Supporting Information

Redox behavior of the SOFC electrode candidate $NdBaMn_2O_{5+\delta}$ investigated by high-temperature *in situ* neutron diffraction: first real-time characterisation of an *Ln*$BaMn_2O_{5.5}$ intermediate phase

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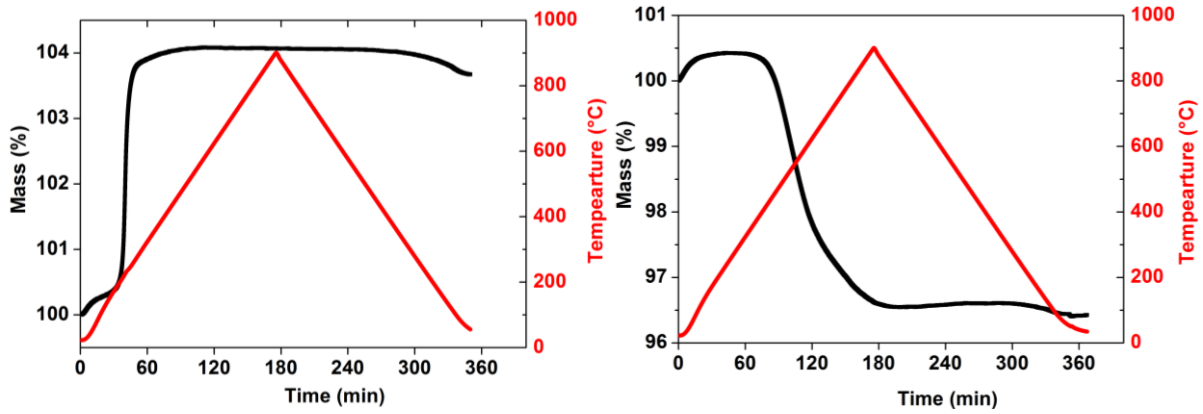

Figure S1. TGA of $NdBaMn_2O_{5+\delta}$ (left) under air (flow rate 40 mL min^{-1}) and (right) subsequent cycle under 5% H_2/N_2 (flow rate 20 mL min^{-1}); heating and cooling rates are 5 °C min^{-1}.

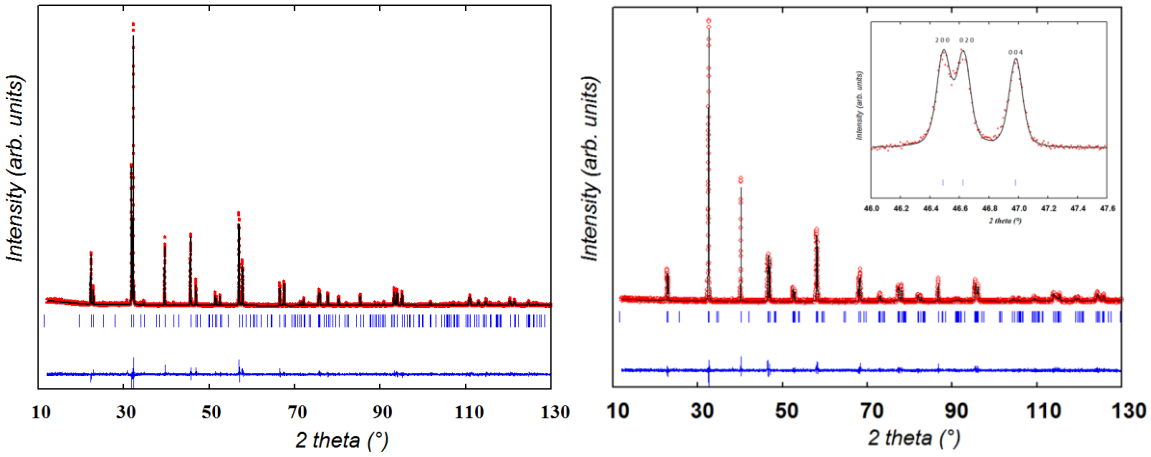

Figure S2. Rietveld refinement plot of the X-ray diffraction data of (let) as-prepared $NdBaMn_2O_5$, S.G. *P*4/*nmm* and (right) $NdBaMn_2O_6$ (S.G. *Pmmm*) part of which is expanded in the inset to highlight the (200/020) doublet.

Table S1. Refined lattice parameters, coordinates, site occupancies and isotropic thermal vibration parameters B_{iso} (Å^2) for $NdBaMn_2O_5$ at 800 °C (NPD, three-phase refinement, cycle 3) in the space group *P4/mmm*; the occupancy of O1 was fixed at 0, B_{iso} for O2 and O3 were contained to be equal.

Atom		
	a (Å)	4.0028(2)
	c (Å)	7.8471(6)
	wt. %	39
Nd	B_{iso}	1.57(7)
Ba	B_{iso}	1.56(9)
Mn	*z*	0.2435(9)
	B_{iso}	1.7(1)
O3	*z*	0.1961(4)
	B_{iso} (O)	2.98(6)
	χ^2	1.11
	R_B %	3.8

Table S2. Refined lattice parameters, coordinates, site occupancies and isotropic vibration parameters B_{iso} (Å^2) for $NdBaMn_2O_6$ at 800 °C (NPD, three-phase refinement, cycle 3) in the space group *Pmmm*, the occupation for the O sites was fixed at 1, B_{iso} for the oxygen sites were constrained to be equal, z coordinate for O2 and O3 were constrained to be equal.

Atom		
	a (Å)	3.9445(2)
	b (Å)	3.9458(4)
	c (Å)	7.8239(5)
	wt. %	36
Nd	B_{iso}	1.57(7)
Ba	B_{iso}	1.569)
Mn	*z*	0.248(1)
	B_{iso}	1.00(6)
O3, O4	*z*	0.2285(5)
	B_{iso} (O)	2.45(4)
	χ^2	1.11
	R_B %	2.6

Table S3. Refined lattice parameters, coordinates, site occupancies and isotropic thermal vibration parameters B_{iso} (Å^2) for $NdBaMn_2O_5$ at 65 °C (NPD, two-phase refinement, cycle 3) in the space group *P4/nmm*; B_{iso} for the O sites were contained to be equal and the occupancy of O2 and O3 has been fixed at 1.

Atom		
	a (Å)	5.6164(1)
	c (Å)	7.7495(3)
	wt. %	68
Nd	B_{iso}	0.49(2)
Ba	B_{iso}	0.56(4)
Mn1	*z*	0.2676(9)
	B_{iso}	0.3(1)
Mn2	*z*	0.748(1)
	B_{iso}	0.4(1)
O1	*occ*	0.02(1)
O2	*z*	0.0078(9)
O3	*x*	0.4923(3)
	z	0.3084(1)
	B_{iso} (O)	0.88(2)
	χ^2	1.16
	R_B %	2.7