Supporting Information

Enhanced crystallinity and thermal stability of Ba^{2+} and Al^{3+}-O^{2-} co-substituted $Sr_2Si_5N_8:Eu^{2+}$

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Figure S1 The SEM micrographs of x=0,0.3,0.6 in three series: Ba-AlO-258, Ca-AlO-258 and AlO-258

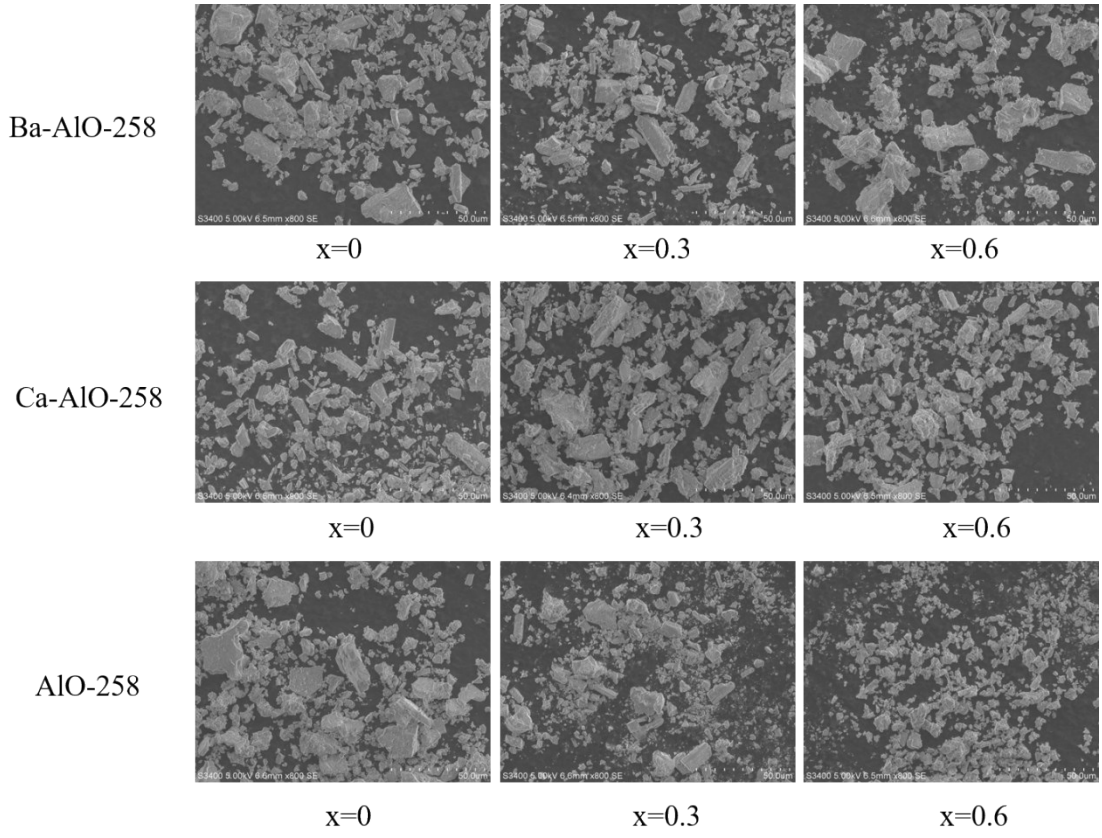


Figure S2 Rietveld refinement of Ba-AlO-258: ($Sr_{1.7}Ba_{0.3}Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9, Black crosses: measured diffractogram; bottom blue line: difference profile; red line: calculated diffractogram; olive green, orange and violet bars: positions of the diffraction

peak.

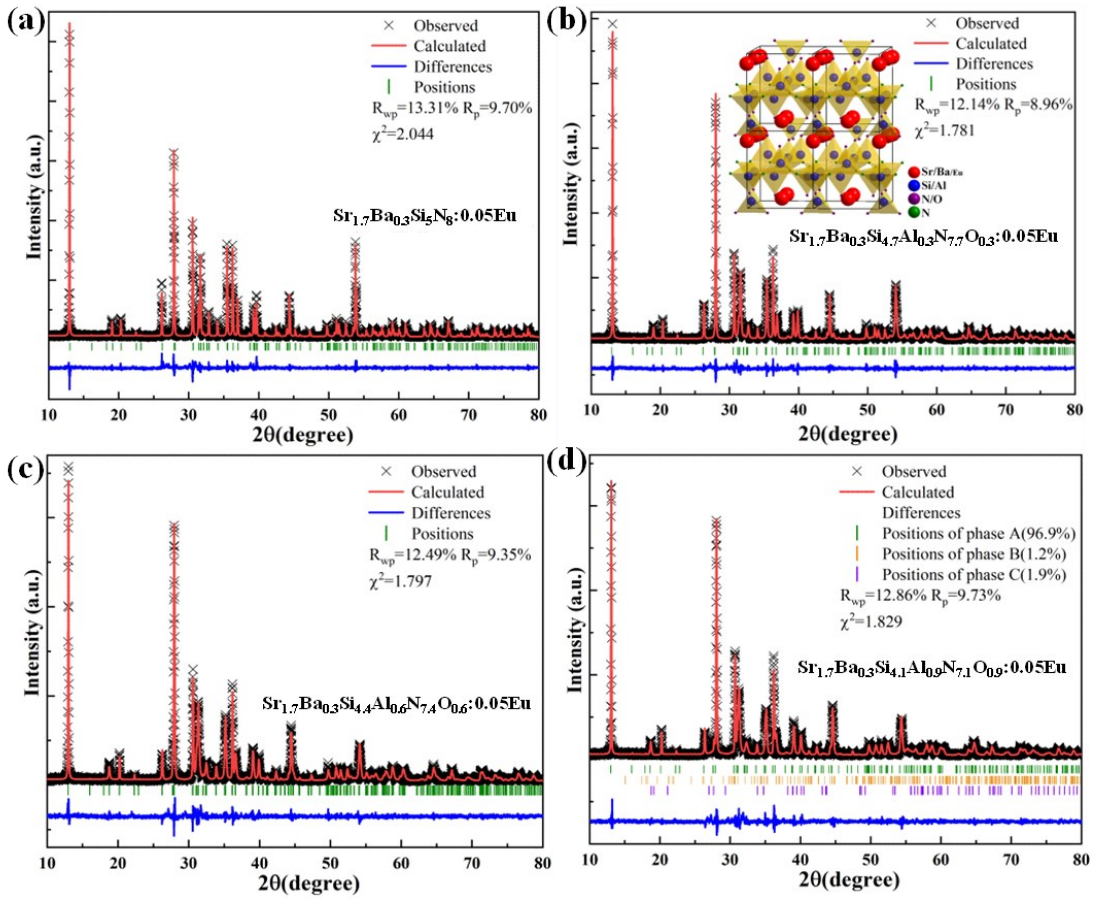


Figure S3 Rietveld refinement of Ca-AlO-258: ($Sr_{1.7}Ca_{0.3}Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9, Black crosses: measured diffractogram; bottom blue line: difference profile; red line: calculated diffractogram; olive green, orange and violet bars: positions of the diffraction peak.

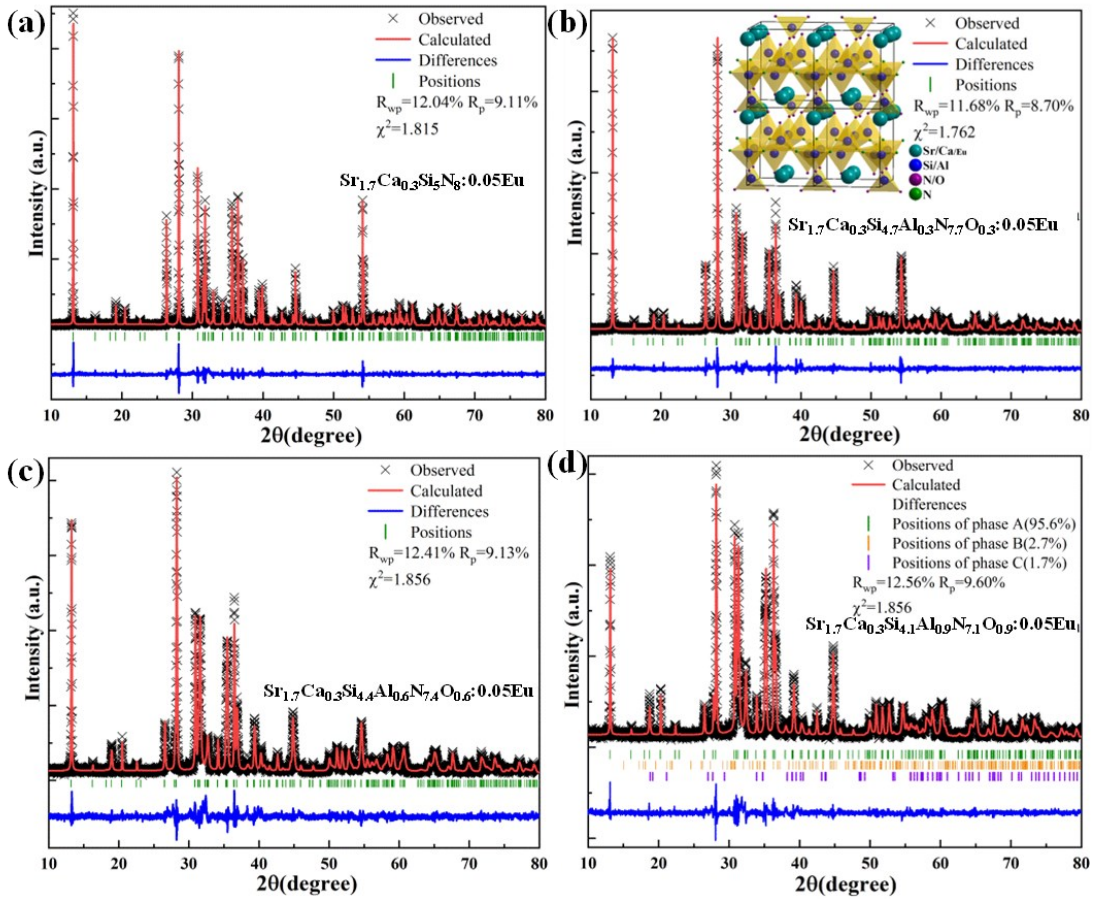


Figure S4 Rietveld refinement of AlO-258 ($Sr_2Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9, Black crosses: measured diffractogram; bottom blue line: difference profile; red line:

calculated diffractogram; olive green, orange and violet bars: positions of the diffraction peak.

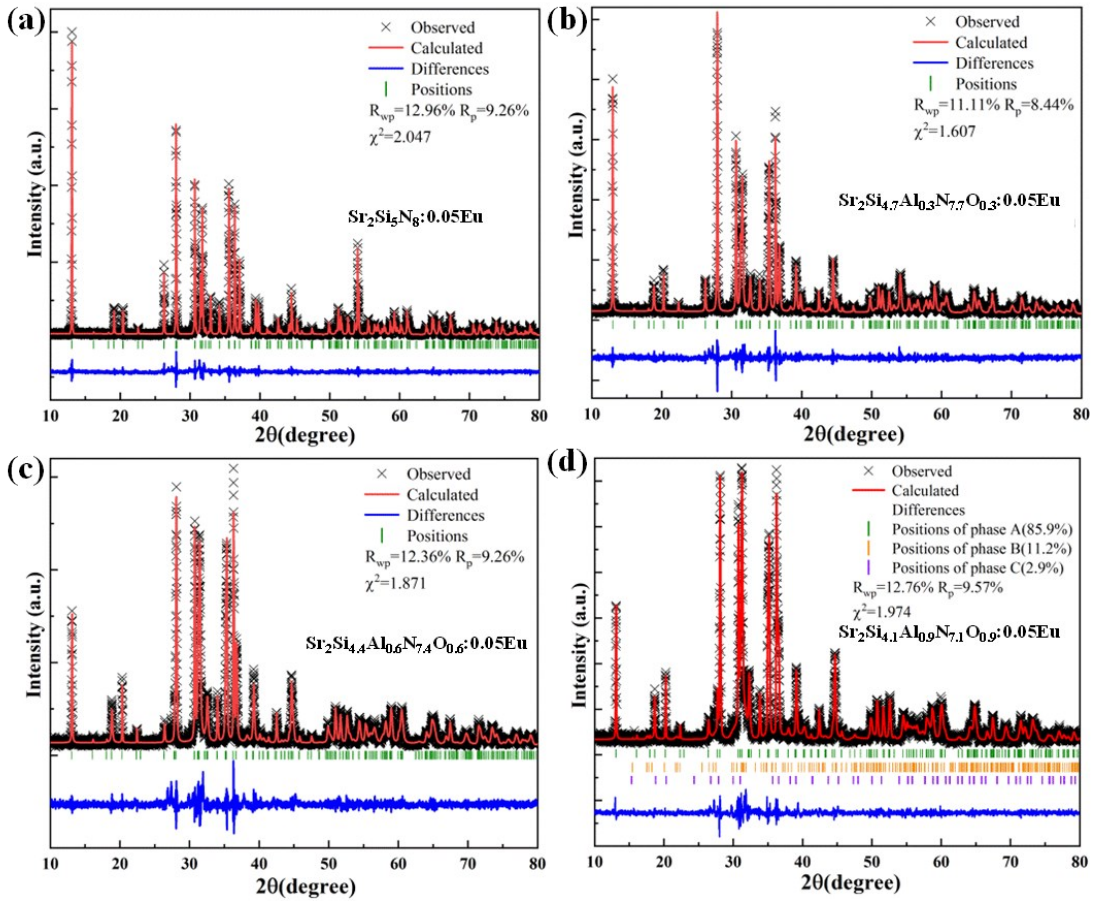


Table S1 The detailed crystal datum of all samples of the three series in the paper from the Rietveld refinement.

Formula sum			$Sr_{1.7}Ba_{0.3}Si_5N_8$:0.05Eu			
Formula weight			445.48 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.6974(1) Å b=6.8127(0) Å c=9.3089(1) Å			
Cell ratio			a/b=0.8363 b/c=0.7319 c/a=1.6339			
Cell volume			361.32(1) Å^3			
Z			2			
Atom	**x/a**	**y/b**	**z/c**	**U [Å^2]**	**Wyck.**	**Frac.**
Sr1	0	0.87032	0.99865	0.0088	2a	0.84981
Sr2	0	0.88247	0.36711	0.0324	2a	0.80769
Si1	0.24826	0.33231	0.1829	0.0198	4b	1
Si2	0	0.05489	0.67753	0.0214	2a	1
Si3	0	0.40179	0.90189	0.0021	2a	1
Si4	0	0.41979	0.46112	0.0032	2a	1
N1	0.24781	0.91352	0.67179	0.0039	4b	1
N2	0.24941	0.44384	0.01098	0.0022	4b	1
N3	0	0.17423	0.83842	0.0023	2a	1
N4	0	0.19029	0.52193	0.0032	2a	1
N5	0	0.42502	0.2718	0.0072	2a	1

N6	0	0.58632	0.77327	0.0049	2a	1
Eu1	0	0.87032	0.99865	0.0088	2a	0.025
Eu2	0	0.88247	0.36711	0.0324	2a	0.025
Ba1	0	0.87032	0.99865	0.0088	2a	0.12519
Ba2	0	0.88247	0.36711	0.0324	2a	0.16731

Formula sum			$Sr_{1.7}Ba_{0.3}Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$:0.05Eu			
Formula weight			445.74 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7178(2) Å b=6.7894(1) Å c=9.3654(2) Å			
Cell ratio			a/b=0.8422 b/c=0.7249 c/a=1.6379			
Cell volume			363.56(1) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.8687	1.00074	0.0069	2a	0.83126
Sr2	0	0.88298	0.36779	0.0231	2a	0.82624
Ba1	0	0.8687	1.00074	0.0069	2a	0.14374
Ba2	0	0.88298	0.36779	0.0231	2a	0.14876
Si1	0.24636	0.33652	0.19182	0.0141	4b	0.94
Si2	0	0.0591	0.68762	0.0291	2a	0.94
Si3	0	0.40463	0.90282	0.0032	2a	0.94
Si4	0	0.42151	0.46116	0.0015	2a	0.94
N1	0.24194	0.9094	0.67734	0.0066	4b	0.925
N2	0.25017	0.44872	0.0086	0.0049	4b	1
N3	0	0.17965	0.83145	0.0005	2a	0.925
N4	0	0.19363	0.52104	0.0013	2a	0.925
N5	0	0.4247	0.27274	0.0018	2a	1
N6	0	0.58227	0.75835	0.0012	2a	1
Eu1	0	0.8687	1.00074	0.0069	2a	0.025
Eu2	0	0.88298	0.36779	0.0231	2a	0.025
Al1	0.24636	0.33652	0.19182	0.0141	4b	0.06
Al2	0	0.0591	0.68762	0.0291	2a	0.06
Al3	0	0.40463	0.90282	0.0032	2a	0.06
Al4	0	0.42151	0.46116	0.0015	2a	0.06
O1	0.24194	0.9094	0.67734	0.0066	4b	0.075
O3	0	0.17965	0.83145	0.0005	2a	0.075
O4	0	0.19363	0.52104	0.0013	2a	0.075

Formula sum	$Sr_{1.7}Ba_{0.3}Si_{4.4}Al_{0.6}N_{7.4}O_{0.6}$:0.05Eu
Formula weight	446.01 g/mol
Crystal system	orthorhombic
Space-group	P m n 21 (31)
Cell parameters	a=5.7391(2) Å b=6.7731(1) Å c=9.4188(3) Å
Cell ratio	a/b=0.8473 b/c=0.7191 c/a=1.6412
Cell volume	366.12(2) $Å^3$
Z	8

Atom	x/a	y/b	z/c	U [Å²]	Wyck.	Frac.
Sr1	0	0.87041	1.00096	0.0153	2a	0.82875
Sr2	0	0.88296	0.36763	0.0159	2a	0.82875
Si1	0.24781	0.33136	0.18401	0.0173	4b	0.88
Si2	0	0.05555	0.68001	0.0077	2a	0.88
Si3	0	0.40376	0.90187	0.0335	2a	0.88
Si4	0	0.42133	0.4613	0.0073	2a	0.88
N1	0.2242(25)	0.9128(8)	0.674(4)	0.0149	4b	0.85
N2	0.245(4)	0.4541(11)	-0.0017(20)	0.0024	4b	1
N3	0	0.1896(15)	0.8157(21)	0.0019	2a	0.85
N4	0	0.1950(15)	0.5160(26)	0.0043	2a	0.85
N5	0	0.42612	0.27092	0.0105	2a	1
N6	0	0.58727	0.77039	0.0393	2a	1
Eu1	0	0.87041	1.00096	0.0153	2a	0.025
Eu2	0	0.88296	0.36763	0.1059	2a	0.025
Al1	0.24781	0.33136	0.18401	0.0173	4b	0.12
Al2	0	0.05555	0.68001	0.0077	2a	0.12
Al3	0	0.40376	0.90187	0.0335	2a	0.12
Al4	0	0.42133	0.4613	0.0073	2a	0.12
O1	0.2242(25)	0.9128(8)	0.674(4)	0.0149	4b	0.15
O3	0	0.1896(15)	0.8157(21)	0.0019	2a	0.15
O4	0	0.1950(15)	0.5160(26)	0.0043	2a	0.15
Ba1	0	0.87041	1.00096	0.0153	2a	0.14625
Ba2	0	0.88296	0.36763	0.0159	2a	0.14625

Formula sum	$Sr_{1.7}Ba_{0.3}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu
Formula weight	446.28/mol
Crystal system	orthorhombic
Space-group	P m n 21 (31)
Cell parameters	a=5.7658(2) Å b=6.7608(1) Å c=9.4855(3) Å
Cell ratio	a/b=0.8528 b/c=0.7128 c/a=1.6451
Cell volume	369.76(2) $Å^3$
Z	8

Atom	x/a	y/b	z/c	U [Å²]	Wyck.	Frac.
Sr1	0	0.87234	0.99998	0.0078	2a	0.82875
Sr2	0	0.88348	0.36812	0.0109	2a	0.82875
Si1	0.24835	0.33205	0.18505	0.0135	4b	0.82
Si2	0	0.05632	0.67705	0.0231	2a	0.82
Si3	0	0.40261	0.90255	0.0112	2a	0.82
Si4	0	0.42051	0.46163	0.0061	2a	0.82
N1	0.23955	0.9039	0.66117	0.0159	4b	0.775
N2	0.24903	0.4479	0.01276	0.0072	4b	1
N3	0	0.18	0.84074	0.0043	2a	0.775
N4	0	0.18721	0.51989	0.0003	2a	0.775
N5	0	0.42742	0.26607	0.004	2a	1
N6	0	0.58152	0.7719	0.0278	2a	1
Eu1	0	0.87234	0.99998	0.0078	2a	0.025

Eu2	0	0.88348	0.36812	0.0109	2a	0.025
Al1	0.24835	0.33205	0.18505	0.0135	4b	0.18
Al2	0	0.05632	0.67705	0.0231	2a	0.18
Al3	0	0.40261	0.90255	0.0112	2a	0.18
Al4	0	0.42051	0.46163	0.0061	2a	0.18
O1	0.23955	0.9039	0.66117	0.0159	4b	0.225
O3	0	0.18	0.84074	0.0043	2a	0.225
O4	0	0.18721	0.51989	0.0003	2a	0.225
Ba1	0	0.87234	0.99998	0.0078	2a	0.14625
Ba2	0	0.88348	0.36812	0.0109	2a	0.14625

Formula sum			$Sr_{1.7}Ca_{0.3}Si_5N_8$:0.05Eu			
Formula weight			417.03g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.6838(1) Å b=6.785() Å c=9.2971(1) Å			
Cell ratio			a/b=0.8377 b/c=0.7298 c/a=1.6357			
Cell volume			358.54(0) $Å^3$			
Z			2			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87344	0.99973	0.0152	2a	0.84419
Sr2	0	0.8849	0.3683	0.0127	2a	0.81331
Si1	0.24832	0.33299	0.18382	0.0173	4b	1
Si2	0	0.05339	0.67507	0.0079	2a	1
Si3	0	0.40642	0.90267	0.0144	2a	1
Si4	0	0.42288	0.46074	0.0045	2a	1
N1	0.24632	0.91324	0.67487	0.0119	4b	1
N2	0.25561	0.44555	0.01239	0.0009	4b	1
N3	0	0.17436	0.83978	0.0068	2a	1
N4	0	0.1885	0.52127	0.0091	2a	1
N5	0	0.42549	0.27339	0.001	2a	1
N6	0	0.58482	0.77147	0.01	2a	1
Eu1	0	0.87344	0.99973	0.0152	2a	0.025
Eu2	0	0.8849	0.3683	0.0127	2a	0.025
Ca1	0	0.87344	0.99973	0.0152	2a	0.13081
Ca2	0	0.8849	0.3683	0.0127	2a	0.16169

Formula sum			$Sr_{1.7}Ca_{0.3}Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$:0.05Eu			
Formula weight			417.30 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7014(1) Å b=6.7575(1) Å c=9.3525(2) Å			
Cell ratio			a/b=0.8437 b/c=0.7225 c/a=1.6404			
Cell volume			360.33(1) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87199	1.00011	0.01	2a	0.82875

Sr2	0	0.88287	0.36803	0.0137	2a	0.82875
Ca1	0	0.87199	1.00011	0.01	2a	0.14625
Ca2	0	0.88287	0.36803	0.0137	2a	0.14625
Si1	0.24719	0.33327	0.18394	0.0144	4b	0.94
Si2	0	0.05561	0.67689	0.0197	2a	0.94
Si3	0	0.40452	0.90225	0.0092	2a	0.94
Si4	0	0.42207	0.4611	0.0009	2a	0.94
N1	0.24682	0.91221	0.67246	0.009	4b	0.925
N2	0.2535	0.44527	0.01091	0.0046	4b	1
N3	0	0.17524	0.83939	0.0037	2a	0.925
N4	0	0.18999	0.52081	0.0001	2a	0.925
N5	0	0.42615	0.27166	0.0021	2a	1
N6	0	0.58518	0.77176	0.0013	2a	1
Eu1	0	0.87199	1.00011	0.01	2a	0.025
Eu2	0	0.88287	0.36803	0.0137	2a	0.025
Al1	0.24719	0.33327	0.18394	0.0144	4b	0.06
Al2	0	0.05561	0.67689	0.0197	2a	0.06
Al3	0	0.40452	0.90225	0.0092	2a	0.06
Al4	0	0.42207	0.4611	0.0009	2a	0.06
O1	0.24682	0.91221	0.67246	0.009	4b	0.075
O3	0	0.17524	0.83939	0.0037	2a	0.075
O4	0	0.18999	0.52081	0.0001	2a	0.075

Formula sum			$Sr_{1.7}Ca_{0.3}Si_{4.4}Al_{0.6}N_{7.4}O_{0.6}$:0.05Eu			
Formula weight			417.57 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7235(2) Å b=6.7362(1) Å c=9.4049(3) Å			
Cell ratio			a/b=0.8497 b/c=0.7162 c/a=1.6432			
Cell volume			362.60(2) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87282	0.99964	0.0111	2a	0.82875
Sr2	0	0.88422	0.36835	0.0113	2a	0.82875
Si1	0.24772	0.33223	0.18448	0.0114	4b	0.88
Si2	0	0.05439	0.67854	0.0109	2a	0.88
Si3	0	0.40367	0.90206	0.0122	2a	0.88
Si4	0	0.42219	0.46096	0.0035	2a	0.88
N1	0.24573	0.91293	0.6744	0.0083	4b	0.85
N2	0.25182	0.44536	0.01176	0.002	4b	1
N3	0	0.17562	0.84001	0.0039	2a	0.85
N4	0	0.18993	0.52003	0.0025	2a	0.85
N5	0	0.42676	0.27174	0.0081	2a	1
N6	0	0.58463	0.77093	0.0169	2a	1
Eu1	0	0.87282	0.99964	0.0111	2a	0.025
Eu2	0	0.88422	0.36835	0.0113	2a	0.025
Al1	0.24772	0.33223	0.18448	0.0114	4b	0.12

Al2	0	0.05439	0.67854	0.0109	2a	0.12
Al3	0	0.40367	0.90206	0.0122	2a	0.12
Al4	0	0.42219	0.46096	0.0035	2a	0.12
O1	0.24573	0.91293	0.6744	0.0083	4b	0.15
O3	0	0.17562	0.84001	0.0039	2a	0.15
O4	0	0.18993	0.52003	0.0025	2a	0.15
Ca1	0	0.87282	0.99964	0.0111	2a	0.14625
Ca2	0	0.88422	0.36835	0.0113	2a	0.14625

Formula sum			$Sr_{1.7}Ca_{0.3}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu			
Formula weight			417.83 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7452(2) Å b=6.7180(1) Å c=9.4542(3) Å			
Cell ratio			a/b=0.8552 b/c=0.7106 c/a=1.6456			
Cell volume			364.90(2) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87344	0.99959	0.011	2a	0.82875
Sr2	0	0.88448	0.36838	0.0108	2a	0.82875
Si1	0.2475	0.33118	0.18524	0.0106	4b	0.82
Si2	0	0.05404	0.67892	0.01	2a	0.82
Si3	0	0.40191	0.90242	0.0149	2a	0.82
Si4	0	0.42058	0.461	0.0117	2a	0.82
N1	0.242(2)	0.9153(11)	0.6732(31)	0.0078	4b	0.775
N2	0.2478(30)	0.4449(15)	0.0101(19)	0.0109	4b	1
N3	0	0.1753(22)	0.8372(21)	0.0109	2a	0.775
N4	0	0.1913(21)	0.5200(23)	0.0039	2a	0.775
N5	0	0.4237(24)	0.2673(19)	0.013	2a	1
N6	0	0.5884(25)	0.7725(19)	0.0252	2a	1
Eu1	0	0.87344	0.99959	0.011	2a	0.025
Eu2	0	0.88448	0.36838	0.0108	2a	0.025
Al1	0.2475	0.33118	0.18524	0.0106	4b	0.18
Al2	0	0.05404	0.67892	0.01	2a	0.18
Al3	0	0.40191	0.90242	0.0149	2a	0.18
Al4	0	0.42058	0.461	0.0117	2a	0.18
O1	0.242(2)	0.9153(11)	0.6732(31)	0.0078	4b	0.225
O3	0	0.1753(22)	0.8372(21)	0.0109	2a	0.225
O4	0	0.1913(21)	0.5200(23)	0.0039	2a	0.225
Ca1	0	0.87344	0.99959	0.011	2a	0.14625
Ca2	0	0.88448	0.36838	0.0108	2a	0.14625

Formula sum	$Sr_2Si_5N_8$:0.05Eu
Formula weight	430.94 g/mol
Crystal system	orthorhombic
Space-group	P m n 21 (31)

Cell parameters			a=5.6900(1) Å b=6.7949(0) Å c=9.3030(1) Å			
Cell ratio			a/b=0.8374 b/c=0.7304 c/a=1.6350			
Cell volume			359.68(1) $Å^3$			
Z			2			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87073	0.99912	0.0109	2a	0.975
Sr2	0	0.88539	0.36848	0.0126	2a	0.975
Si1	0.24783	0.334	0.1879	0.0141	4b	1
Si2	0	0.05489	0.67759	0.0101	2a	1
Si3	0	0.40416	0.90193	0.0102	2a	1
Si4	0	0.42399	0.46116	0.0055	2a	1
N1	0.2433	0.91192	0.67754	0.0133	4b	1
N2	0.25801	0.44385	0.00947	0.0046	4b	1
N3	0	0.17447	0.83812	0.0097	2a	1
N4	0	0.18772	0.5224	0.0103	2a	1
N5	0	0.42342	0.27202	0.0007	2a	1
N6	0	0.58575	0.76965	0.0096	2a	1
Eu1	0	0.87073	0.99912	0.0109	2a	0.025
Eu2	0	0.88539	0.36848	0.0126	2a	0.025

Formula sum			$Sr_2Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$:0.05Eu			
Formula weight			431.21 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7078(1) Å b=6.7729(1) Å c=9.3568(2) Å			
Cell ratio			a/b=0.8427 b/c=0.7239 c/a=1.6393			
Cell volume			361.72(1) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87252	0.99799	0.0148	2a	0.975
Sr2	0	0.87937	0.36512	0.0244	2a	0.975
Si1	0.24659	0.33182	0.18043	0.024	4b	0.9349
Si2	0	0.05407	0.67717	0.0131	2a	0.9451
Si3	0	0.40376	0.90397	0.0119	2a	0.97299
Si4	0	0.42109	0.46022	0.0034	2a	0.90701
N1	0.24705	0.91247	0.67021	0.0016	4b	0.925
N2	0.24762	0.44403	0.00972	0.0011	4b	1
N3	0	0.17072	0.83607	0.0169	2a	0.93559
N4	0	0.18897	0.52118	0.0081	2a	0.91442
N5	0	0.42853	0.2692	0.0112	2a	1
N6	0	0.58551	0.77436	0.0155	2a	1
Eu1	0	0.87252	0.99799	0.0148	2a	0.025
Eu2	0	0.87937	0.36513	0.0244	2a	0.025
Al1	0.24659	0.33182	0.18043	0.024	4b	0.05491
Al2	0	0.05407	0.67716	0.0131	2a	0.06509
Al3	0	0.40375	0.90397	0.0119	2a	0.02701
Al4	0	0.42109	0.46022	0.0034	2a	0.09299

O1	0.24705	0.91247	0.67021	0.0016	4b	0.075
O3	0	0.17072	0.83607	0.0169	2a	0.06442
O4	0	0.18897	0.52118	0.0081	2a	0.08558

Formula sum			$Sr_2Si_{4.4}Al_{0.6}N_{7.4}O_{0.6}$:0.05Eu			
Formula weight			431.47 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7268(1) Å b=6.7473(1) Å c=9.4114(3) Å			
Cell ratio			a/b=0.8488 b/c=0.7169 c/a=1.6434			
Cell volume			363.66(1) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87305	0.99966	0.0102	2a	0.975
Sr2	0	0.88386	0.36834	0.01	2a	0.975
Si1	0.24772	0.33309	0.18604	0.01	4b	0.88016
Si2	0	0.05469	0.68025	0.0103	2a	0.87984
Si3	0	0.40344	0.9023	0.0101	2a	0.77866
Si4	0	0.41979	0.46112	0.0096	2a	0.98134
N1	0.24681	0.91239	0.67155	0.0097	4b	0.85
N2	0.24836	0.44382	0.01067	0.0077	4b	1
N3	0	0.17487	0.83803	0.0101	2a	0.90236
N4	0	0.19164	0.52217	0.0095	2a	0.79764
N5	0	0.42646	0.27071	0.0095	2a	1
N6	0	0.58546	0.77325	0.0147	2a	1
Eu1	0	0.87305	0.99966	0.0102	2a	0.025
Eu2	0	0.88386	0.36834	0.01	2a	0.025
Al1	0.24772	0.33309	0.18604	0.01	4b	0.12016
Al2	0	0.05469	0.68025	0.0103	2a	0.11983
Al3	0	0.40344	0.9023	0.0101	2a	0.22134
Al4	0	0.41979	0.46112	0.0096	2a	0.01866
O1	0.24681	0.91239	0.67155	0.0097	4b	0.15
O3	0	0.17487	0.83803	0.0101	2a	0.09764
O4	0	0.19164	0.52217	0.0095	2a	0.20235

Formula sum			$Sr_2Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu			
Formula weight			431.74 g/mol			
Crystal system			orthorhombic			
Space-group			P m n 21 (31)			
Cell parameters			a=5.7488(2) Å b=6.7227(1) Å c=9.4603(3) Å			
Cell ratio			a/b=0.8551 b/c=0.7106 c/a=1.6456			
Cell volume			365.61(2) $Å^3$			
Z			8			
Atom	**x/a**	**y/b**	**z/c**	**U [$Å^2$]**	**Wyck.**	**Frac.**
Sr1	0	0.87311	0.99978	0.0102	2a	0.975
Sr2	0	0.88377	0.36821	0.0103	2a	0.975

Si1	0.24826	0.33249	0.18322	0.0099	4b	0.82
Si2	0	0.05475	0.67952	0.0106	2a	0.82
Si3	0	0.40207	0.90205	0.009	2a	0.82
Si4	0	0.4198	0.46093	0.0089	2a	0.82
N1	0.23952	0.91206	0.67534	0.0077	4b	0.775
N2	0.24037	0.44148	0.00754	0.0086	4b	1
N3	0	0.17356	0.83943	0.0095	2a	0.775
N4	0	0.19424	0.51918	0.0063	2a	0.775
N5	0	0.42444	0.26859	0.0072	2a	1
N6	0	0.59296	0.77833	0.0142	2a	1
Eu1	0	0.87311	0.99978	0.0102	2a	0.025
Eu2	0	0.88377	0.36821	0.0103	2a	0.025
Al1	0.24826	0.33249	0.18322	0.0099	4b	0.18
Al2	0	0.05475	0.67952	0.0106	2a	0.18
Al3	0	0.40207	0.90205	0.009	2a	0.18
Al4	0	0.4198	0.46093	0.0089	2a	0.18
O1	0.23952	0.91206	0.67534	0.0077	4b	0.225
O3	0	0.17356	0.83943	0.0095	2a	0.225
O4	0	0.19424	0.51918	0.0063	2a	0.225

Table S2 The amount of impurities in $(Sr_{0.85}Ba_{0.15})_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu , $(Sr_{0.85}Ca_{0.15})_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu and $Sr_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu from the results of Rietveld refinements.

Imputiry Phase	Sr_2SiO_4(wt%)	Al_2SiO_5(wt%)
$(Sr_{0.85}Ba_{0.15})_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu	1.2	1.9
$(Sr_{0.85}Ca_{0.15})_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu	2.7	1.7
$Sr_{1.95}Si_{4.1}Al_{0.9}N_{7.1}O_{0.9}$:0.05Eu	11.2	2.9

Figure S5 The particle size distribution of Ba-AlO-258 ($Sr_{1.7}Ba_{0.3}Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9

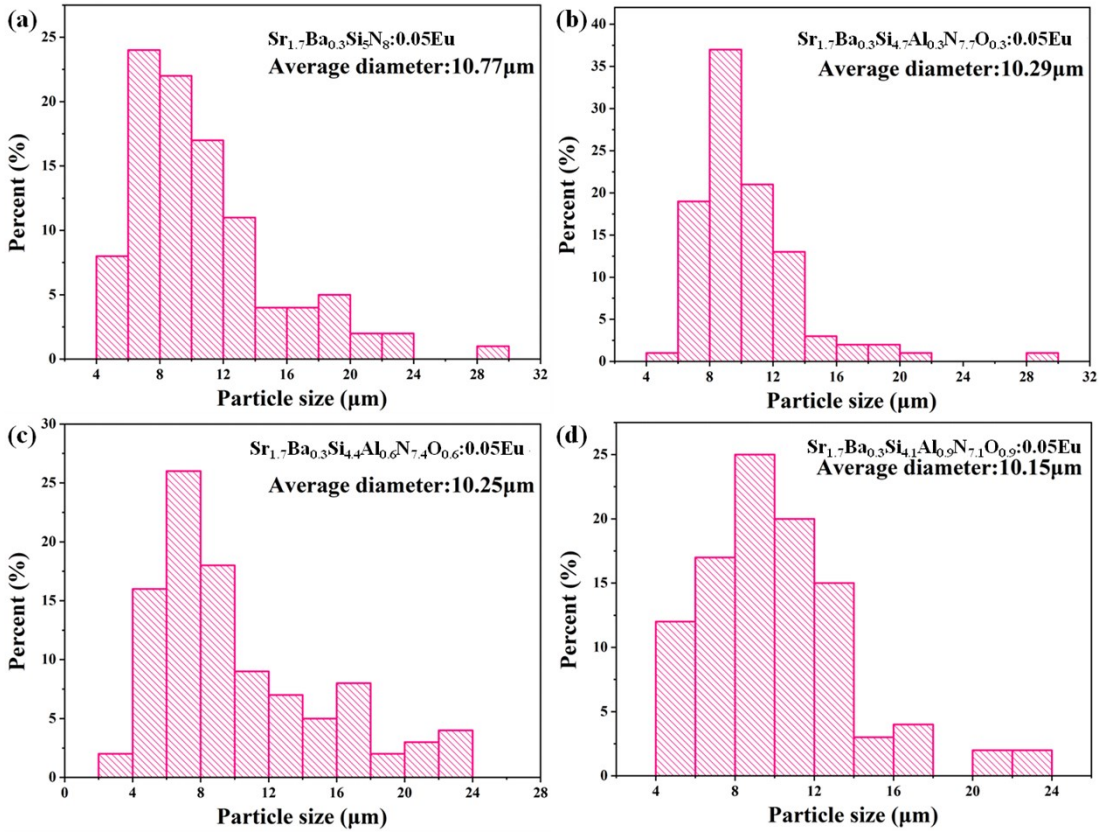


Figure S6 The particle size distribution of Ca-AlO-258 ($Sr_{1.7}Ca_{0.3}Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9

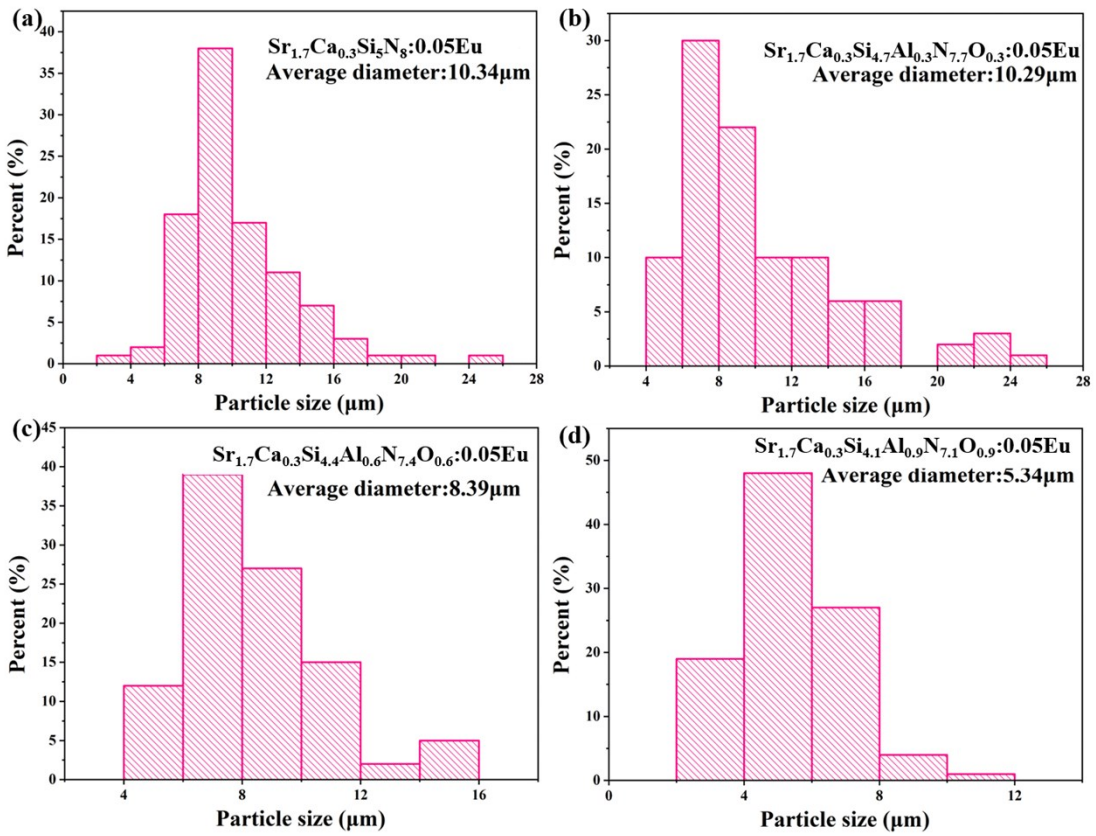


Figure S7 The particle size distribution of AlO-258 ($Sr_2Si_{5-x}Al_xN_{8-x}O_x$:0.05Eu) : (a-d) x=0, 0.3, 0.6, 0.9

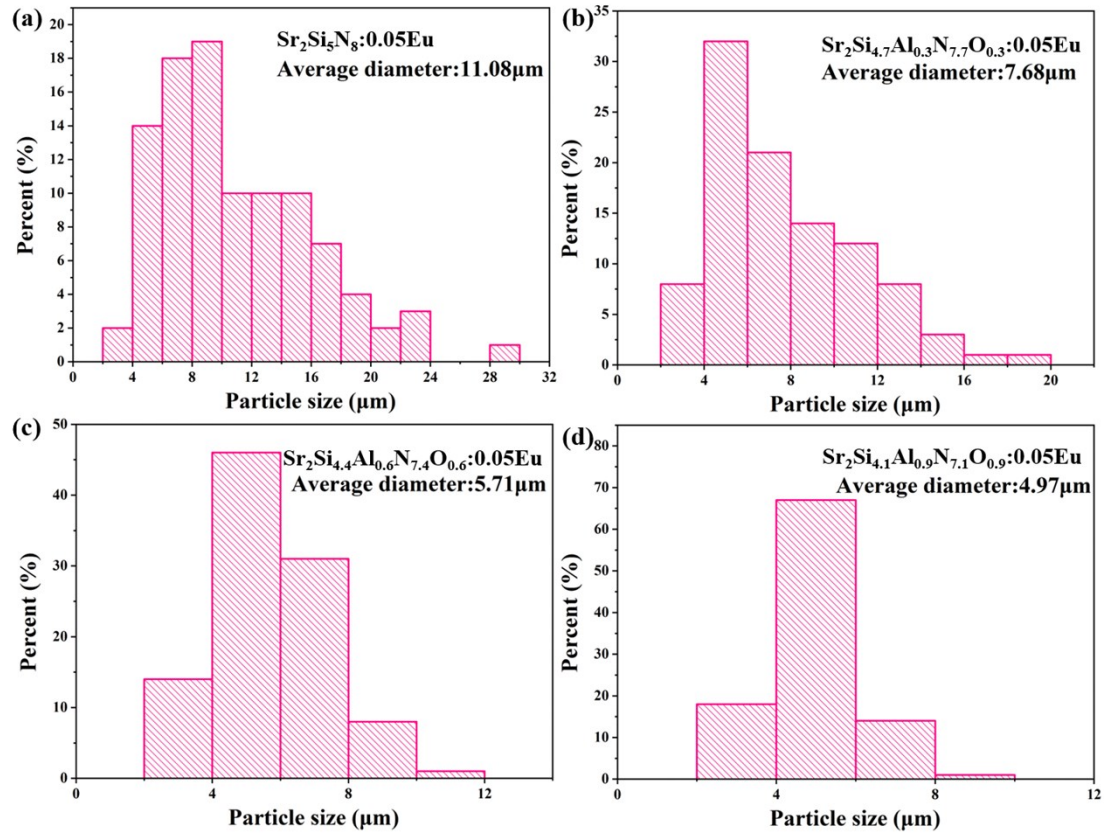
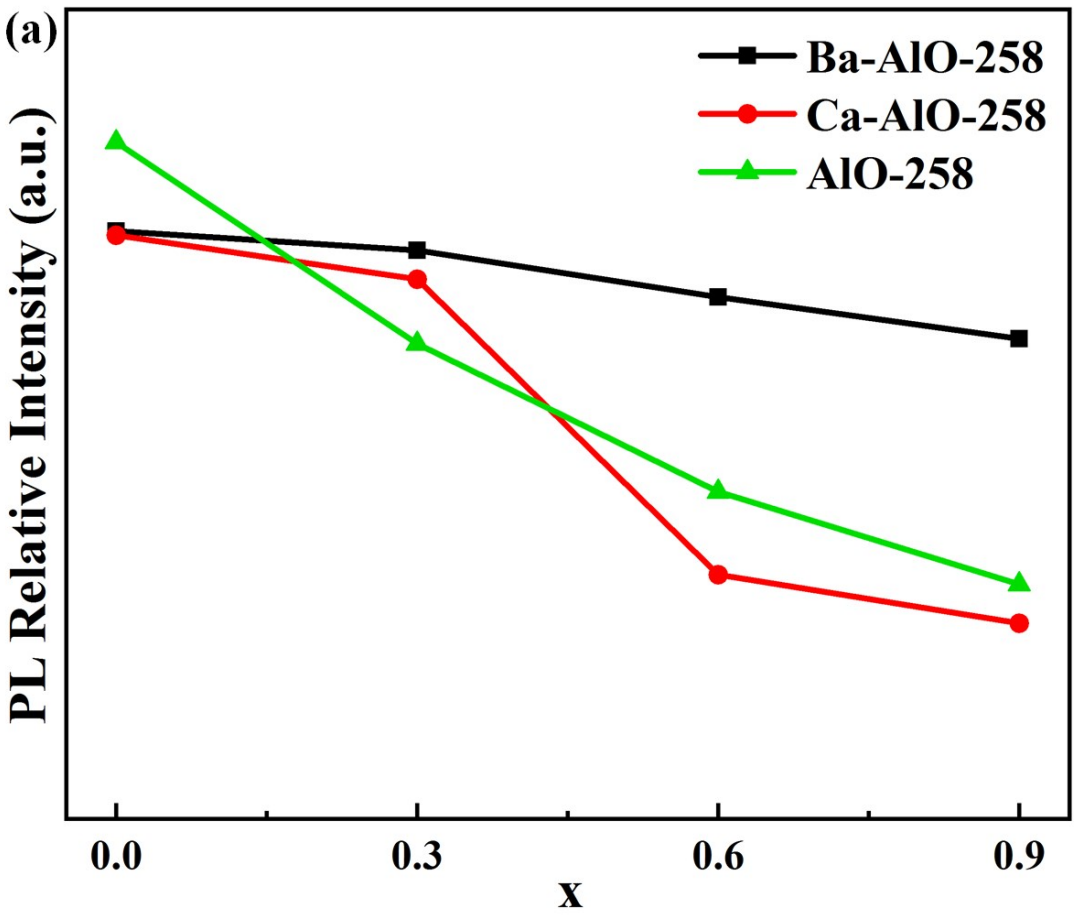


Figure S8 (a) the PL relative intensity and (b)the internal quantum efficiency of the three series. It can be seen that in the three series, the PL intensity of each series decreases with the increasing content of $(AlO)^+$ substitution i.e. x. Among them, the decrease of PL intensity of Ba-AlO-258 series is relatively small; that of Ca-AlO-258 series when $x \leq 0.3$ is relatively small, and that of Ca-AlO-258 series when $x > 0.3$ is relatively large; that of AlO-258 series is relatively large. The change of PL intensity of each series with the increasing x is related to the change of its crystallinity. Compared with the single substitution of $(AlO)^+$, the increase of crystallinity by co-substitution of Ba^{2+}-$(AlO)^+$ and Ca^{2+}-$(AlO)^+$ ($x \leq 0.3$) thus can increase the PL intensity.


(a)
PL Relative Intensity (a.u.)
Ba-AlO-258
Ca-AlO-258
AlO-258
0.0
0.3
0.6
0.9
x

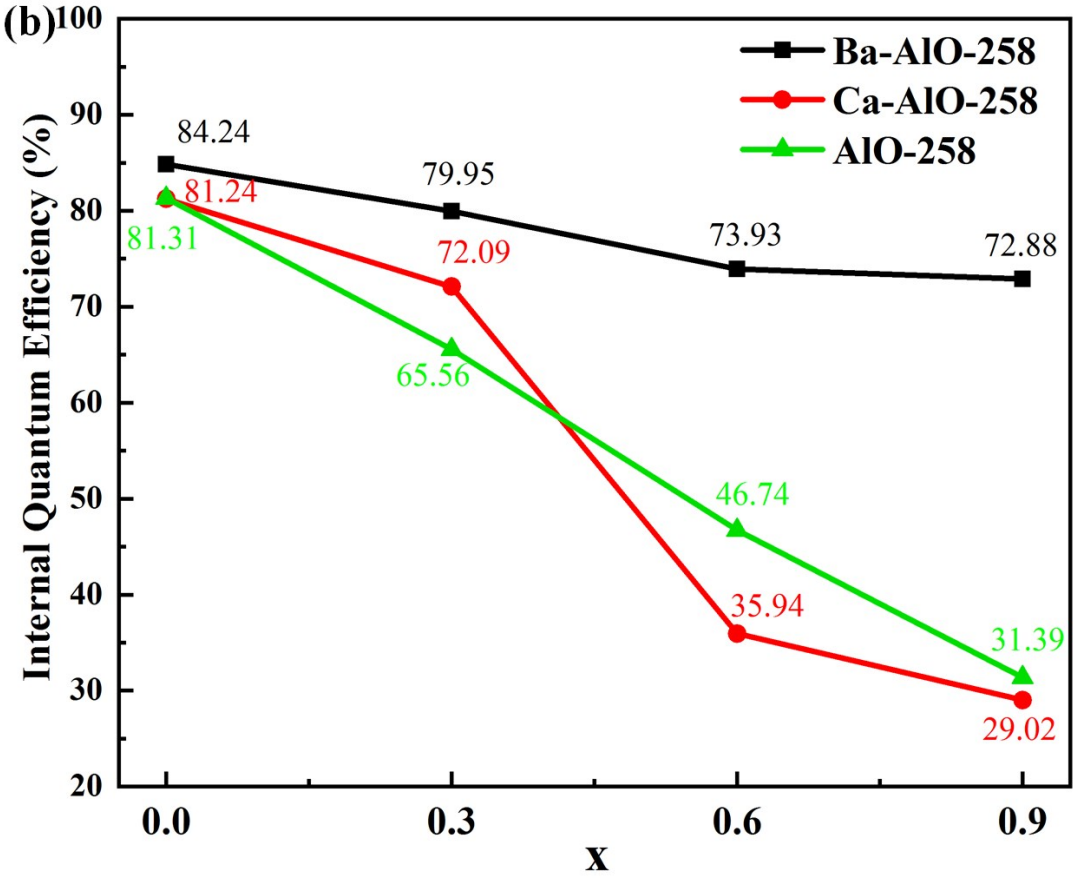

(b)
Internal Quantum Efficiency (%)
Ba-AlO-258
Ca-AlO-258
AlO-258
84.24
79.95
73.93
72.88
81.24
72.09
35.94
29.02
81.31
65.56
46.74
31.39
100
90
80
70
60
50
40
30
20
0.0
0.3
0.6
0.9
x

Table S3 Absorption of three series. The absorption of the three series of samples are very close, so the external quantum efficiency has the same change trend with the internal quantum efficiency. With the increasing content of $(AlO)^+$ substitution i.e. x, the change of quantum efficiency of the each series is consistent with the change of PL relative intensity.

Ba-AlO-258	x=0	x=0.3	x=0.6	x=0.9
Absorption (%)	57.53	61.12	63.38	59.89
Ca-AlO-258	x=0	x=0.3	x=0.6	x=0.9
Absorption (%)	63.67	57.43	61.56	59.11
AlO-258	x=0	x=0.3	x=0.6	x=0.9
Absorption (%)	61.16	60.91	58.83	56.64

Figure S9 Temperature-dependent PL spectra of different kinds of substituted samples : no ion substituted samples ,Ba^{2+}, Ca^{2+},AlO^+ single substituted samples , Ba^{2+}-AlO^+ co-

substituted and Ca^{2+}-AlO^{+} co-substituted samples.

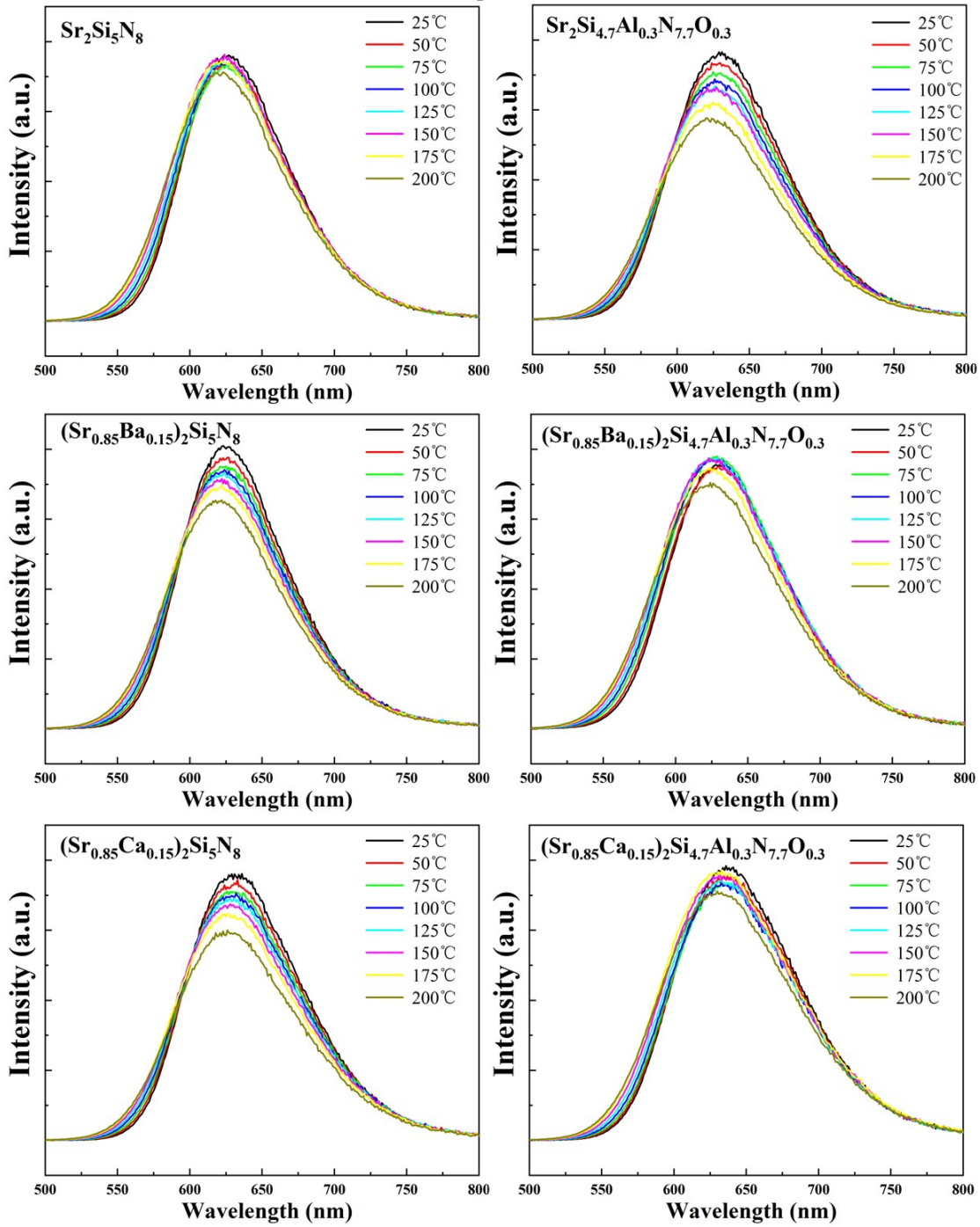


Figure S10 The fluorescence decay curves at room temperature of all samples in the three series: (a)Ba-AlO-258: x=0, 0.3, 0.6, 0.9;(b)Ca-AlO-258: x=0, 0.3, 0.6, 0.9;(c) AlO-258: x=0, 0.3, 0.6, 0.9 .It can be seen that in the three series, with the increase of the content of $(AlO)^{+}$ substitution, the fluorescence decay time decreases. This shows that the substitution of $(AlO)^{+}$ has the same effect on the local coordination environment of Eu^{2+} in three series.

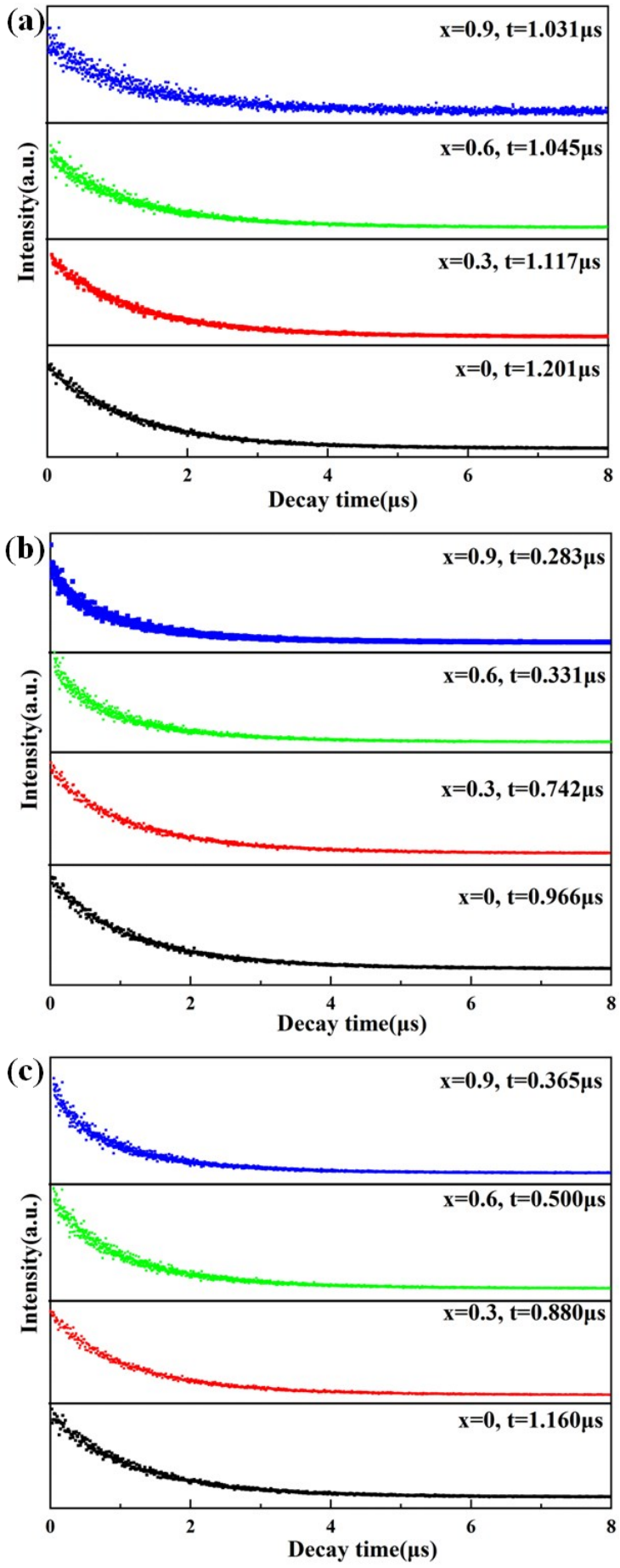

(a)
x=0.9, t=1.031μs
x=0.6, t=1.045μs
x=0.3, t=1.117μs
x=0, t=1.201μs
Intensity(a.u.)
0
2
4
6
8
Decay time(μs)
(b)
x=0.9, t=0.283μs
x=0.6, t=0.331μs
x=0.3, t=0.742μs
x=0, t=0.966μs
Intensity(a.u.)
0
2
4
6
8
Decay time(μs)
(c)
x=0.9, t=0.365μs
x=0.6, t=0.500μs
x=0.3, t=0.880μs
x=0, t=1.160μs
Intensity(a.u.)
0
2
4
6
8
Decay time(μs)

Figure S11 the X-ray diffraction patterns of the phosphors of $(Sr_{1-X},Ba_X)_2Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$: Eu^{2+}, X=0.30,0.45,0.60. With the increase of the substitution of Ba^{2+}, the diffraction peaks in XRD shift to a smaller angle, proving that the lattice expands, indicating that a substitution of Ba^{2+} for Sr^{2+} enters the lattice and the substitution content increases gradually. When the substitution content of Ba^{2+} is 0.30 or more, the impurities of $BaSi_4O_9$ and Al_2SiO_5 appear, but the content is very small.

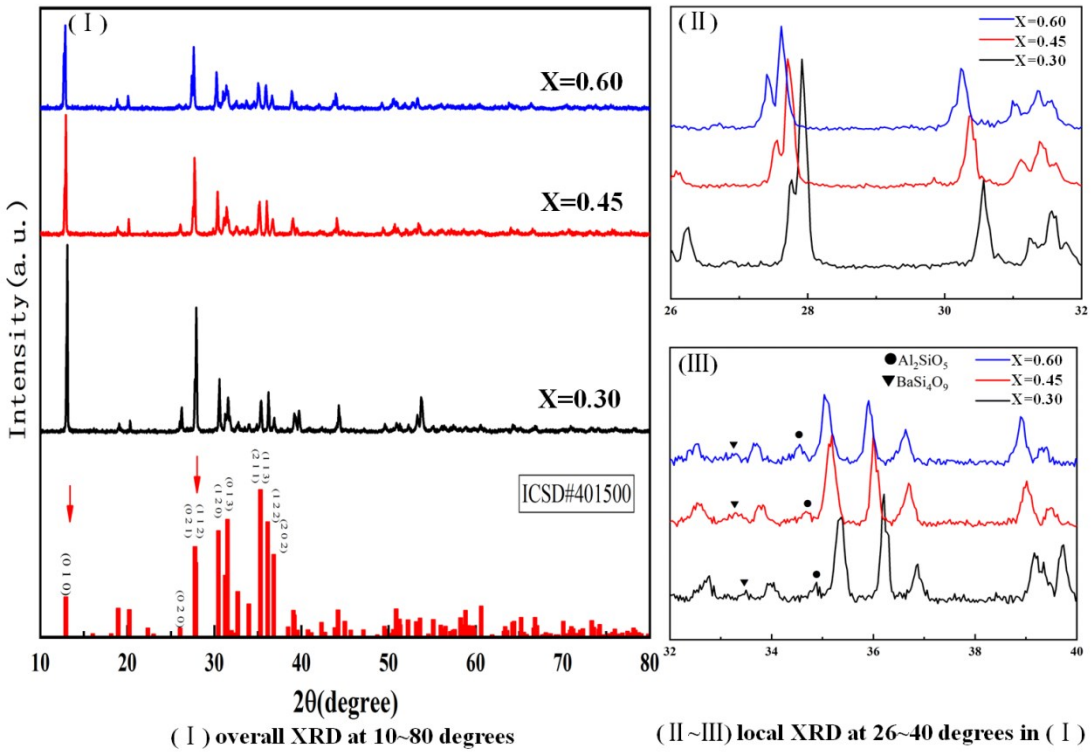


Figure S12 the X-ray diffraction patterns of the phosphors of $(Sr_{1-X},Ca_X)_2Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$: Eu^{2+}, X=0.30,0.45,0.60. With the increase of the substitution of Ca^{2+}, the diffraction peaks in XRD shift to a larger angle, proving that the lattice shrinks, indicating that a substitution of Ca^{2+} for Sr^{2+} enters the lattice and the substitution content increases gradually. When the substitution content of Ca^{2+} is 0.60 or more, the impurities of $Ca_3Al_2O_6$ appear, and the content is small.

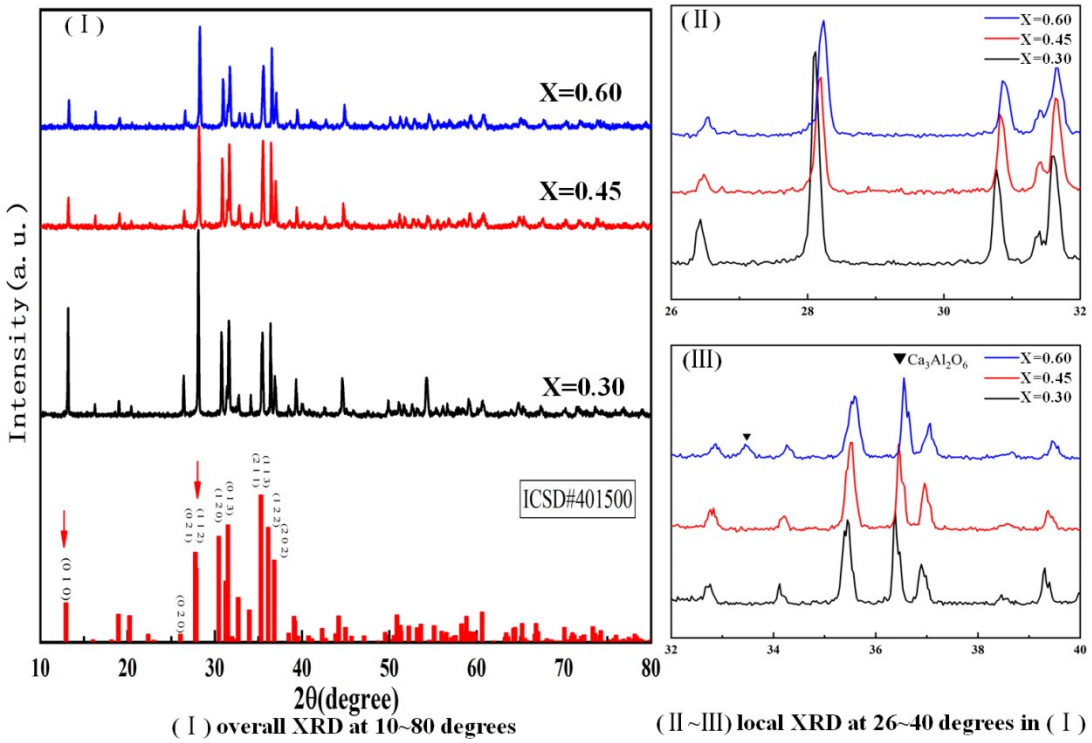


Figure S13 Photoluminescence of the two series: (a)$(Sr_{1-X},Ba_X)_2Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$: Eu^{2+}, X=0.30,0.45,0.60; (b)$(Sr_{1-X},Ca_X)_2Si_{4.7}Al_{0.3}N_{7.7}O_{0.3}$: Eu^{2+}, X=0.30,0.45,0.60. With the increase of the substitution of Ba^{2+}, the emission peak position shifted to blue, which was consistent with that in $(Sr_{1-X}Ba_X)_2Si_5N_8$:Eu^{2+} without $(AlO)^+$ substitution[1]. With the increase of Ca^{2+} substitution, the emission peak redshift is consistent with that in $(Sr_{1-X}Ca_X)_2Si_5N_8$:Eu^{2+} without $(AlO)^+$ substitution[2]. Ba^{2+} replaces Sr^{2+} into the lattice, which expands the lattice, makes the average bond length longer, and the crystal field splitting decreases, so as to cause the blue shift of the spectrum. In the same way, Ca^{2+} replaces Sr^{2+} into the lattice, which shrinks the lattice, makes the average bond length shorter, and the crystal field splitting increases, so as to cause the red shift of the spectrum.

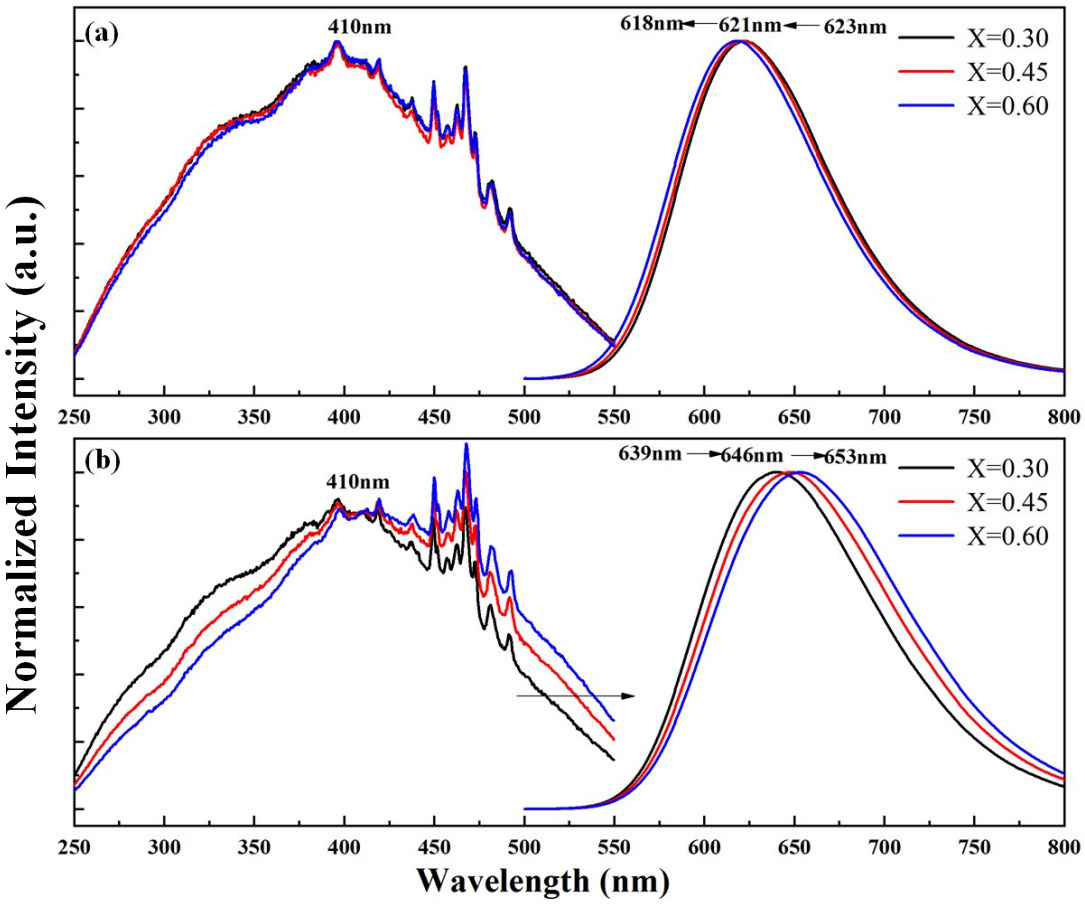
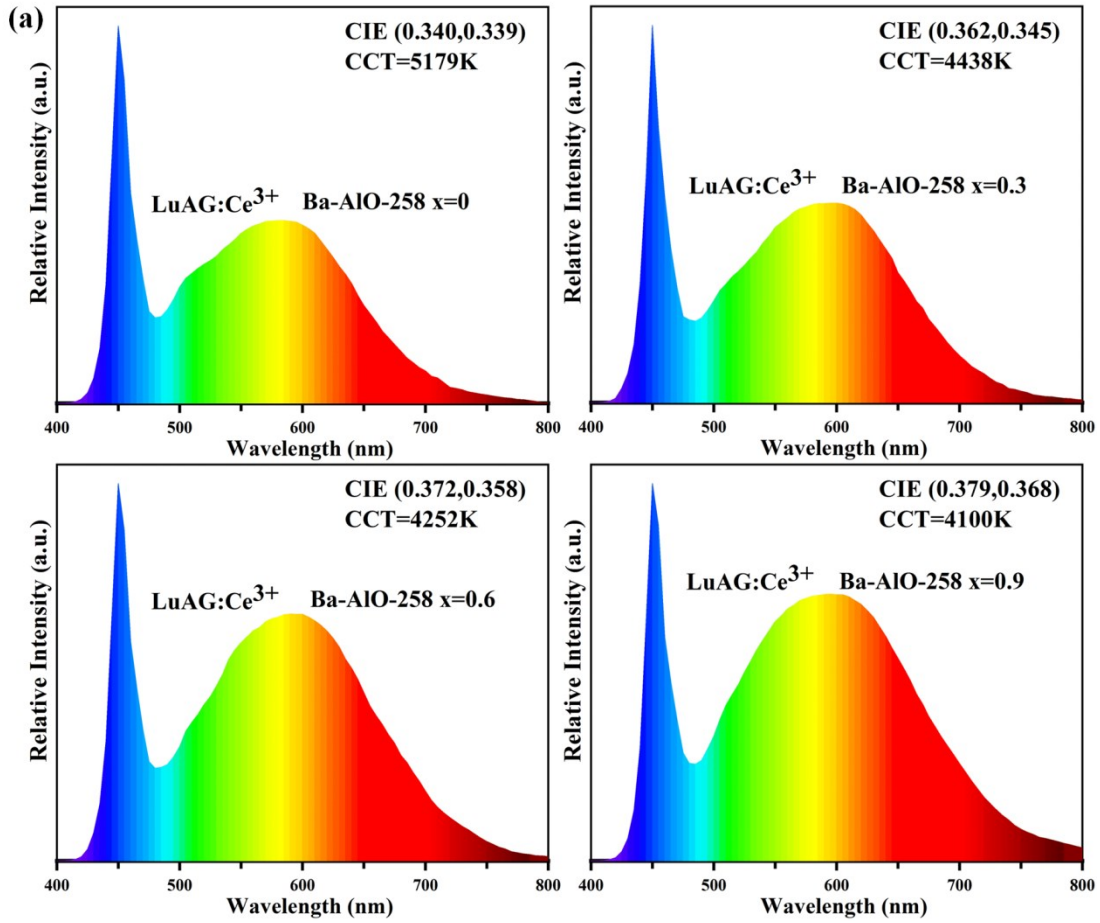
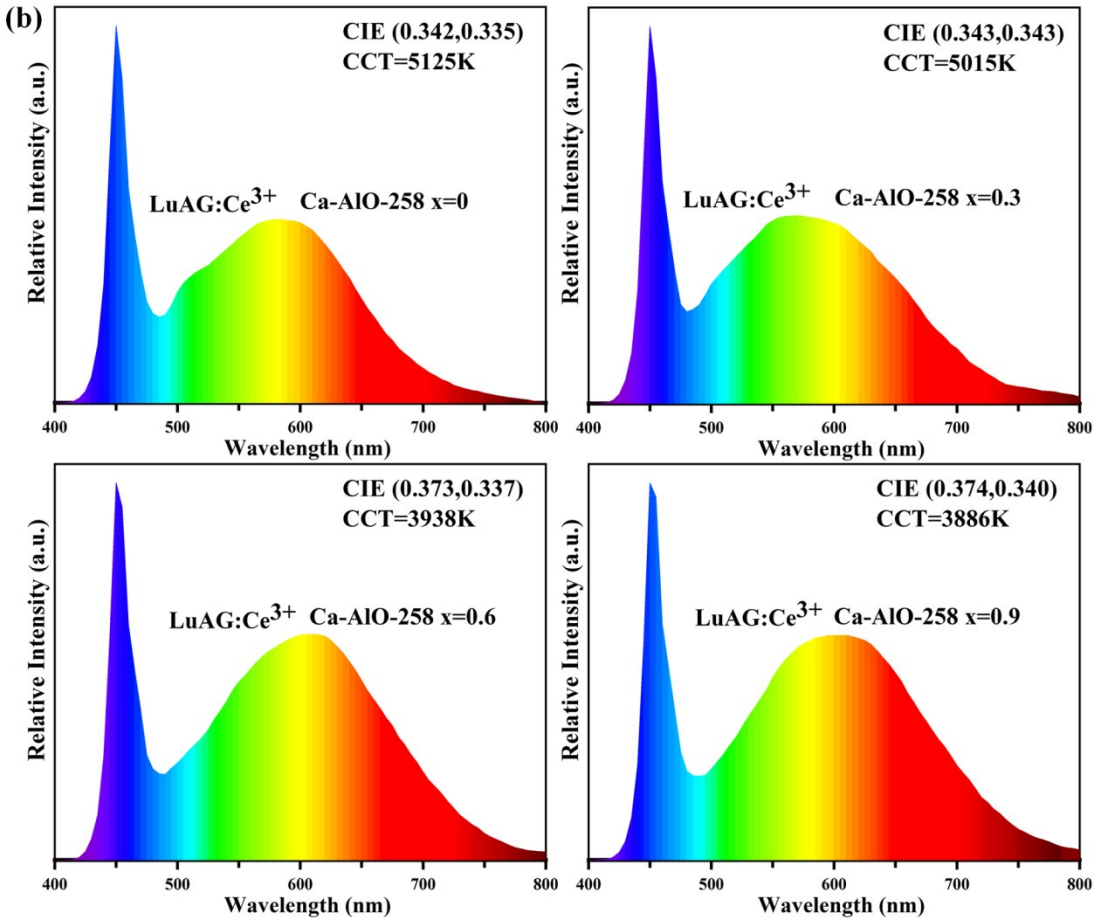


Figure S14 The electroluminescence (EL) spectrums of the WLED lamps fabricated by coating commercial green (LuAG:Ce^{3+}) phosphor and the three series phosphors on a 450nm blue LED chip.


(a)
CIE (0.340,0.339)
CCT=5179K
LuAG:Ce3+ Ba-AlO-258 x=0
CIE (0.362,0.345)
CCT=4438K
LuAG:Ce3+ Ba-AlO-258 x=0.3
CIE (0.372,0.358)
CCT=4252K
LuAG:Ce3+ Ba-AlO-258 x=0.6
CIE (0.379,0.368)
CCT=4100K
LuAG:Ce3+ Ba-AlO-258 x=0.9
Relative Intensity (a.u.)
Wavelength (nm)
400
500
600
700
800


(b)
CIE (0.342,0.335)
CCT=5125K
LuAG:Ce3+ Ca-AlO-258 x=0
CIE (0.343,0.343)
CCT=5015K
LuAG:Ce3+ Ca-AlO-258 x=0.3
CIE (0.373,0.337)
CCT=3938K
LuAG:Ce3+ Ca-AlO-258 x=0.6
CIE (0.374,0.340)
CCT=3886K
LuAG:Ce3+ Ca-AlO-258 x=0.9
Relative Intensity (a.u.)
Wavelength (nm)
400
500
600
700
800

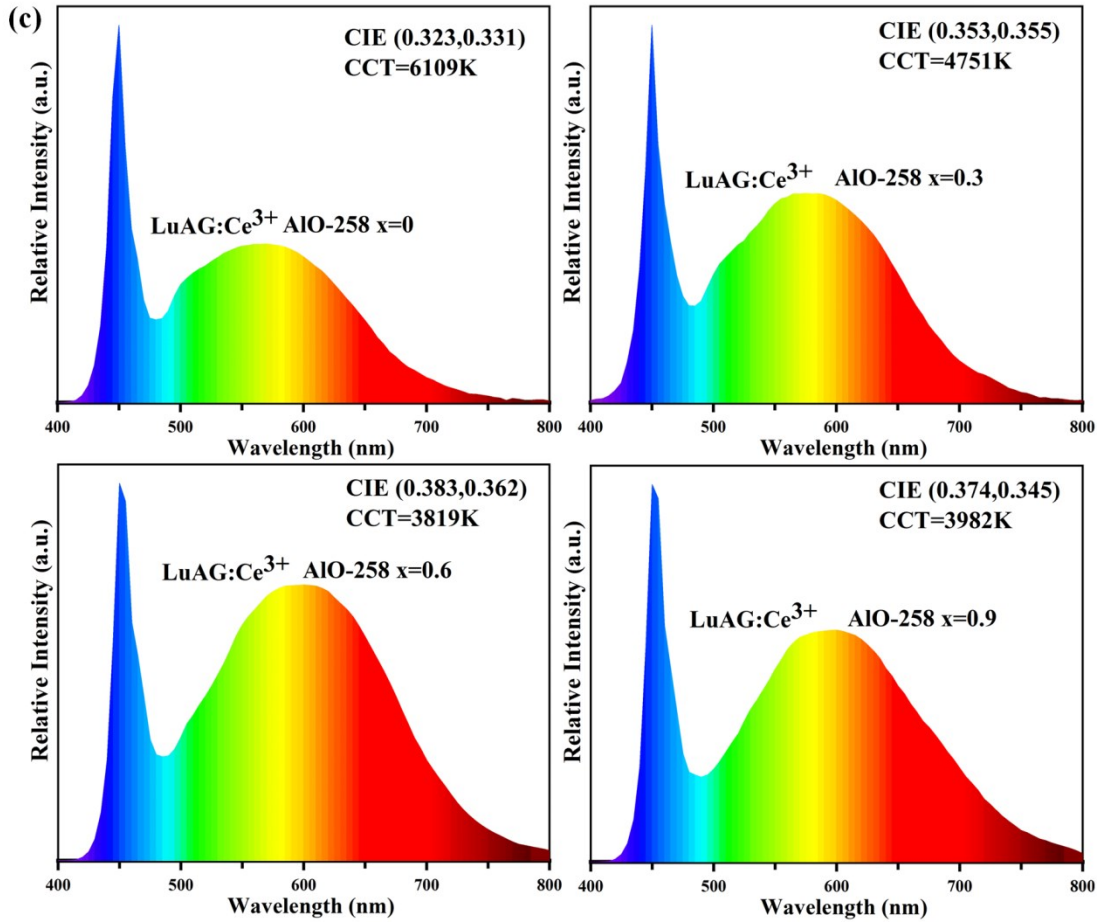


Table S4 luminous efficiency and CRI of three series WLED lamps. With the increasing content of $(AlO)^+$ substitution i.e. x, the change of luminous efficiency of the each series is consistent with the change of quantum efficiency. With the increasing content of $(AlO)^+$ substitution i.e. x, the CRI of each series increases slightly due to the red shift of emission spectrum and the increase of FWHM.

Ba-AlO-258	x=0	x=0.3	x=0.6	x=0.9
luminous efficiency (lm/W)	100.20	94.03	82.00	80.07
Ra	87.1	87.5	88.0	88.6
Ca-AlO-258	x=0	x=0.3	x=0.6	x=0.9
luminous efficiency (lm/W)	94.40	81.70	47.50	44.39
Ra	87.2	88.5	90.9	91.9
AlO-258	x=0	x=0.3	x=0.6	x=0.9
luminous efficiency (lm/W)	96.10	78.10	59.72	49.23
Ra	86.7	87.2	88.7	89.1

Reference

1. Y. H. Liu, L. Chen, X. F. Zhou, R. H. Liu and W. D. Zhuang, *Journal of Solid State Chemistry*, 2017, **246**, 145-149.
2. X. Teng, Y. Liu, Y. Liu, H. Yunsheng, H. He and W. Zhuang, *Journal of Rare Earths*, 2009, **27**, 58-61.