Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Discovery of a Ce^{3+}-Activated Red Nitride Phosphor for High-Brightness Solid-State Lighting

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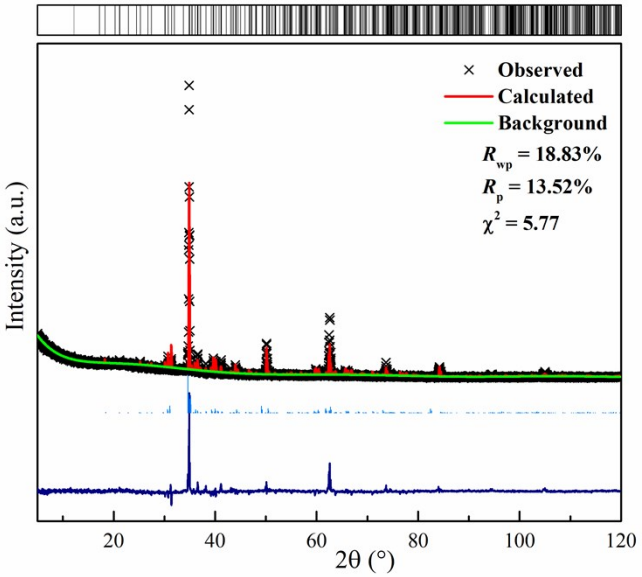

Figure S1. Rietveld refinement of observed HP-$CaSiN_2$ XRD pattern on the basis of AP-$CaSiN_2$ structure.

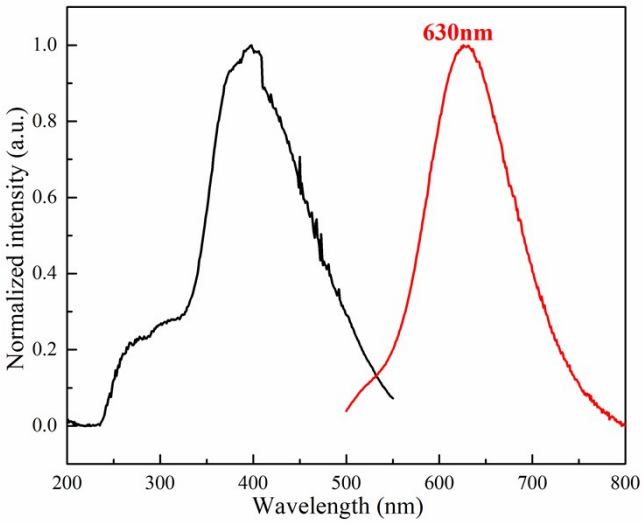

Figure S2. Excitation and emission spectra of none-doped HP-$CaSiN_2$.

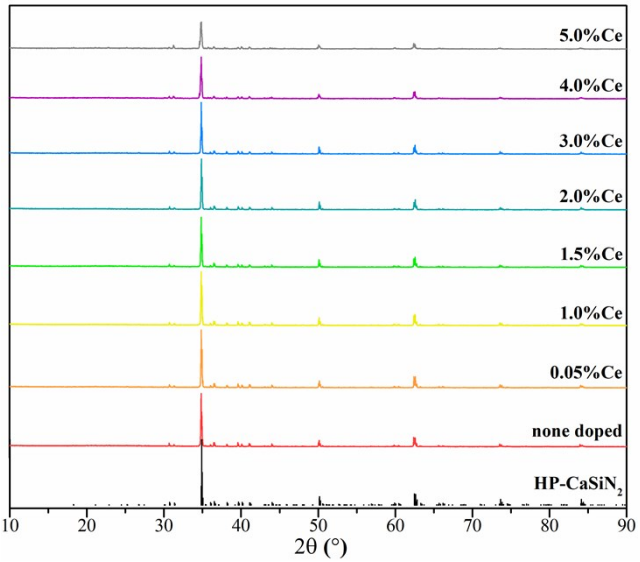

Figure S3. XRD patterns for HP-$Ca_{(1-x)}SiN_2$:xCe^{3+} (x = 0-5.0%).

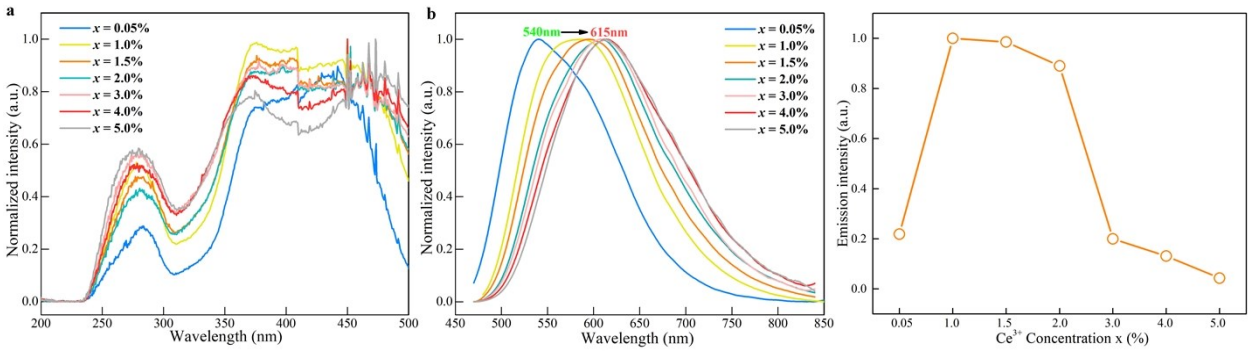

Figure S4. (a) The excitation (λ_{em} = 610 nm), (b) emission spectra (λ_{ex} = 450 nm) and emission intensity of HP-$Ca_{(1-x)}SiN_2$:xCe^{3+}.

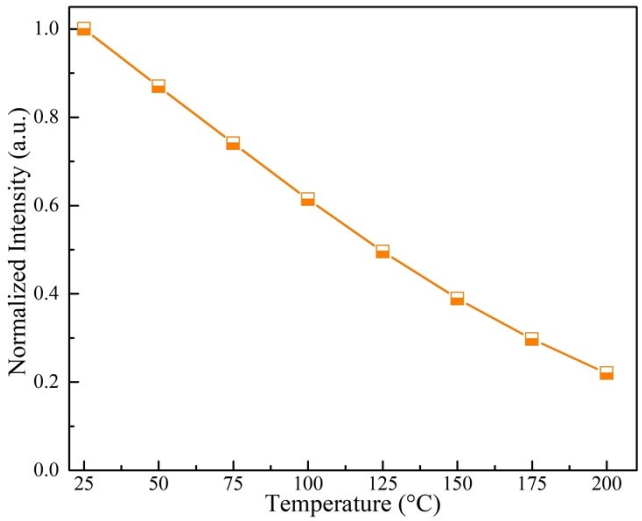

Figure S5. Temperature-dependent normalized integrated PL intensities of HP-$Ca_{0.98}SiN_2$:0.02Ce^{3+}.

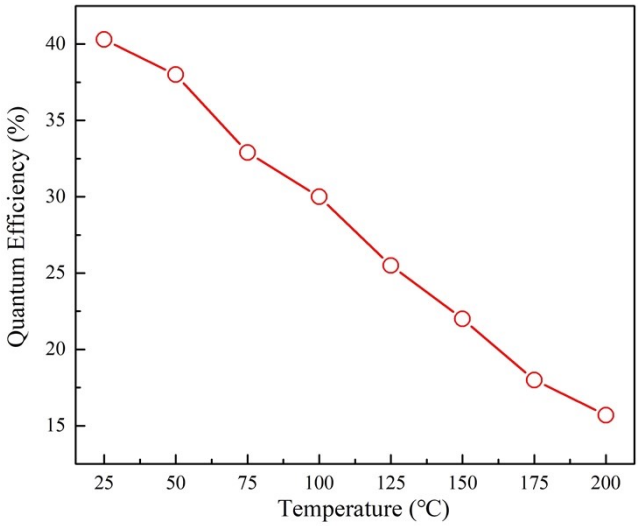

Figure S6. Temperature-dependent quantum efficiency of HP-$Ca_{0.98}SiN_2$:0.02Ce^{3+}.

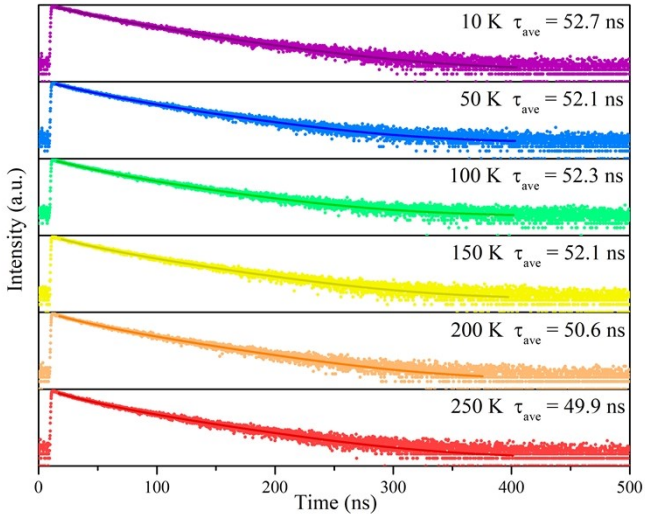

Figure S7. Temperature-dependent decay curves of HP-$Ca_{0.98}SiN_2$:0.02Ce^{3+} monitored at 610 nm, excited at 450 nm.

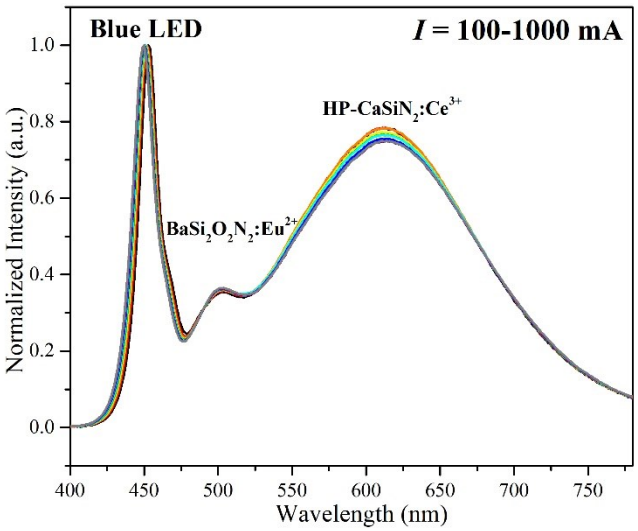

Figure S8. Luminescence spectra of the fabricated white LED driven by current increasing from 100 to 1000 mA.

Table S1. The space group, band gap, average bond length (R_{av}) and distortion index (D) of potential nitridosilicate candidates.

Nitride	Space group	Band gap (PBE)[a]	R_{av}[b]	distortion index (D)[b]
$SrSi_6N_8$	*Imm*2	3.21[1]	3.028	0.048
$BaSi_6N_8$	*Imm*2	3.25[1]	3.062	0.040
$SrSi_7N_{10}$	*Pc*	4.28[1]	3.268	0.082
$BaSi_7N_{10}$	*Pc*	4.11[1]	3.247	0.043
$Ca_2Si_5N_8$	*Cc*	3.65[1]	2.638	0.082
$Sr_2Si_5N_8$	$Pmn2_1$	3.20[1]	2.866	0.068
$Ba_2Si_5N_8$	$Pmn2_1$	2.88[1]	2.917	0.046
$LaSi_3N_5$	$P2_12_12_1$	3.19[1]	2.700	0.049
$La_3Si_6N_{11}$	P4*bm*	2.97[1]	2.647	0.001
$ZnSiN_2$	$Pna2_1$	4.83[2]	2.109	0.010
$MgSiN_2$	$Pna2_1$	3.97[1]	2.090	0.011
Cubic-$CaSiN_2$ ($Ca_{16}Si_{17}N_{34}$)	F3m	4.50[3] (optical band gap)	2.481	0.079
AP-$CaSiN_2$ (ambient-pressure phase)	*Pbca*	3.44	2.506	0.046
HP-$CaSiN_2$ (high-pressure phase)	*Pbca*	**3.44**	**2.570**	**0.086**
$SrSiN_2$	$P2_1/c$	2.97[1]	2.843	0.069
$BaSiN_2$	*Cmca*	2.92[1]	2.986	0.061
$La_5Si_3N_9$	*Cmca*	1.97[1]	2.553	0.074
$Ca_5Si_2N_6$	*C*2/*c*	2.46[1]	2.447	0.023
$Ba_5Si_2N_6$	$P2_12_12_1$	1.40[1]	2.977	0.073
Ca_4SiN_4	$P2_1/c$	1.45[1]	2.454	0.034

$SrMg_3SiN_4$	$I4_1/a$	4.90[4] (optical band gap)	2.863	0.049
$CaMg_3SiN_4$	$I4_1/a$	2.60[5]	2.733	0.052
BaMg3SiN4	P	4.00[4] (optical band gap)	2.953	0.015
$Ca_3Li_4Si_2N_6$	$C2/m$	2.29[1]	2.580	0.045
$Sr_3Li_4Si_2N_6$	$C2/m$	unknown	2.706	0.022
$SrAlSi_4N_7$	$Pna2_1$	3.58[1]	2.713	0.054
$Ba_2AlSi_5N_9$	$P1$	unknown	2.955	0.030
$SrYSi_4N_7$	$P6_3mc$	2.74[1]	3.012	0.015
$CaLi_2Si_2N_4$	Pa	3.46[6]	2.499	0.004
$SrLi_2Si_2N_4$	Pa	3.44[1]	2.689	0.001
$CaAlSiN_3$	$Cmc21$	3.36[7]	2.470	0.053
$Ca_5Al_2Si_2N_8$	Pbcn	2.79[1]	2.526	0.033
$Sr_8Mg_7Si_9N_{22}$	$C2/m$	2.90[8]	2.764	0.035
$Ca_3LiSi_2N_5$	$C2/c$	2.44[1]	2.511	0.039
$La_{17}Al_4Si_9N_{33}$	$F3m$	1.88[1]	2.578	0.011
$Li_5La_5Si_4N_{12}$	$Pb2$	unknown	2.580	0.032
$Sr_4Li_2Si_2N_6$	Pa	2.20[1]	2.671	0.029
$Ca_2Ba_3Si_2N_6$	$C2/c$	1.55[1]	2.513	0.040
$MgBa_4Si_2N_6$	$Fddd$	1.59[1]	2.845	0.047
$Li_2Ca_2Mg_2Si_2N_6$	$C2/m$	4.26[9] (optical band gap)	2.588	0.041
$Li_2Ca_3MgSi_2N_6$	$C2/m$	unknown	2.513	0.038
$Li_4Ca_2MgSi_2N_6$	$C2/m$	unknown	2.482	0.015
$BaLi_2Al_2Si_2N_6$	$P4/ncc$	4.60[10] (optical band gap)	3.017	0.027
$KLaSi(CN_2)_4$	$P2_122_1$	unknown	2.609	0.018
$RbLaSi(CN_2)_4$	I	unknown	2.607	0.019

[a] (The optical band gap is multiplied by 67% to convert to an approximate PBE band gap.);
[b](The values of R_{av} and D are collected from ICSD; For multi-lattice structures, the smallest R_{av} and corresponding D values are selected.)

Table S2. Selected bond lengths of HP-$CaSiN_2$:Ce^{3+}.

Atom	Distance (Å)	Atom	Distance (Å)
(Ca1/Ce1) – N1	2.371(34)	(Ca2/Ce2) – N1	2.546(26)
(Ca1/Ce1) – N2	2.497(30)	(Ca2/Ce2) – N1	2.971(28)
(Ca1/Ce1) – N2	2.928(29)	(Ca2/Ce2) – N2	2.824(31)
(Ca1/Ce1) – N3	2.559(31)	(Ca2/Ce2) – N3	2.507(30)
(Ca1/Ce1) – N3	2.068(30)	(Ca2/Ce2) – N4	2.598(29)
(Ca1/Ce1) – N4	2.927(27)	(Ca2/Ce2) – N4	2.601(30)
		(Ca2/Ce2) – N4	2.965(29)

Table S3. CIE color coordinates, CRI and CCT of the fabricated white LED.

Current (mA)	CIE color coordinates	CRI	CCT (K)
100	(0.4012, 0.3527)	89.8	3243
200	(0.4003, 0.3515)	90.1	3254
300	(0.3993, 0.3504)	90.4	3268
400	(0.3975, 0.3486)	90.7	3294
500	(0.3960, 0.3468)	90.9	3312
600	(0.3952, 0.3459)	91.1	3322
700	(0.3937, 0.3438)	91.2	3341
800	(0.3927, 0.3423)	91.4	3351
900	(0.3919, 0.3413)	91.5	3360
1000	(0.3915, 0.3408)	91.5	3365

Notes and references

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