Supplementary Information (SI) for RSC Advances.
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Supporting Information

Novel Efficient Deep-Red Emitting Phosphor $SrCa_2Ga_2O_6:Mn^{4+}$ with Tululite-Related Structure

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To verify the validity of single crystal XRD analysis including manganese, the PL spectra determination was performed. As shown in Fig. S1, the normalized PL spectra of $SrCa_2Ga_2O_6:Mn^{4+}$ single crystal was well matched to that of $SrCa_2Ga_2O_6:0.03Mn^{4+}$ powder. In a similar purpose, EPMA measurement was acted into $SrCa_2Ga_2O_6:Mn^{4+}$ single crystals. Semiquantitative measurement for carbon-coated single crystals was performed with an electron probe microanalyzer *EPMA-1720* (Shimadzu corp.). The EPMA measurement result is summarized in Table S1. The selected elements were Sr, Ca, Ga, Mn, O, Al, and Cl. Manganese was detected in all points. Aluminum was expected to be derived from the alumina boat, and chlorine from the residue of $SrCl_2$ flux. The structural analysis excluding Al and Cl was appropriate since those atoms were barely detected. From the above results, the single crystal XRD analysis including manganese was valid.

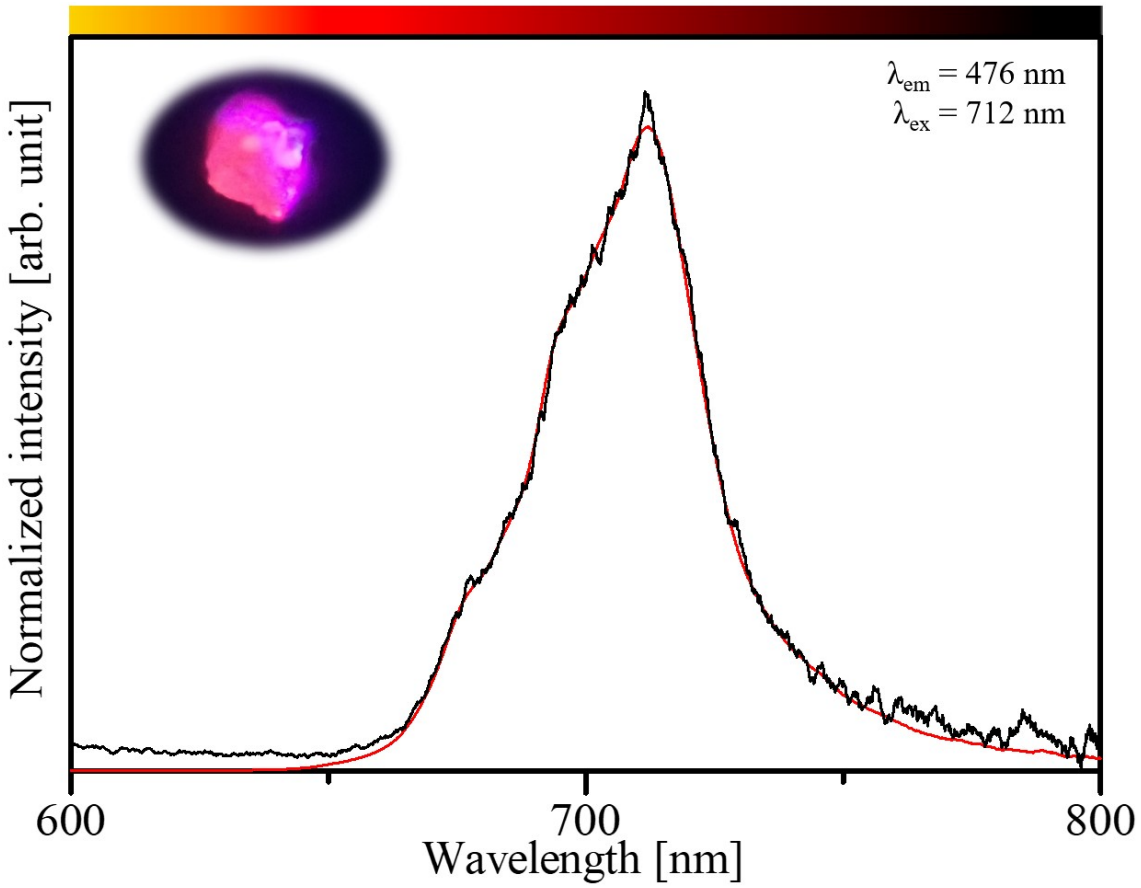

Figure S1. Normalized PL spectra of $SrCa_2Ga_2O_6:Mn^{4+}$ single crystals (black line) and $SrCa_2Ga_2O_6:0.03Mn^{4+}$ powder (red line). The insert photograph is the deep-red emitting $SrCa_2Ga_2O_6:Mn^{4+}$ single crystal under 365 nm excitation

Table S1. EPMA measurement result for $SrCa_2Ga_2O_6:Mn^{4+}$ single crystals.

	Sr(mol%)	Ca(mol%)	Ga(mol%)	Mn(mol%)	O(mol%)	Al(mol%)	Cl(mol%)
1	12.29	27.63	16.28	0.97	42.36	0.18	0.29
2	9.04	23.41	11.07	1.34	54.75	0.13	0.26
3	13.94	30.87	7.07	0.58	47.28	0.08	0.17
4	21.05	19.71	27.76	0.78	29.02	0	1.68
5	17.06	37.4	27.99	3.27	14.03	0.08	0.17

Table S2. Crystallographic data of $SrCa_2Ga_2O_6$:0.03Mn^{4+} powder obtained by Rietveld refinement.

Chemical formula	$SrCa_2Ga_{1.94}Mn_{0.06}O_6$
Crystal system	Cubic
Space group	*F*432 (#209)
a (Å)	15.4888(6)
V (Å^3)	3715.8(2)
Z	24
R_{wp} (%)	9.316
R_p (%)	6.556
R_e (%)	3.705
S	2.515
Phase 1 - SCG:0.03Mn^{4+} (Mass%)	93.74
Phase 2 - $Sr_3Ga_2O_6$ (Mass%)	0.40
Phase 3 - $Ca_5Ga_6O_{14}$ (Mass%)	5.86

Table S3. Atomic positions, occupancies, and isotropic displacement parameters for $SrCa_2Ga_2O_6$:0.03Mn^{4+} powder obtained by Rietveld refinement.

Atom	Site	Occ.*	*x*	*y*	*z*	B_{eq} (Å^2)
Sr1	24e	1	0.2082(1)	0	0	1
Ca1	32f	1	0.3896(1)	0.3896(1)	0.3896(1)	1
Ca2	32f	0.5	0.1556(2)	0.1556(2)	0.1556(2)	1
Ga1/Mn1	4b	0.64/0.36	0.5	0.5	0.5	1
Ga2	32f	0.5	0.1781(1)	0.1781(1)	0.1781(1)	1
Ga3	24d	1	0	0.25	0.25	1
Ga4	4a	1	0	0	0	1
O1	24e	1	0.3762(7)	0	0	1
O2	8c	1	0.25	0.25	0.25	1
O3	96j	1	0.2511(6)	0.9350(3)	0.1495(4)	1
O4	32f	0.5	0.0694(6)	0.0694(6)	0.0694(6)	1

*Occ. means occupancy.

Table S4. Results of XRF quantitative analysis for $SrCa_2Ga_2O_6$:0.03Mn^{4+}.

x	**Sr (at%)**	**Ca (at%)**	**Ga (at%)**	**Mn (at%)**
1	20.03	40.80	37.95	1.22
2	19.95	40.95	37.88	1.22
3	19.99	40.93	37.88	1.20

Table S5. Results of PL decay curve fitting for $SrCa_2Ga_2O_6$:*x*Mn^{4+} ($0.005 \leq x \leq 0.07$).

x	τ_1 **(ms)**	τ_2 **(ms)**	τ_{ave} **(ms)**
0.005	2.490	-	2.573
0.01	2.423	-	2.516
0.03	2.342	4.909	2.443
0.05	2.493	1.712	2.331
0.07	2.163	0.954	2.026

The morphology of SCG:0.03Mn^{4+} was investigated by SEM measurement. Fig. S2 displays the SEM image of SCG:0.03Mn^{4+} phosphor ground by the agate mortar. The particles ranging from sub-micrometer to 10 micrometers were obtained in this study. The morphology of the particles was unevenly polygonal-shape.

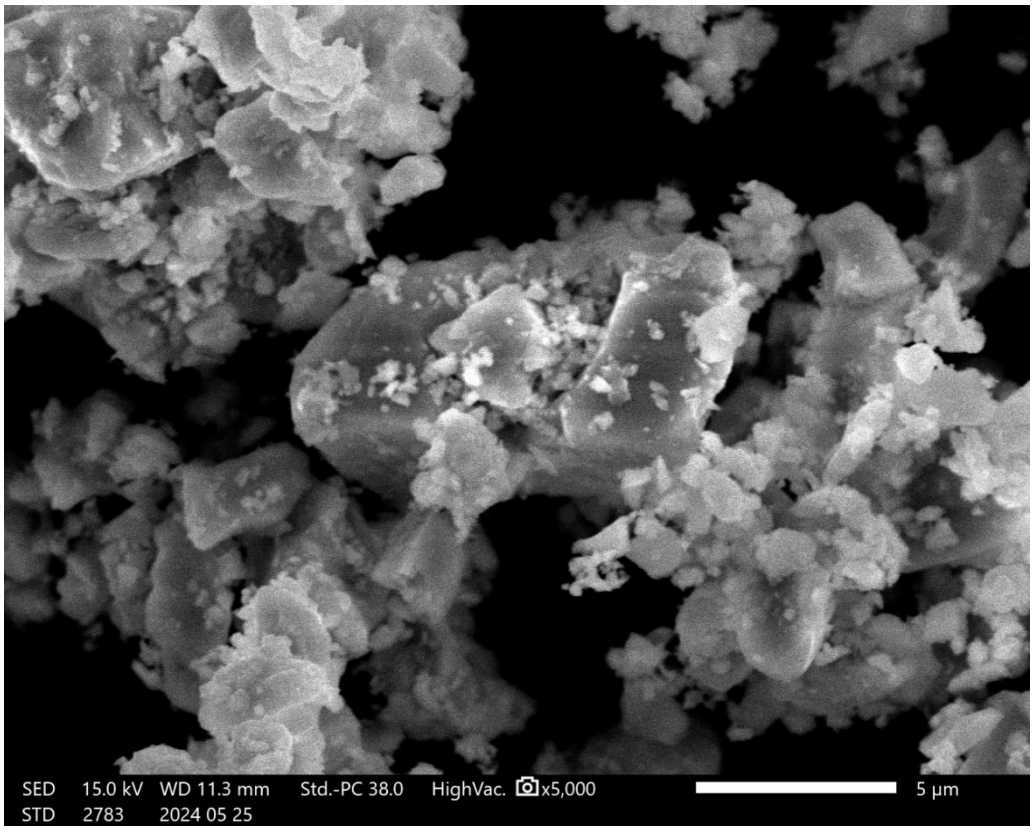

Figure S2. SEM image of $SrCa_2Ga_2O_6$:0.03Mn^{4+} powder after grinding.

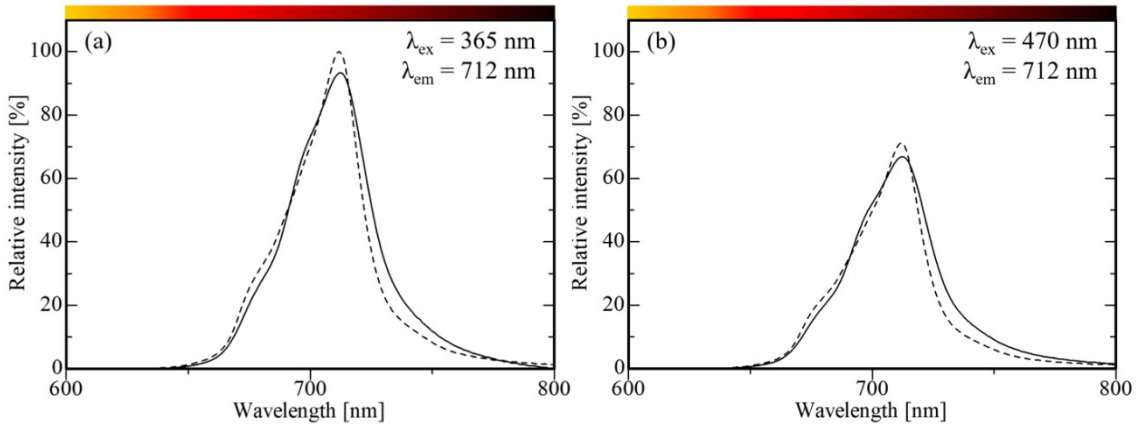

Figure S3. Normalized PL spectra of $SrCa_2Ga_2O_6$:0.03Mn^{4+} (solid lines) and $Ca_{14}Zn_6Ga_{10}O_{35}$:Mn^{4+} (dashed lines) phosphors excited by (a) 365 nm n-UV light and (b) 470 nm blue light.

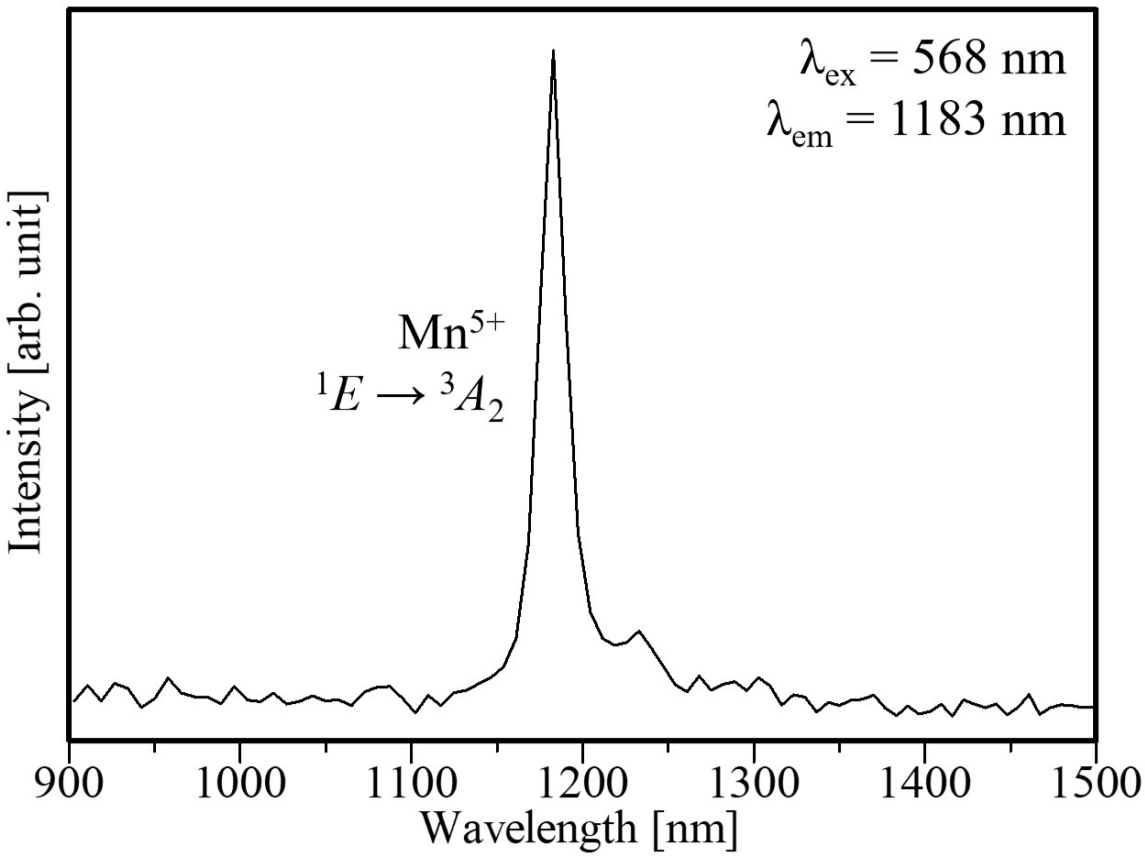

Figure S4. PL spectrum of tetrahedrally coordinated Mn^{5+} in

$SrCa_2Ga_2O_6$:0.03Mn^{4+} phosphor excited under 568 nm yellow LED light.