Electronic supplementary information

Understanding phase stability of yttria stabilized zirconia electrolyte under solid oxide electrolysis cell operation condition

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The derivation of van der Pauw's technique for measuring resistivity is described as follows:

$R_A = R_{21,34} + R_{12,43} + R_{43,12} + R_{34,21}$ (1)

$R_B = R_{32,41} + R_{23,14} + R_{14,23} + R_{41,32}$ (2)

$e^{\frac{-\pi R_A}{R_S}} + e^{\frac{-\pi R_B}{R_S}} = 1$ (3)

$\rho = R_S \times t$ (4)

$R_{ab,cd}$ indicates that a and b represent the I^+ and I^-, respectively, while c and d represent the V^+ and V^-. The resistivity of the sample can be calculated using equation (4), where t denotes the sample's thickness. For in-plane measurements, the conductivity was measured using the van der Pauw technique, which is commonly employed for samples with uniform thickness. This method is well-suited for measuring the conductivity of our thick film specimens, which have a consistent thickness.

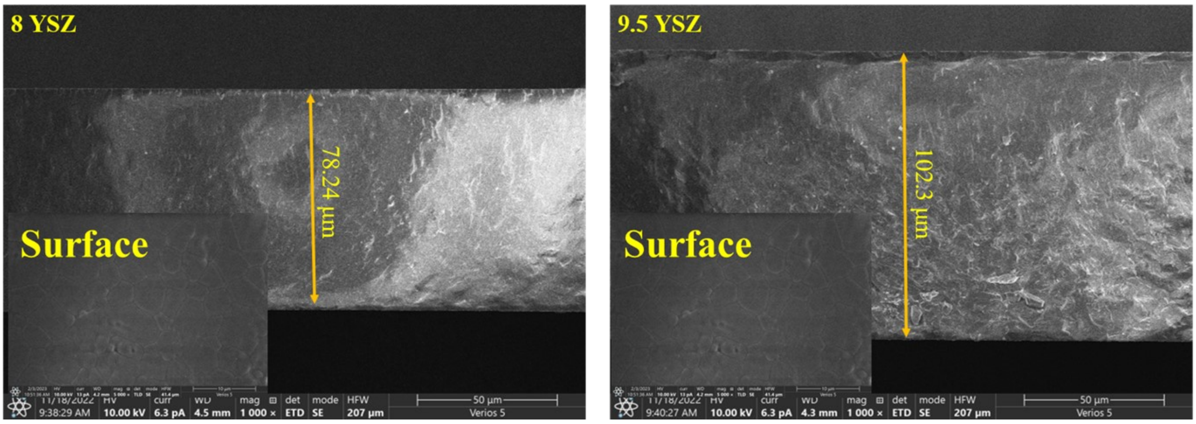

Fig. S1 SEM images of 8YSZ and 9.5YSZ.

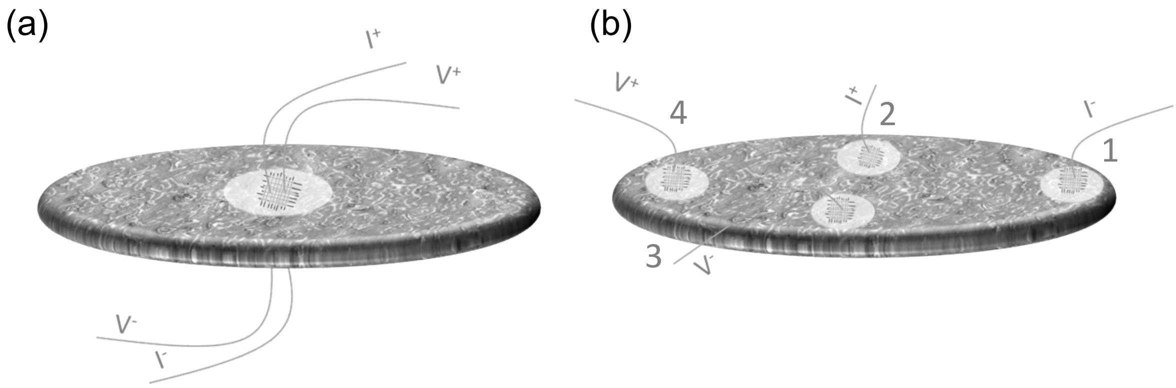

Fig. S2 The configuration of electrodes in the (a) across-plane and (b) van-der Pauw method.

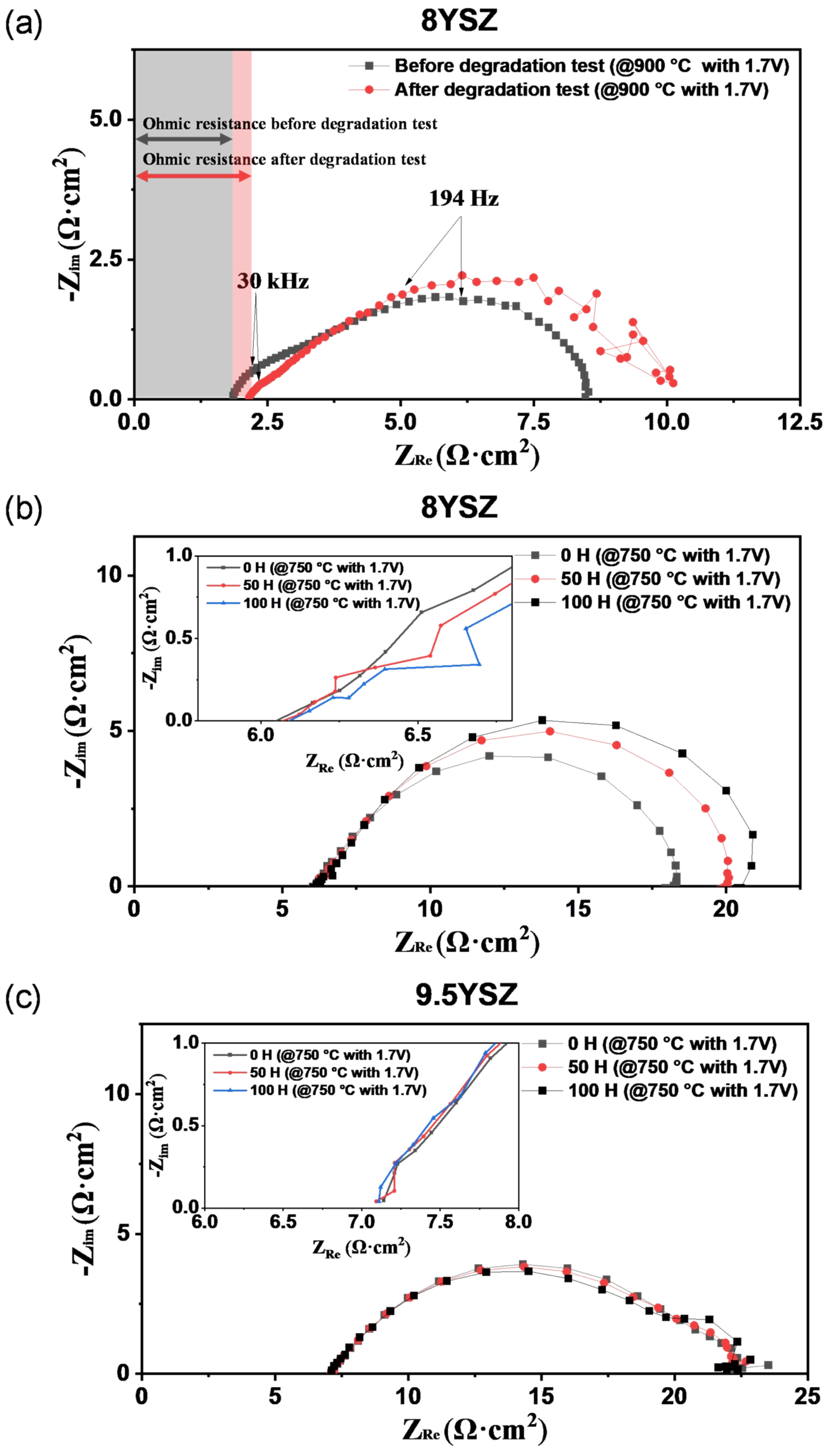

Fig. S3 (a) Variations in ohmic resistance before and after the 900 ℃ degradation test with 1.7V and (b, c) changes in impedance spectroscopy over time at 750 ℃ with 1.7V.

Temperature (1000/T(K^{-1}))	Conductivity measured by in-plane method ($log\sigma(S \cdot cm^{-1})$)		Conductivity measured by across-plane method ($log\sigma(S \cdot cm^{-1})$)	
	8YSZ	9.5YSZ	8YSZ	9.5YSZ
0.85241	-0.968	-1.00	-1.15	-1.12
0.89035	-1.13	-1.16	-1.28	-1.31
0.93184	-1.29	-1.33	-1.44	-1.45
0.97737	-1.49	-1.55	-1.64	-1.72
1.02759	-1.71	-1.78	-1.87	-1.95
1.08325	-1.98	-2.06	-2.14	-2.19
1.14528	-2.29	-2.35	-2.49	-2.44
1.21485	-2.62	-2.76	-2.79	-2.82
1.29341	-2.94	-3.13	-3.09	-3.21

Table S1 Conductivities measured using the in-plane and across-plane methods as a function of temperature.

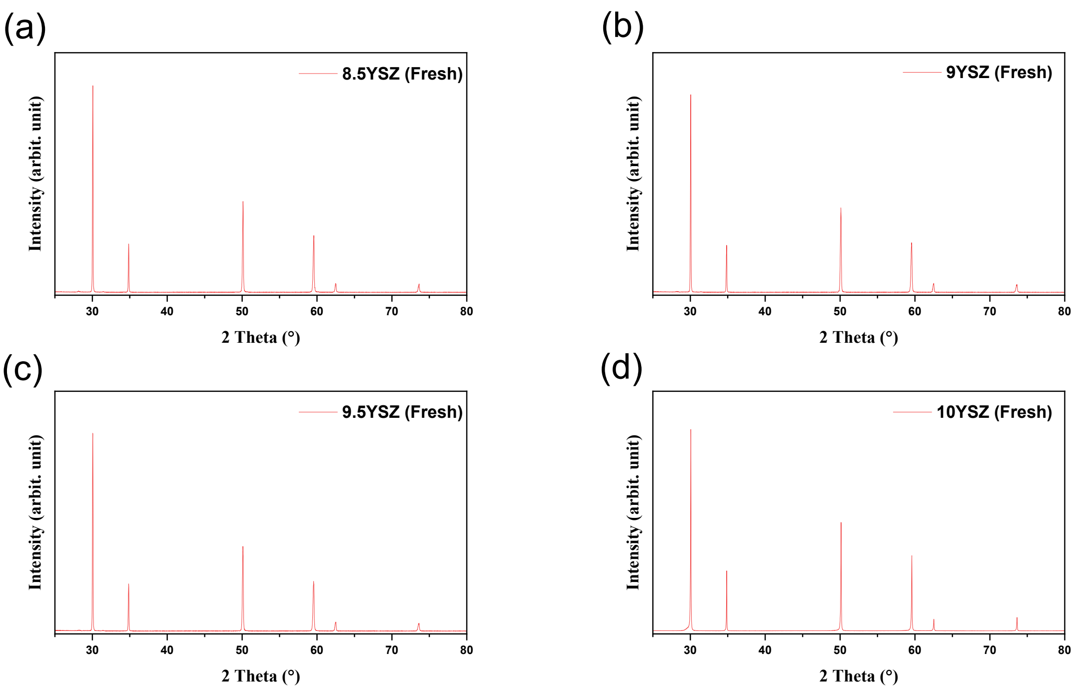

Fig. S4 XRD data of pristine 8.5YSZ, 9YSZ, 9.5YSZ and 10YSZ.

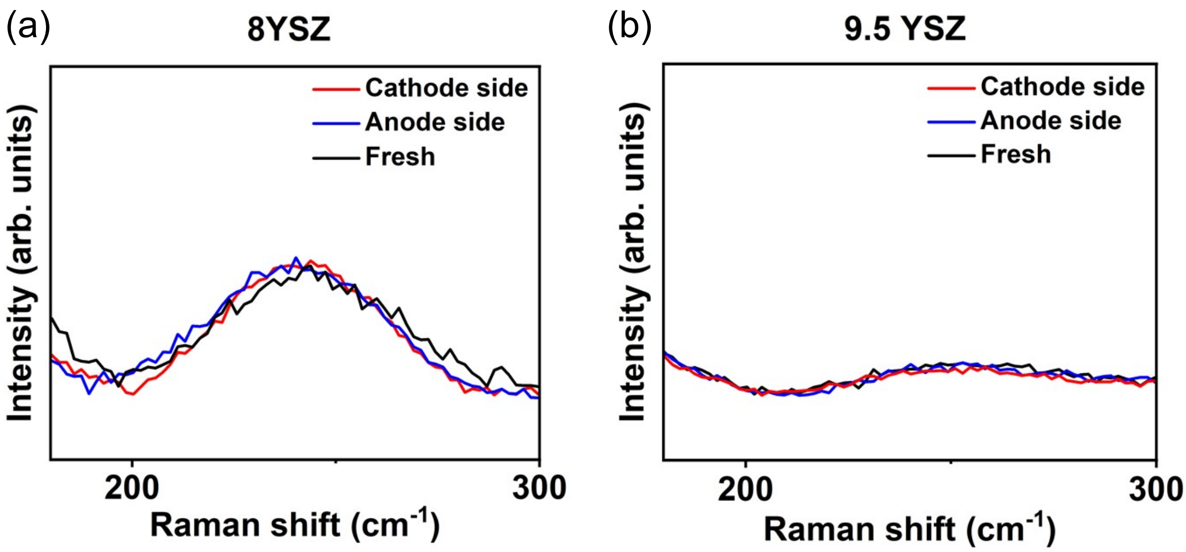

Fig. S5 Raman spectra of (a) 8YSZ and (b) 9.5YSZ at 750 ℃ under 1.7V for 100 hours.

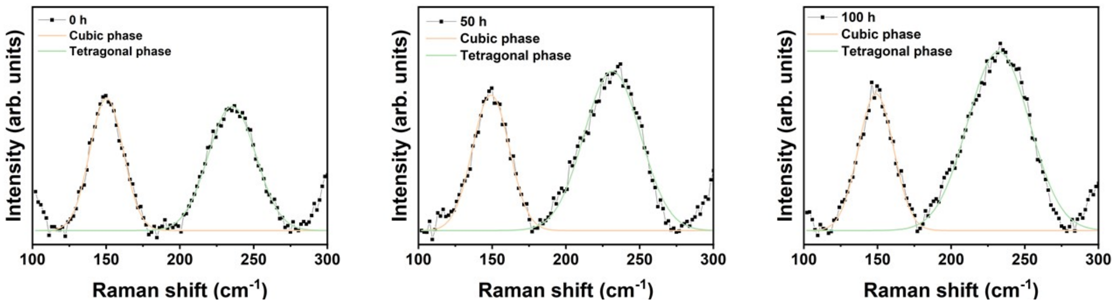

Fig. S6 Raman spectra indicating the variation in the tetragonal 8YSZ ratio with increasing annealing time.

	8YSZ				9.5YSZ		
	Centroid coordinates (at%)		Zr/Y		Centroid coordinates (at%)		Zr/Y
	Zr	Y			Zr	Y	
Fresh	**36.74**	**7.14**	**5.15**	**Fresh**	**32.53**	**7.61**	**4.27**
	27.39	**8.25**	**3.32**		**40.26**	**6.66**	**6.05**
	46.28	**5.84**	**7.92**		**24.05**	**7.48**	**3.21**
Anode side	**37.19**	**5.88**	**6.32**	**Anode side**	**30.00**	**10.50**	**2.86**
	29.93	**6.17**	**4.85**		**46.65**	**7.61**	**6.13**
	47.08	**6.72**	**7.00**		**38.26**	**9.08**	**4.21**
Cathode side	**31.27**	**5.13**	**6.10**	**Cathode side**	**39.63**	**10.15**	**3.90**
	25.66	**5.39**	**4.76**		**30.19**	**10.77**	**2.80**
	37.89	**4.79**	**7.90**		**48.26**	**8.47**	**5.70**

Table S2 Centroid coordinates of Zr and Y for samples.

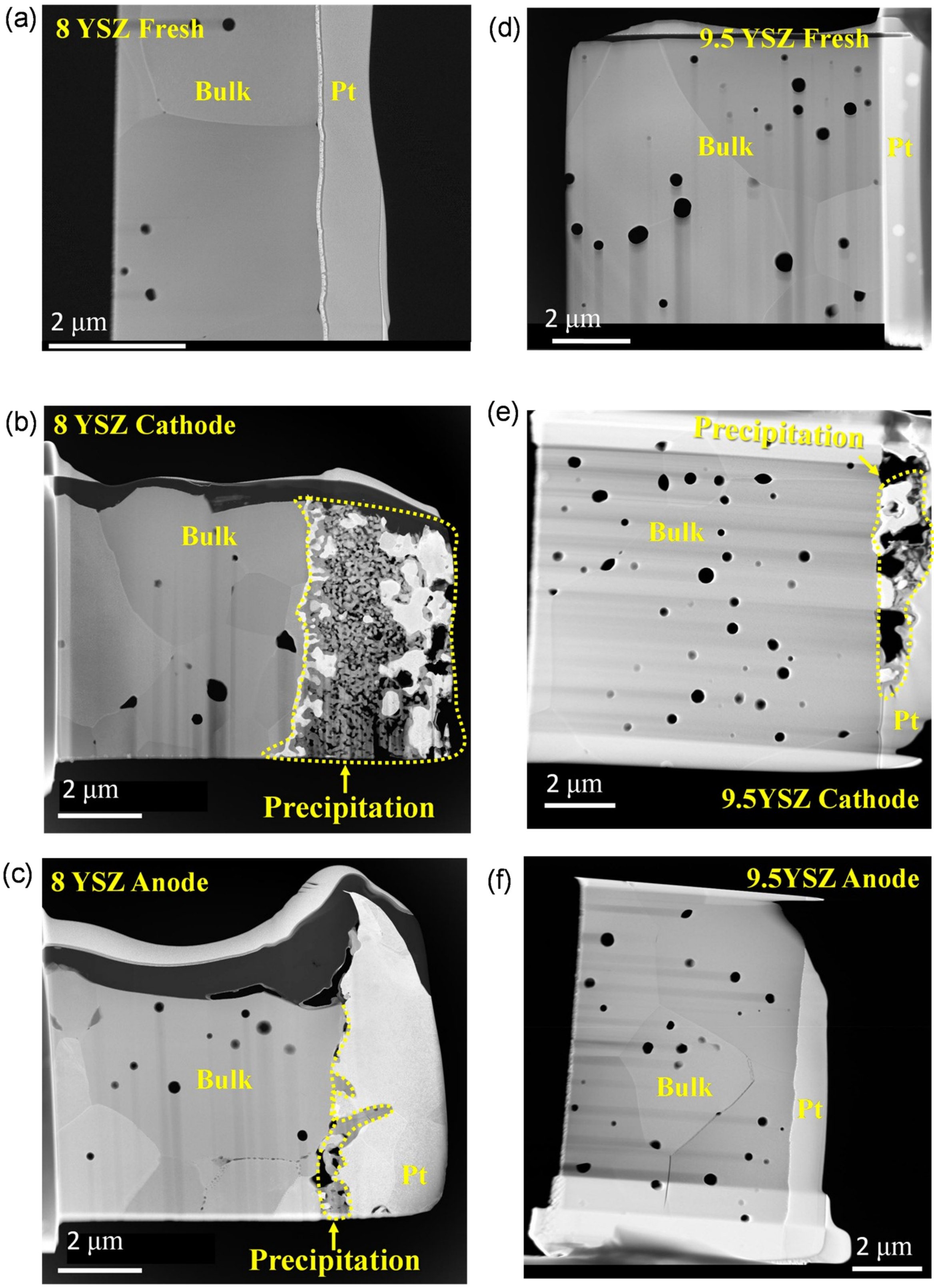

Fig. S7 STEM images of (a) 8YSZ fresh, (b) 8YSZ cathode side, (c) 8YSZ anode side, (d) 9.5YSZ fresh, (e) 9.5YSZ cathode side and (f) 9.5YSZ anode side.

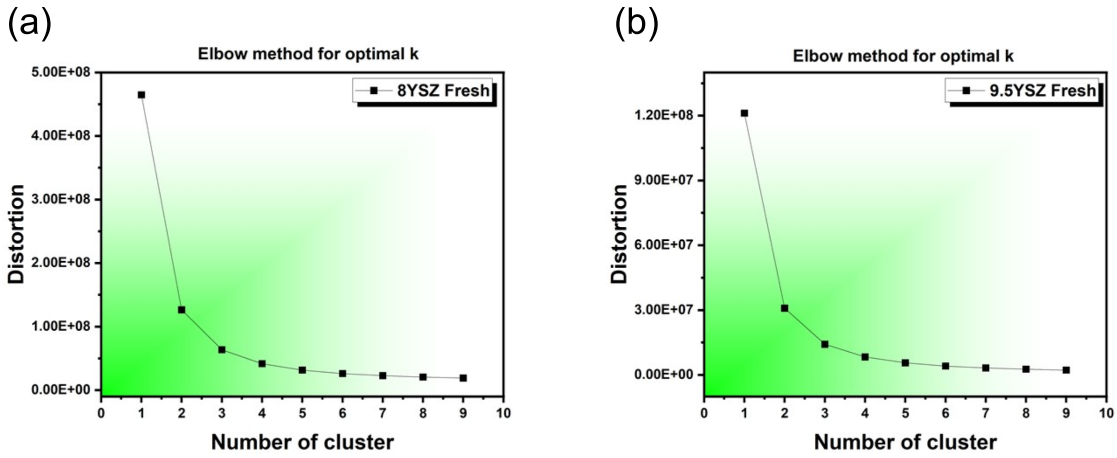

Fig. S8 Elbow method for optimal k values of (a) 8YSZ and (b) 9.5YSZ fresh samples.

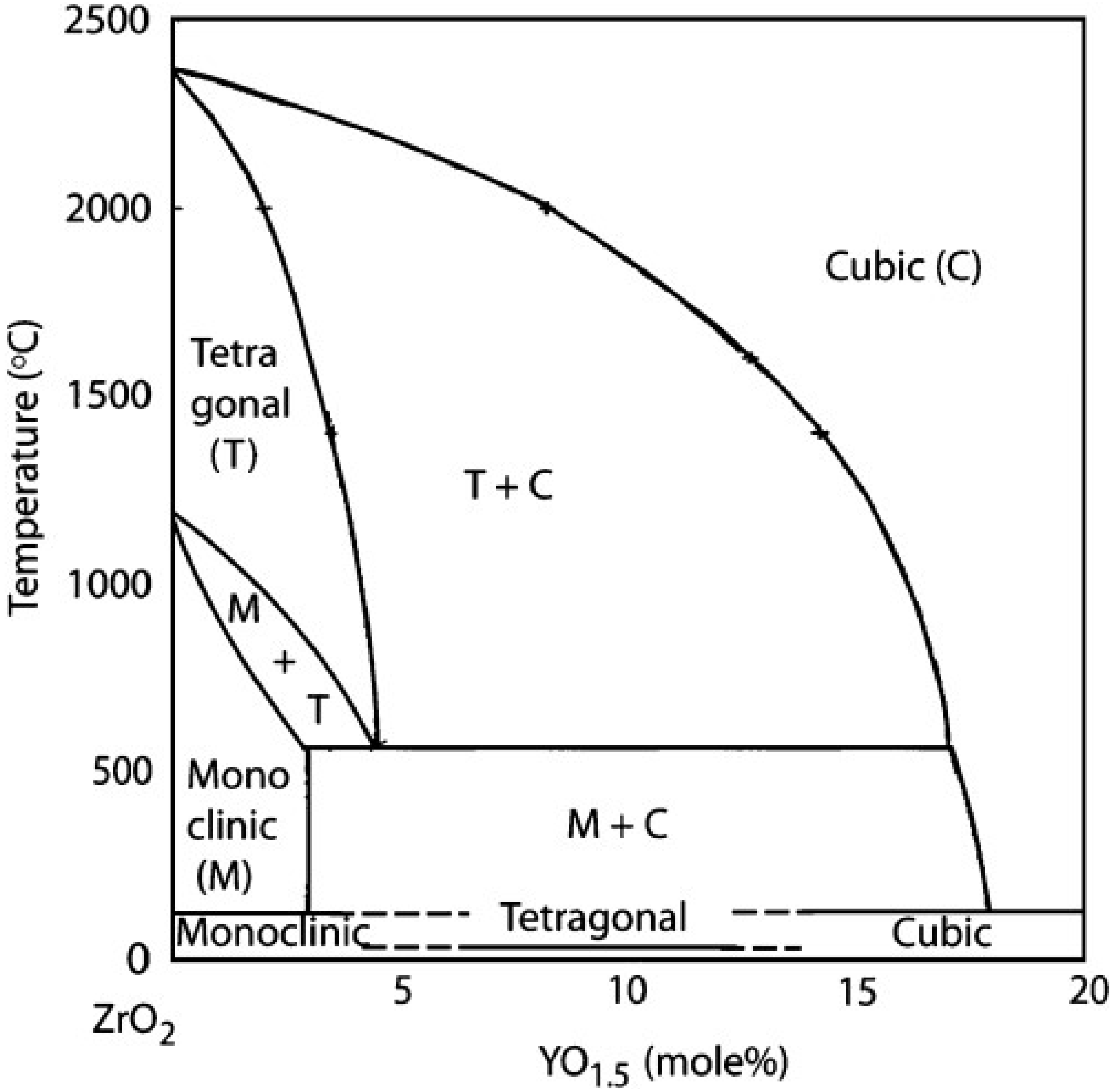

Fig. S9 Y_2O_3-ZrO_2 phase diagram.[41] Copyright 2007, American Chemical Society.