Supporting Information for

Chemical-Substitution-Induced Successive Symmetry Descent and Structure-Property Correlation Investigation for "114" Oxides $CaBa_{1-x}Sr_xZn_2Al_2O_7$

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Table S1. Comparison of reliable factors and isotropic thermal factors for $CaBa_{1-x}Sr_xZn_2Al_2O_7$ (x = 0.2, 0.4, and 0.6) refined with different structure models.

	$CaBa_{0.8}Sr_{0.2}Zn_2Al_2O_7$			$CaBa_{0.6}Sr_{0.4}Zn_2Al_2O_7$			$CaBa_{0.4}Sr_{0.6}Zn_2Al_2O_7$		
Refinement Models	$P6_3mc$ model	Splitting $P6_3mc$ model	$P31c$ model	$P6_3mc$ model	Splitting $P6_3mc$ model	$P31c$ model	$P6_3mc$ model	Splitting $P6_3mc$ model	$P31c$ model
R_{wp} (%)	4.79	4.48	4.53	5.25	4.72	4.79	6.60	5.83	5.70
R_p (%)	3.57	3.35	3.37	3.87	3.48	3.54	4.85	4.32	4.18
$\mathbf{U_{iso.}}$ of O3 (Å^2)	0.053(2)	0.012(2)	0.011(2)	0.064(2)	0.007(1)	0.007(2)	0.080(2)	0.009(2)	0.010(2)

Table S2. Atomic coordinates, occupancy factors, and isotropic thermal displacement parameters obtained from the Rietveld refinements.

$CaBaZn_2Al_2O_7$	x	y	z	Occ.	$\mathbf{U_{iso.}}$ (Å^2)
Ca	2/3	1/3	0.8715(2)	1	0.0112(6)
Ba	2/3	1/3	0.5	1	0.0206(3)
Al1/Zn1	0	0	0.4353(5）	0.942(4)/0.058(4)	0.014(1)
Al2/Zn2	0.17259(8)	0.82741(8)	0.6833(1）	0.353(4)/0.647(4)	0.0133(4)
O1	0.5032(6)	0.4968(6)	0.7332(5)	1	0.025(1)
O2	0	0	0.2614(9)	1	0.023(2)
O3	0.1510(3)	0.8490(3)	0.4940(6)	1	0.033(1)
$CaBa_{0.8}Sr_{0.2}Zn_2Al_2O_7$	x	y	z	Occ.	$\mathbf{U_{iso.}}$ (Å^2)
Ca	2/3	1/3	0.8711(2)	1	0.0056(6)
Ba/Sr	2/3	1/3	0.5	0.8/0.2	0.0197(4)
Al1/Zn1	0	0	0.4450(5)	0.926(4)/0.074(4)	0.0132(9)
Al2/Zn2	0.1692(7)	0.8261(8)	0.68950(9)	0.358(4)/0.642(4)	0.0132(4)
O1	0.503(3)	0.501(3)	0.7374(4)	1	0.027(2)
O2	0	0	0.2725(8)	1	0.016(2)
O3	0.2005(8)	0.8998(7)	0.5010(4)	1	0.011(2)

$CaBa_{0.6}Sr_{0.4}Zn_2Al_2O_7$	*x*	*y*	*z*	Occ.	$\mathbf{U_{iso.}}$ (Å)
Ca	2/3	1/3	0.8783(2)	1	0.0073(6)
Ba/Sr	2/3	1/3	0.5	0.6/0.4	0.0243(4)
Al1/Zn1	0	0	0.4394(5)	0.932(4)/0.068(4)	0.0187(9)
Al2/Zn2	0.1682(6)	0.8249(6)	0.6892(1)	0.356(4)/0.644(4)	0.0135(4)
O1	0.506(2)	0.500(2)	0.7364(4)	1	0.034(1)
O2	0	0	0.2754(8)	1	0.027(2)
O3	0.2088(7)	0.9078(7)	0.4974(4)	1	0.006(1)
$CaBa_{0.4}Sr_{0.6}Zn_2Al_2O_7$	*x*	*y*	*z*	Occ.	$\mathbf{U_{iso.}}$ (Å^2)
Ca	2/3	1/3	0.8709(2)	1	0.0056(7)
Ba/Sr	2/3	1/3	0.5	0.4/0.6	0.0214(5)
Al1/Zn1	0	0	0.4310(6)	0.940(4)/0.060(4)	0.0150(9)
Al2/Zn2	0.17246(9)	0.82754(9)	0.6835(2)	0.353(4)/0.647(4)	0.0118(4)
O1	0.508(2)	0.505(2)	0.7295(5)	1	0.041(2)
O2	0	0	0.2676(8)	1	0.028(3)
O3	0.2142(7)	0.9142(7)	0.4930(5)	1	0.010(2)
$CaBa_{0.2}Sr_{0.8}Zn_2Al_2O_7$	*x*	*y*	*z*	Occ.	$\mathbf{U_{iso.}}$ (Å^2)
Ca	0.6700(5)	1.0082(8)	0.8949(2)	1	0.0048(8)
Ba/Sr	0.6661(3)	0.0007(3)	0.52210(9)	0.8/0.2	0.0207(6)
Al11/Zn11	0.997(1)	0.003(1)	0.9617(7)	0.929(6)/0.071(6)	0.015(1)
Zn1/Al1	0.1740(3)	1.0045(6)	0.7067(6)	0.74(1)/0.26(1)	0.009(1)
Zn2/Al2	0.0906(3)	0.2549(6)	0.2128(4)	0.80(1)/0.20(1)	0.017(2)
Zn3/Al3	0.9207(3)	0.2598(8)	0.6962(5)	0.45(1)/0.55(1)	0.011(2)
O2	1.005(2)	0.988(3)	0.2815(7)	1	0.015(1)
O11	0.487(1)	0.012(3)	0.246(1)	1	0.015(1)
O12	0.245(1)	0.792(2)	0.810(1)	1	0.015(1)
O13	0.734(1)	0.738(2)	0.227(2)	1	0.015(1)

O31	0.145(1)	0.917(2)	0.511(2)	1	0.015(1)
O32	0.104(1)	0.202(2)	1.026(2)	1	0.015(1)
O33	0.9653(9)	0.260(2)	0.511(2)	1	0.015(1)
$CaSrZn_2Al_2O_7$	*x*	*y*	*z*	Occ.	$\mathbf{U_{iso.}}$ ($Å^2$)
Ca	0.6726(6)	1.000(1)	0.8713（3）	1	0.013(1)
Sr	0.6658(3)	0.0044(5)	0.5	1	0.018(1)
Al11/Zn11	0.004(1)	0.017(1)	0.935(2)	0.95(1)/0.05(1)	0.010(2)
Zn1/Al1	0.1758(3)	0.9925(7)	0.6840(7)	0.87(1)/0.13(1)	0.07(1)
Zn2/Al2	0.0893(3)	0.2541(8)	0.1903(7)	0.85(1)/0.15(1)	0.003(1)
Zn3/Al3	0.9238(5)	0.261(1)	0.6680(8)	0.22(1)/0.78(1)	0.009(3)
O2	1.003(2)	0.969(3)	0.257(1)	1	0.008(2)
O11	0.487(1)	0.006(4)	0.231(2)	1	0.008(2)
O12	0.258(2)	0.784(3)	0.792(1)	1	0.008(2)
O13	0.741(1)	0.721(2)	0.178(2)	1	0.008(2)
O31	0.146(1)	0.902(2)	0.482(2)	1	0.008(2)
O32	0.116(1)	0.185(2)	1.00(2)	1	0.008(2)
O33	0.972(1)	0.249(2)	0.509(2)	1	0.008(2)

Table S3 Selected bond lengths obtained from Rietveld refinements for $CaBa_{1-x}Sr_xZn_2Al_2O_7$ (x = 0, 0.2, 0.4, 0.6).

$CaBaZn_2Al_2O_7$					
Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
Ca-O1 × 3	2.259(6)	Ba-O1 × 3	3.254(6)	T2-O1 × 2	1.872(4)
Ca-O3 × 3	2.337(4)	<Ba-O>	3.125(3)	T2-O2	2.038(4)
<Ca-O>	2.298(6)	T1-O2	1.75(1)	T2-O3	1.913(6)
Ba-O1 × 3	2.941(6)	T1-O3 × 3	1.748(4)	<T2-O>	1.924(4)

Ba-O3 × 6	3.15322(9)	<T1-O>	1.749(6)		

$CaBa_{0.8}Sr_{0.2}Zn_2Al_2O_7$

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
Ca-O1 × 3	2.283(6)	Ba-O3 × 3	3.461(3)	T2-O1	1.842(15)
Ca-O3 × 3	2.361(6)	<Ba-O>	3.120(2)	T2-O1	1.876(15)
<Ca-O>	2.322(6)	T1-O2	1.698(9)	T2-O2	2.043(6)
Ba-O3 × 3	2.836(3)	T1-O3 × 3	1.770(6)	T2-O3	1.929(6)
Ba-O1 × 3	2.931(1)	<T1-O>	1.752(7)	<T2-O>	1.923(11)
Ba-O1 × 3	3.253(1)				

$CaBa_{0.6}Sr_{0.4}Zn_2Al_2O_7$

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
Ca-O1 × 3	2.27(1)	Ba-O3 × 3	3.507(3)	T2-O1	1.861(14)
Ca-O3 × 3	2.343(5)	<Ba-O>	3.112(7)	T2-O1	1.837(13)
<Ca-O>	2.307(8)	T1-O2	1.635(9)	T2-O2	2.054(5)
Ba-O3 × 3	2.780(3)	T1-O3 × 3	1.772(5)	T2-O3	1.965 (6)
Ba-O1 × 3	2.89(1)	<T1-O>	1.738(6)	<T2-O>	1.929(9)
Ba-O1 × 3	3.27(1)				

$CaBa_{0.4}Sr_{0.6}Zn_2Al_2O_7$

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
Ca-O1 × 3	2.28(1)	Ba-O3 × 3	3.539(3)	T2-O1	1.844(12)
Ca-O3 × 3	2.364(5)	<Ba-O>	3.107(7)	T2-O1	1.853(13)
<Ca-O>	2.307(8)	T1-O2	1.624(9)	T2-O2	2.044(5)
Ba-O3 × 3	2.737(3)	T1-O3 × 3	1.785(6)	T2-O3	1.947(5)
Ba-O1 × 3	2.90(1)	<T1-O>	1.745(7)	<T2-O>	1.922(9)
Ba-O1 × 3	3.25(1)				

$CaBa_{0.2}Sr_{0.8}Zn_2Al_2O_7$

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)

Ca-O11	2.257(13)	Sr-O33	2.644(11)	Zn1-O31	2.039(18)
Ca-O12	2.208(13)	Sr-O32	3.473(13)	Zn1-O2	2.076(18)
Ca-O13	2.429(14)	Sr-O13	3.436(14)	<Zn1-O>	1.965(16)
Ca-O31	2.357(15)	Sr-O12	3.482(11)	Zn2-O11	1.867(17)
Ca-O32	2.339(14)	< Sr-O>	3.00(1)	Zn2-O32	1.884(16)
Ca-O33	2.424(14)	Al11-O31	1.696(17)	Zn2-O2	2.027(19)
< Ca-O>	2.34(4)	Al11-O33	1.758(15)	Zn2-O12	2.038(15)
Sr-O11	2.768(13)	Al11-O2	1.785(10)	<Zn2-O>	1.954(17)
Sr-O12	2.656(11)	Al11-O32	1.812(17)	Zn3-O13	1.713(15)
Sr-O11	3.357(13)	<Al11-O>	1.763(14)	Zn3-O33	1.895(18)
Sr-O4	2.729(14)	Zn1-O12	1.844(13)	Zn3-O11	1.926(18)
Sr-O31	2.620(10)	Zn1-O13	1.900(13)	Zn3-O2	1.937(19)
Sr-O32	2.796(13)			<Zn3-O>	1.868(18)

$CaSrZn_2Al_2O_7$

Bond	Length (Å)	Bond	Length (Å)	Bond	Length (Å)
Ca-O11	2.217(17)	Sr-O11	2.814(16)	<Zn1-O>	2.022(18)
Ca-O12	2.144(18)	Sr-O11	3.291(16)	Zn2-O32	1.94(2)
Ca-O13	2.528(19)	<Sr-O>	2.702(15)	Zn2-O2	2.109(18)
Ca-O31	2.331(16)	Al11-O31	1.77(2)	Zn2-O12	1.943(19)
Ca-O32	2.421(17)	Al11-O33	1.827(18)	Zn2-O11	1.90(2)
Ca-O33	2.602(16)	Al11-O2	1.763(19)	<Zn2-O>	1.973(19)
<Ca-O>	2.374(17)	Al11-O32	1.73(2)	Zn3-O13	1.811(16)
Sr-O13	2.432(19)	<Al11-O>	1.773	Zn3-O33	1.65(2)
Sr-O31	2.548(12)	Zn1-O12	1.902(17)	Zn3-O11	1.91(2)
Sr-O12	2.578(15)	Zn1-O13	2.002(15)	Zn3-O2	1.854(19)
Sr-O33	2.603(13)	Zn1-O31	2.095(19)	<Zn3-O>	1.806(19)
Sr-O32	2.650(15)	Zn1-O2	2.09(2)		

Table S4. The lattice parameters for $CaBa_{1-x}Sr_xZn_2Al_2O_7$ (x = 0, 0.2, 0.4, 0.6, 0.8, and 1.0) obtained from Rietveld refinements and $Ca_{0.98}Eu_{0.02}Ba_{1-y}Sr_yZn_{2.02}Al_{1.98}O_7$ (y = 0, 0.4, 0.8) obtained from Le-bail fitting.

	a (Å)	b (Å)	c (Å)	V (Å^3)
x = 0	6.29603(2)	6.29603(2)	10.02682(5)	344.213(3)
x = 0.2	6.28600(6)	6.28600(6)	10.0036(1)	342.324(8)
x = 0.4	6.27357(5)	6.27357(5)	9.9710(1)	339.791(6)
x = 0.6	6.26264(4)	6.26264(4)	9.93947(7)	337.605(5)
x = 0.8	10.8487(1)	6.23985(7)	9.8979(1)	670.03(1)
x = 1.0	10.84872(9)	6.22609(5)	9.86609 (9)	666.40(1)
y = 0	6.2960	6.2960	10.0268	344.2130
y = 0.4	6.2767	6.2767	9.9815	340.5532
y = 0.8	10.8421	6.2388	9.8984	669.5441

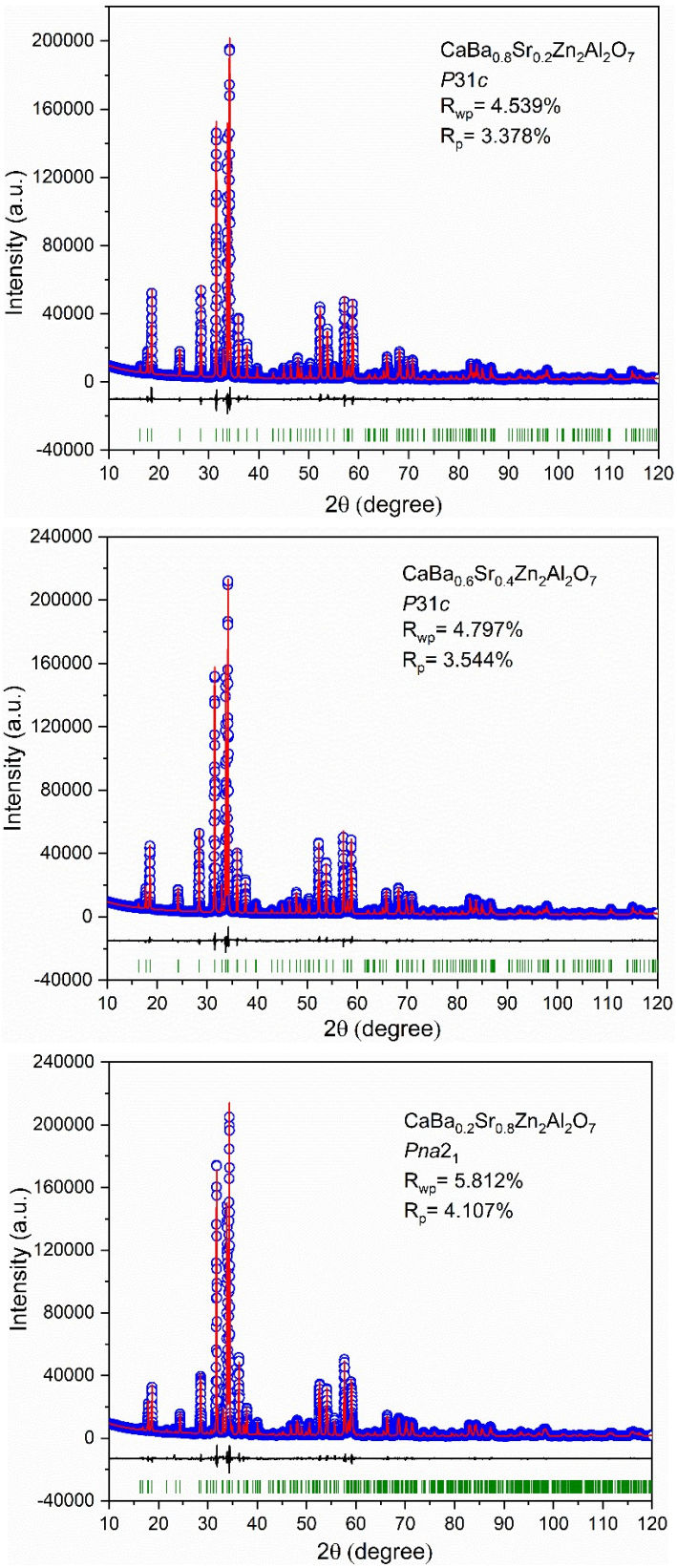


Fig. S1 Rietveld refinement patterns for $CaBa_{1-x}Sr_xZn_2Al_2O_7$ (x = 0.2, 0.4 and 0.8).

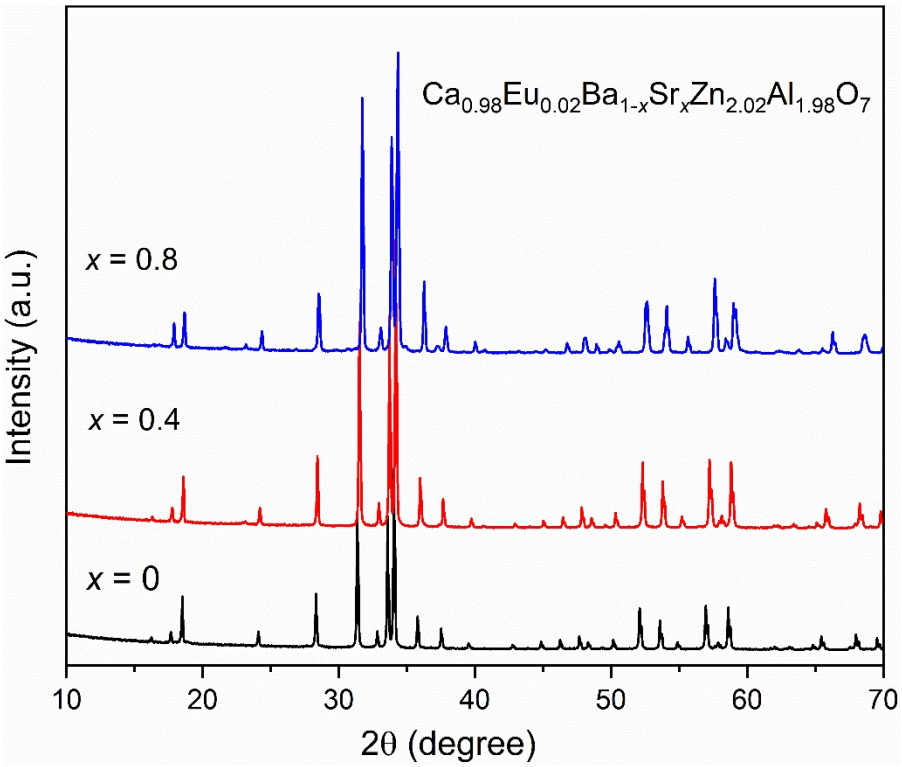
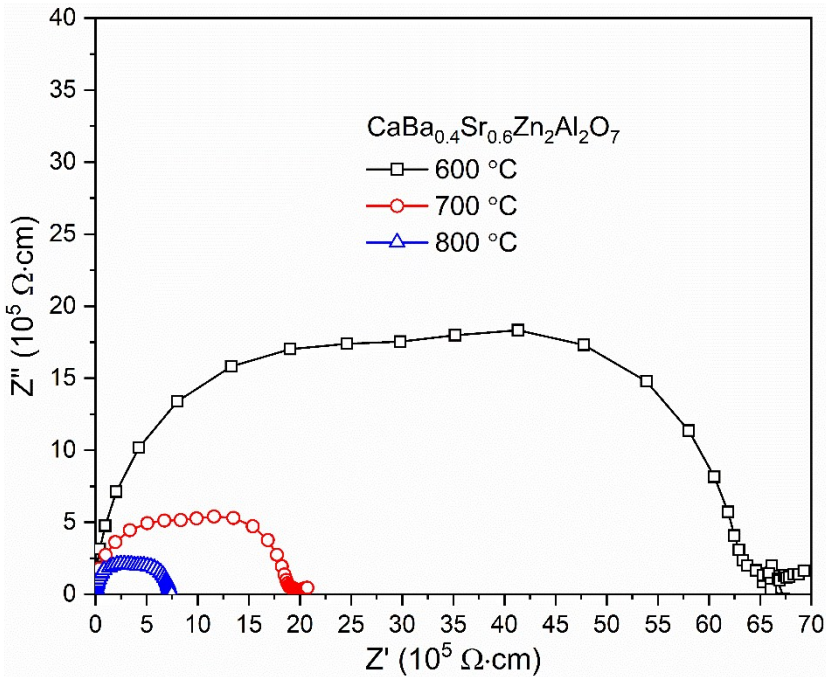


Fig. S2 XRD patterns for $Ca_{0.98}Eu_{0.02}Ba_{1-x}Sr_xZn_{2.02}Al_{1.98}O_7$ (x = 0, 0.4, 0.8).



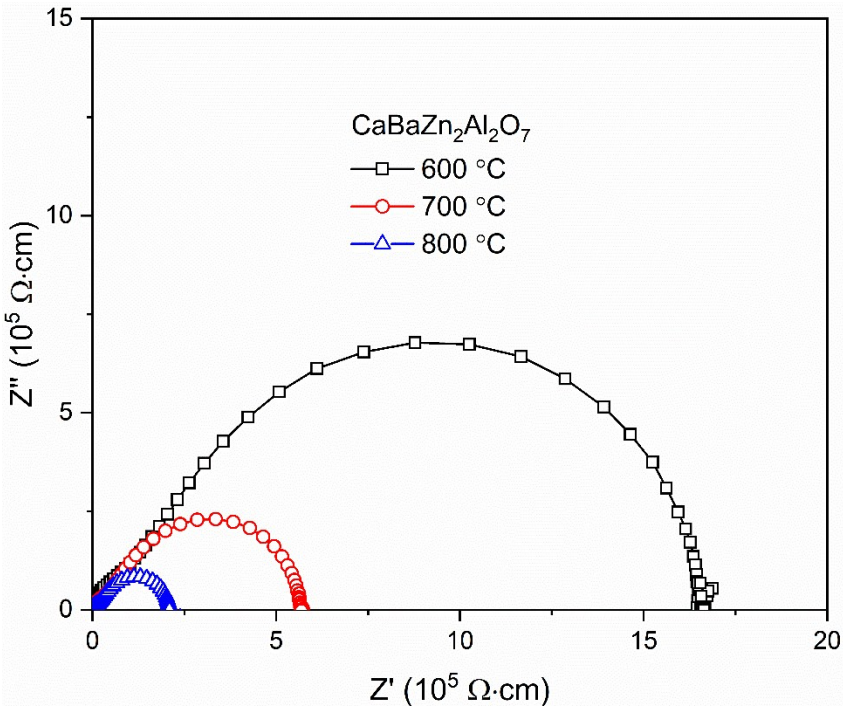


Fig. S3 Complex *ac* impedance spectra for $CaBa_{0.4}Sr_{0.6}ZnAlO_7$ and $CaBaZnAlO_7$ measured in air at 600, 700, and 800 ºC.