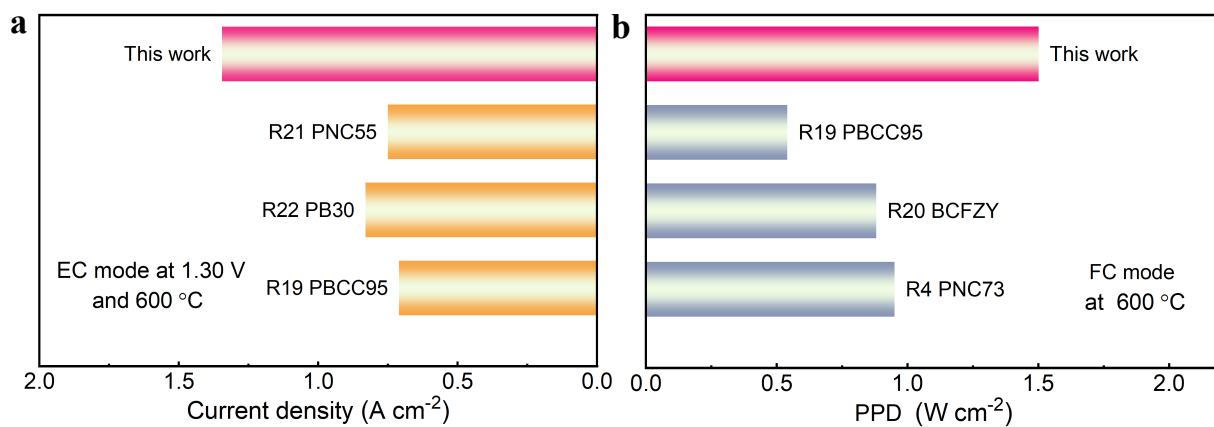
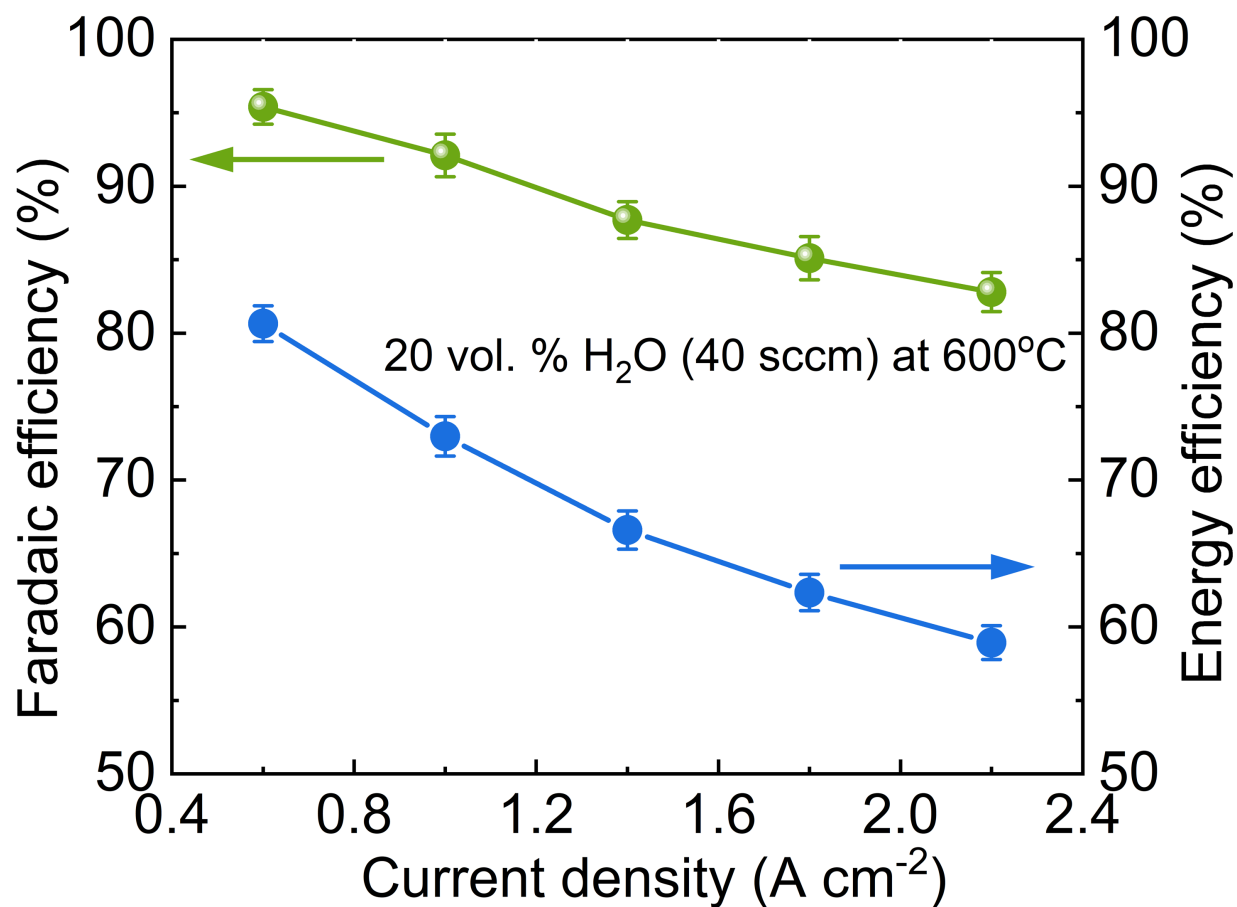


Fig. S9. Comparison of NAUP-PNC73 full cell performances in both FC and EC modes with some recent literatures^{4,19–22}. BCFZY, $\text{BaCo}_{0.4}\text{Fe}_{0.4}\text{Zr}_{0.1}\text{Y}_{0.1}\text{O}_{3-\delta}$; PNC73, $\text{PrNi}_{0.7}\text{Co}_{0.3}\text{O}_{3-\delta}$; PNC55, $\text{PrNi}_{0.5}\text{Co}_{0.5}\text{O}_{3-\delta}$; PB30, $\text{Pr}_{1.7}\text{Ba}_{0.3}\text{NiO}_{4+\delta}$; PBCC95, $(\text{PrBa}_{0.8}\text{Ca}_{0.2})_{0.95}\text{Co}_2\text{O}_{6-\delta}$.



Current density (A cm ⁻²)	FE (%)	Standard deviation of FE	EE (%)	Standard deviation of EE
0.60	95.4	±1.17	80.6	±1.22
1.00	92.1	±1.45	72.9	±1.34
1.40	87.7	±1.25	66.6	±1.30
1.80	85.1	±1.47	62.3	±1.24
2.20	82.8	±1.33	58.9	±1.15

Fig. S10. Faradic efficiency (FE, %) and Energy efficiency (EE, %) of NAUP-PNC full cells in electrolysis mode at 600°C (top). Table of statistical standard deviations for FE and EE (bottom). Each test was conducted for six times to get the statistical standard deviations, presented as error bars.

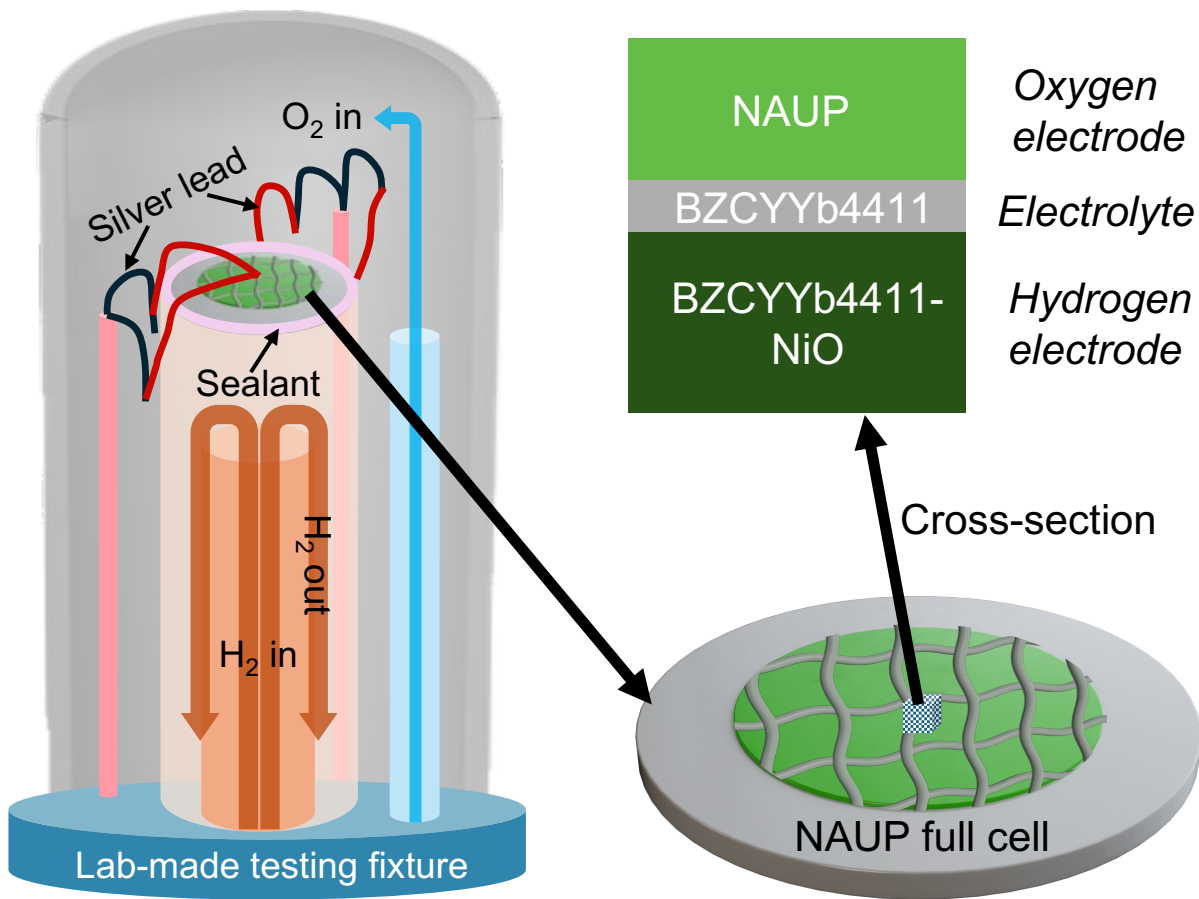


Fig. S11. Lab-made testing fixture used in this work.

PPD (W cm ⁻²)	Temperature (°C)								<i>Experimental condition</i>	Fabrication
	700	650	600	550	500	450	400	350		
This work	-	-	1.50	1.17	0.88	0.61	0.41	-	<i>Under H₂/O₂</i>	NAUP
Ref. 1	-	-	0.36	0.22	0.09	-	-	-	<i>A steam partial pressure of 12%/Pure H₂/FC at 0.7 V</i>	3D self-architected steam electrode
Ref. 2	-	-	0.33	0.20	-	-	-	-	<i>Cathode: 75% CO₂+25%O₂/Anode : Ar</i>	3D architected electrode
Ref. 3	-	-	0.92	-	0.58	-	0.21	-	<i>Cathode: pure O₂/Anode: pure H₂</i>	3D engineering electrode
Ref. 4	-	-	0.95	-	-	-	-	-	<i>Wet H₂ (3% steam) on the fuel side and pure oxygen on the oxygen side</i>	Powder-packed electrode
Ref. 5	-	-	0.65	0.50	0.38	0.24	0.14	0.06	<i>Under H₂/air</i>	Powder-packed electrode
Ref. 6	1.30	0.90	0.40	0.20	-	-	-	-	<i>Under H₂/O₂</i>	Powder-packed electrode
Ref. 7	-	-	0.46	-	-	-	-	-	<i>Under H₂/O₂</i>	Powder-packed electrode
Ref. 8	-	-	-	-	-	-	-	-	-	Powder-packed electrode
Ref. 9	0.40	0.28	0.19	0.11	-	-	-	-	<i>Under H₂/air</i>	Powder-packed electrode
Ref. 10	0.34	-	0.21	-	0.12	-	-	-	<i>Humidified H₂/O₂</i>	Powder-packed electrode
Ref. 11	0.24	0.16	0.10	-	-	-	-	-	<i>Humidified hydrogen (~3% H₂O) /O₂</i>	Powder-packed electrode
Ref. 12	-	-	-	-	0.65	0.34	0.18	-	<i>Humidified hydrogen (~3% H₂O) /air</i>	Nano-network electrode
Ref. 13	0.80	-	-	-	-	-	-	-	<i>Under H₂/O₂</i>	Nano-tailoring electrode
Ref. 14	1.37	-	-	-	-	-	-	-	<i>Humidified hydrogen (~6% H₂O) /air</i>	Nanocatalyst electrode
Ref. 15	0.70	-	-	-	-	-	-	-	<i>Humidified hydrogen (~3% H₂O) /air</i>	Infiltrated nano-electrode
Ref. 16	0.49	0.38	0.29	-	-	-	-	-	<i>Humidified hydrogen (~3% H₂O) /air</i>	Infiltrated multiscale electrode
Ref. 17	-	-	0.15	-	-	-	-	-	<i>Humidified hydrogen (~3% H₂O) /air</i>	Nanostructured electrode
Ref. 18	0.72	0.68	-	-	-	-	-	-	<i>Humidified hydrogen (~3% H₂O) /air</i>	Nanostructured electrode

Table. S1. Comparison of peak power density between this work and other literature results.

R_p (Ω cm^2)	Temperature ($^{\circ}\text{C}$)								Experimental condition	Fabrication
	700	650	600	550	500	450	400	350		
This work	-	-	0.07	0.09	0.13	0.19	0.22	-	Under H_2/O_2 /at OCV	NAUP
Ref. 1	-	-	0.72	1.13	1.86	-	-	-	A steam partial pressure of 12%/Pure H_2 /FC at OCV	3D self- architected steam electrode
Ref. 2	-	-	1.55	2.10	2.57	-	-	-	Cathode: 75% $\text{CO}_2 + 25\% \text{O}_2$ /Anode : Ar/at OCV	3D architected electrode
Ref. 3	-	-	-	-	-	-	0.71	-	Cathode: pure O_2 /Anode: pure H_2 /at OCV	3D engineering electrode
Ref. 4	-	-	0.38	-	-	-	-	-	Wet O_2 (3% steam) at OCV	Powder-packed electrode
Ref. 5	-	-	-	-	-	-	-	-	-	Powder-packed electrode
Ref. 6	-	-	-	-	-	-	-	-	-	Powder-packed electrode
Ref. 7	-	-	0.44	-	-	-	-	-	Under H_2/O_2 /at OCV	Powder-packed electrode
Ref. 8	0.31	-	0.40	-	-	-	-	-	Steam in anode: 0.4 atm	Powder-packed electrode
Ref. 9	0.17	-	-	2.12	-	-	-	-	Under H_2 /air /at OCV	Powder-packed electrode
Ref. 10	0.62	-	0.81	-	-	-	-	-	Humidified H_2/O_2 /at OCV	Powder-packed electrode
Ref. 11	0.32	-	2.09	-	-	-	-	-	Humidified hydrogen (~3% H_2O)/ O_2 /at OCV	Powder-packed electrode
Ref. 12	-	-	-	-	0.25	0.42	1.06	-	Humidified hydrogen (~3% H_2O)/air /at OCV	Nano-network electrode
Ref. 13	-	-	-	-	-	-	-	-	-	Nano-tailoring electrode
Ref. 14	0.24	-	-	-	-	-	-	-	Humidified hydrogen (~6% H_2O)/air /at OCV	Nanocatalyst electrode
Ref. 15	-	-	-	0.67	-	-	-	-	Humidified hydrogen (~3% H_2O)/air /at OCV	Infiltrated nano- electrode
Ref. 16	0.06	0.16	0.39	-	-	-	-	-	Humidified hydrogen (~3% H_2O)/air /at OCV	Infiltrated multiscale electrode
Ref. 17	-	-	-	-	-	-	-	-	-	Nanostructured electrode
Ref. 18	0.99	1.63	-	-	-	-	-	-	Humidified hydrogen (~3% H_2O)/air /at OCV	Nanostructured electrode

Table. S2. Comparison of R_p between this work and other literature results.

R_o	E_a (eV)	Standard deviation
Powder-packed PNC at OCV	0.1139	± 0.01399
NAUP-PNC at OCV	0.1055	± 0.01197
NAUP-PNC at 1.30 V 1 st cell	0.1059	± 0.01323
NAUP-PNC at 1.30 V 2 nd cell	0.1064	± 0.01378

Table. S3. Calculated $E_{a,o}$ with standard errors of NAUP-PNC cells and powder-packed PNC cells under both SOFC and SOEC modes.

R_p	E_a (eV)	Standard deviation
Powder-packed PNC at OCV	0.32935	± 0.01807
NAUP-PNC at OCV	0.30406	± 0.01371
NAUP-PNC at 1.30 V 1 st cell	0.30768	± 0.01742
NAUP-PNC at 1.30 V 2 nd cell	0.30817	± 0.01728

Table. S4. Calculated $E_{a,p}$ with standard errors of NAUP-PNC cells and powder-packed PNC cells under both SOFC and SOEC modes.

NAUP-PNC cells					
Temperature (°C)	400	450	500	550	600
Peak power density (W cm ⁻²)	0.4140	0.6055	0.8762	1.1684	1.5012
Powder-packed PNC cells					
Temperature (°C)	400	450	500	550	600
Peak power density (W cm ⁻²)	0.3031	0.3619	0.5481	0.7364	0.9056

Table. S5. Comparison of peak power density between NAUP-PNC cells (top) and powder-packed PNC cells (bottom) under SOFC mode at 400-600 °C.

NAUP-PNC cells					
Temperature (°C)	400	450	500	550	600
Current density (A cm ⁻²)	-0.6512	-1.5245	-2.4985	-3.8627	-5.0423
Powder-packed PNC cells					
Temperature (°C)	400	450	500	550	600
Current density (A cm ⁻²)	-0.4932	-1.1012	-1.5321	-2.6021	-3.3605

Table. S6. Comparison of current density of NAUP-PNC cells (top) and powder-packed PNC cells (bottom) under SOEC mode at 400-600 °C.

Relative humidity (vol.% H ₂ O)	3	10	20	30
Current density (A cm ⁻²)	-4.0265	-4.5124	-5.0397	-4.7397
Gas flow rate (sccm)	10	20	40	60
Current density (A cm ⁻²)	-4.2151	-4.6001	-5.0258	-4.8225

Table. S7. Comparison of current density of NAUP-PNC cells in terms of relative humidity (top) and gas flow rate (bottom) under SOEC mode at 600 °C.

PPD (W cm ⁻²)				
Ref.	R4 PNC73	R20 BCFZY	R19 PBCC95	This work
Conditions	H ₂ -Humidified O ₂ (50% H ₂ O)	Humidified H ₂ (3% H ₂ O) -Humidified O ₂ (3% H ₂ O)	Humidified H ₂ (3% H ₂ O) -O ₂	H ₂ -Humidified O ₂ (3% H ₂ O)
PPD (W cm ⁻²)	0.95	0.88	0.54	1.50

CD at 1.30V (A cm ⁻²)				
Ref.	R22 PB30	R19 PBCC95	R21 PNC55	This work
Conditions	H ₂ - Humidified O ₂ (60% H ₂ O)	Humidified H ₂ (3% H ₂ O) -Humidified O ₂ - (20% H ₂ O)	Humidified H ₂ (3% H ₂ O) -Humidified O ₂ (50% H ₂ O)	H ₂ - Humidified O ₂ (20% H ₂ O)
CD at 1.30V (A cm ⁻²)	0.83	0.71	0.75	1.35

Table. S8. Comparison of peak power density and current density for the reference data in Fig. S9.

X th	$R_o (\Omega \text{ cm}^2)$	$R_t (\Omega \text{ cm}^2)$	$R_p (\Omega \text{ cm}^2)$
1st	0.1577	0.2099	0.05220
2nd	0.1579	0.2084	0.05055
3rd	0.1582	0.2098	0.05158
4th	0.1586	0.2116	0.05304
5th	0.1592	0.2095	0.05029
6th	0.1583	0.2085	0.05026
7th	0.1572	0.2093	0.05214
8th	0.1576	0.2084	0.05083
9th	0.1574	0.2081	0.05061
10th	0.1558	0.2091	0.05330
11th	0.1554	0.2099	0.05450
12th	0.1571	0.2095	0.05235
13th	0.1563	0.2100	0.05368
14th	0.1535	0.2095	0.05599
15th	0.1535	0.2095	0.05596
16th	0.1548	0.2095	0.05465
17th	0.1524	0.2109	0.05851
18th	0.1547	0.2079	0.05313
19th	0.1586	0.2116	0.05304
20th	0.1554	0.2087	0.05336

Table. S9. Results of R_o , R_t and R_p of NAUP-PNC cells at 1.30 V and 600 °C for 20 cycles.

Time (h)	Current density (A cm^{-2})			Mean %
	-Value	Difference	% of degradation	
0	1.2889			
10	1.2763	0.0126	0.98	
20	1.2632	0.0131	1.03	
30	1.2499	0.0133	1.05	1.03
40	1.2369	0.0130	1.04	
50	1.2239	0.0130	1.05	
Altering applied voltage from 1.30 V to 1.40 V				
50	1.9958			
60	1.9729	0.0230	1.15	
70	1.9482	0.0247	1.25	
80	1.9242	0.0240	1.23	1.24
90	1.9002	0.0241	1.25	
100	1.8753	0.0249	1.31	

Table. S10. % of degradation calculation on current density of NAUP-PNC cells at 1.30 V/1.40 V and 600 °C for 100 hours.

Time (h)	Current density (A cm^{-2})			Mean %
	Mode	Standard deviation	% of degradation	
0-10	SOEC	0.02036	1.69	1.87
20-30		0.02168	1.83	
40-50		0.02244	1.93	
60-70		0.02144	1.88	
80-90		0.02204	1.97	
10-20	SOFC	0.02823	1.48	1.55
30-40		0.02969	1.58	
50-60		0.02977	1.61	
70-80		0.02893	1.59	
90-100		0.02668	1.49	

Table. S11. % of degradation calculation on current density of NAUP-PNC cells for reversible tests at 600 °C for 100 hours.

Xth	Current density (A cm ⁻²)			Mean %
	Voltage (V)	Standard deviation	% of degradation	
1	1.50	0.05634	1.85	1.94
2		0.05410	1.81	
3		0.05605	1.91	
4		0.05239	1.82	
5		0.05257	1.86	
6		0.05104	1.84	
7		0.05935	2.18	
8		0.05620	2.11	
9		0.05423	2.08	
10		0.04902	1.92	
11		0.04732	1.89	
12		0.04889	1.99	
13		0.04623	1.92	

Table. S12. % of degradation calculation on current density of NAUP-PNC cells for transient tests (step-voltage based) at 1.50 V and 600 °C.

Xth	Current density (A cm ⁻²)			Mean %
	Voltage (V)	Standard deviation	% of degradation	
1&2	1.35	0.02270	1.35	1.28
3&4		0.02273	1.37	
5&6		0.02176	1.33	
7&8		0.02164	1.34	
9&10		0.02023	1.27	
11&12		0.01903	1.21	
13&14		0.01989	1.28	
15&16		0.01856	1.21	
17&19		0.01955	1.29	
19&20		0.01959	1.31	
21&22		0.01757	1.19	
23&24		0.01823	1.25	

Table. S13. % of degradation calculation on current density of NAUP-PNC cells for transient tests (step-voltage based) at 1.35 V and 600 °C.

Xth	Current density (A cm ⁻²)			Mean %
	Voltage (V)	Standard deviation	% of degradation	
1	1.20	0.00327	0.45	0.45
2		0.00304	0.42	
3		0.00303	0.42	
4		0.00352	0.49	
5		0.00343	0.48	
6		0.00313	0.44	
7		0.00318	0.45	
8		0.00303	0.43	
9		0.00358	0.51	
10		0.00272	0.39	
11		0.00306	0.44	
12		0.00311	0.45	

Table. S14. % of degradation calculation on current density of NAUP-PNC cells for transient tests (step-voltage based) at 1.20 V and 600 °C.

Xth	Current density (A cm ⁻²)			Mean %
	Voltage (V)	Standard deviation	% of degradation	
1		0.00722	0.24	
2		0.04665	1.56	
3		0.00393	0.13	
4		0.03728	1.26	
5		0.03791	1.30	
6		0.01210	0.41	
7		0.05000	1.69	
8	1.50	0.03888	1.33	1.07
9		0.03888	1.32	
10		0.07500	2.57	
11		0.04155	1.39	
12		0.03321	1.13	
13		0.00118	0.04	
14		0.01784	0.60	
15		0.03333	1.12	

Table. S15. % of degradation calculation on current density of NAUP-PNC cells for transient tests (time-interval based) at 1.50 V and 600 °C.

Xth	Current density (A cm^{-2})			Mean %
	Voltage (V)	Standard deviation	% of degradation	
1	1.20	0.00747	0.75	4.11
2		0.00333	0.33	
3		0.02334	2.34	
4		0.00666	0.68	
5		0.06167	6.38	
6		0.08500	9.39	
7		0.00500	0.51	
8		0.06833	6.87	
9		0.05000	5.40	
10		0.01333	1.36	
11		0.08500	8.59	
12		0.05333	5.89	
13		0.04000	4.17	
14		0.08666	8.68	
15		0.04000	4.39	
16		0.02916	3.06	

Table. S16. % of degradation calculation on current density of NAUP-PNC cells for transient tests (time-interval based) at 1.20 V and 600 °C.

Voltage (V)	% of degradation	Mean %
1.50 V	1.07	2.59
1.20 V	4.11	

Table. S17. Mean % of degradation calculation on current density of NAUP-PNC cells for transient tests (time-interval based) at 600 °C.

<i>X</i> th	CD at SOFC (A cm ²)	CD at SOEC (A cm ²)	Temperature (°C)
1st	4.68322	-4.87872	600
2nd	3.34564	-2.94545	550
3rd	3.09026	-2.40625	500
4th	3.28819	-2.89412	550
5th	4.58335	-4.78633	600
6th	3.21531	-2.76239	550
7th	2.92903	-1.90668	500
8th	2.95254	-2.49223	550
9th	4.4722	-4.78633	600
10th	2.78259	-2.32329	550
11th	2.76781	-1.7373	500
12th	2.66128	-2.20302	550
13th	4.47308	-4.67502	600
14th	2.52947	-2.06924	550
15th	2.40252	-1.1122	500

Table. S18. Results of maximum current density of NAUP-PNC cells for thermal cycling tests.

X th	R_o (Ω cm ²)	R_t (Ω cm ²)	R_p (Ω cm ²)	Standard deviation for R_p	Temperature (°C)
1st	0.16555	0.22275	0.05720	0.00134	600
2nd	0.18043	0.25075	0.07032	0.00115	550
3rd	0.19417	0.30703	0.11286	0.00107	500
4th	0.18049	0.25687	0.07638	0.00129	550
5th	0.16531	0.22604	0.06073	0.00135	600
6th	0.1806	0.2615	0.08090	0.00147	550
7th	0.19399	0.31509	0.12110	0.00151	500
8th	0.18176	0.26166	0.07990	0.00147	550
9th	0.16625	0.22853	0.06228	0.00132	600
10th	0.18162	0.2627	0.08108	0.00132	550
11th	0.19703	0.32481	0.12778	0.00128	500
12th	0.18127	0.26595	0.08468	0.00121	550
13th	0.16689	0.23154	0.06465	0.00145	600
14th	0.18162	0.26827	0.08665	0.00133	550
15th	0.19725	0.33375	0.13650	0.00129	500

Table. S19. Results of R_o , R_t and R_p of NAUP-PNC cells at 1.30 V for thermal cycling tests.

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