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

Supporting Information

Tailoring photoluminescence stability in double perovskite red phosphors $A_2BAlF_6:Mn^{4+}$ (A = Rb, Cs; B = K, Rb) via neighboring-cation modulation

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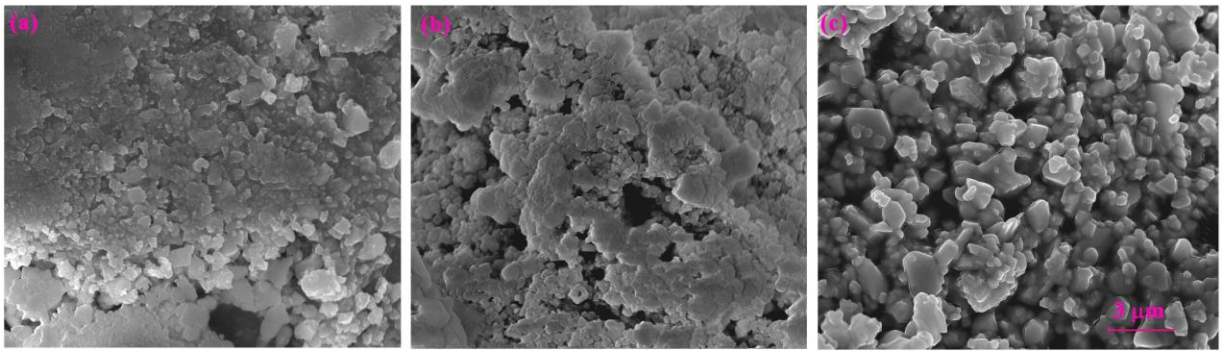

Fig. S1 SEM images of $Rb_2KAlF_6:Mn^{4+}$ (a), $Cs_2KAlF_6:Mn^{4+}$ (b), and $Cs_2RbAlF_6:Mn^{4+}$ (c) synthesized via same reaction environment. The scale bar in the SEM images is fixed at 3 μm.

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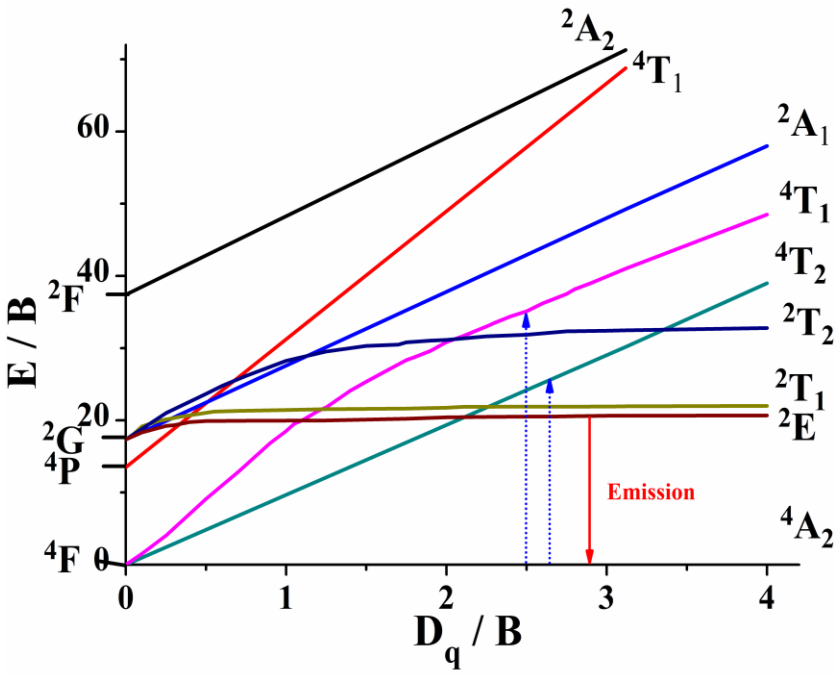

Fig. S2 Tanabe−Sugano energy-level diagram of Mn^{4+} in an octahedral crystal field.

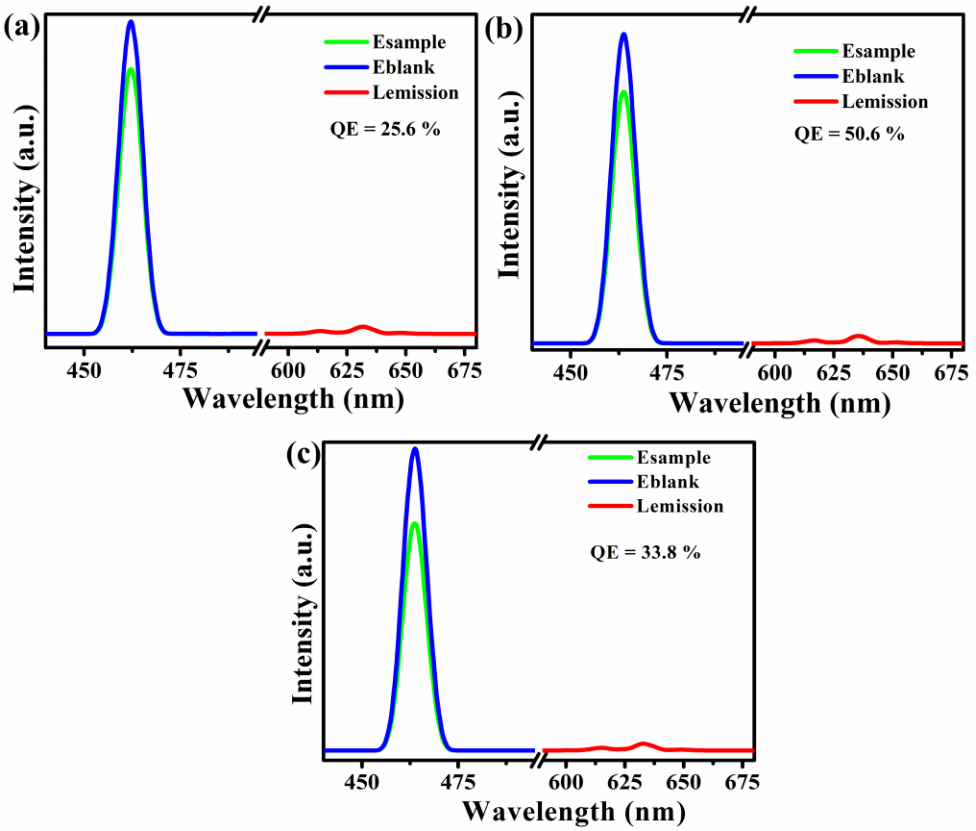

Fig. S3 Excitation line of blank ($BaSO_4$) (*E*blank) and excitation (*E*sample) and emission (*L*emission) spectra of $Rb_2KAlF_6:Mn^{4+}$ (a), $Cs_2KAlF_6:Mn^{4+}$ (b), and $Cs_2RbAlF_6:Mn^{4+}$ (c). The quantum efficiency (QE) can be calculated as follows:

$$QE = \frac{\int Lemission}{\int Eblank - \int Esample} \quad (1)$$

where *L*emission and *E*sample are the emission and excitation spectra of objective samples; *E*blank is the spectrum of the excitation light without samples in integrating sphere. All of spectra were recorded by an optical integrating sphere coupled to the FLS920 fluorescence spectrophotometer at room temperature.

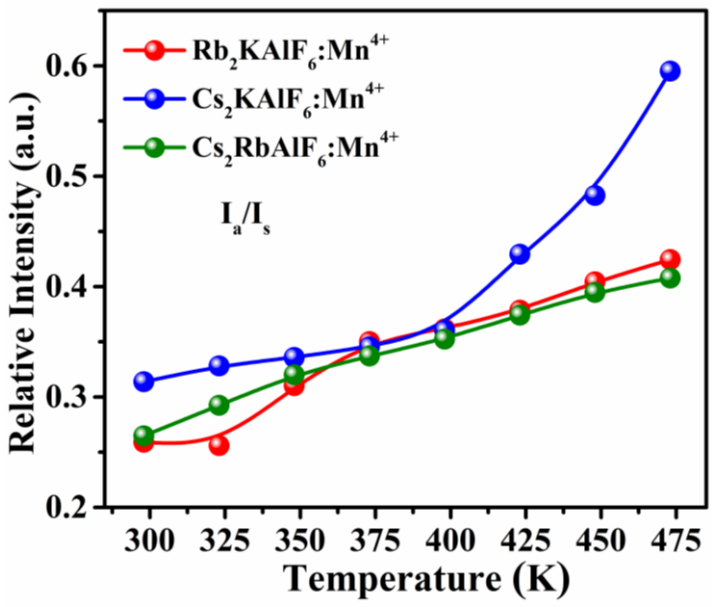

Fig. S4 The ratio of I_a / I_s for $A_2BAlF_6:Mn^{4+}$ (A= Rb, Cs; B = K, Rb) as a function of temperature at 298 - 473 K.

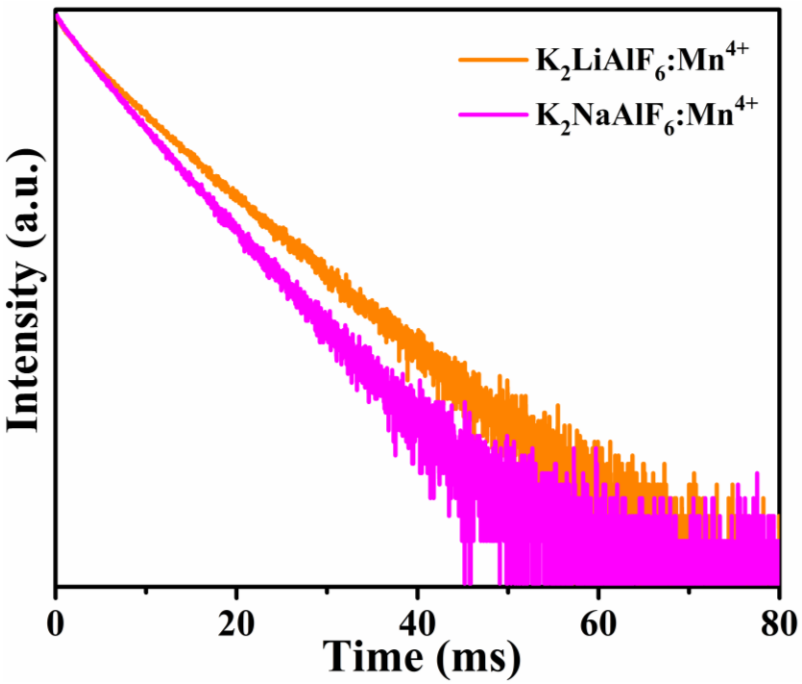

Fig. S5 PL decay curves of $K_2BAlF_6:Mn^{4+}$ (B = Li, Na) at room temperature.

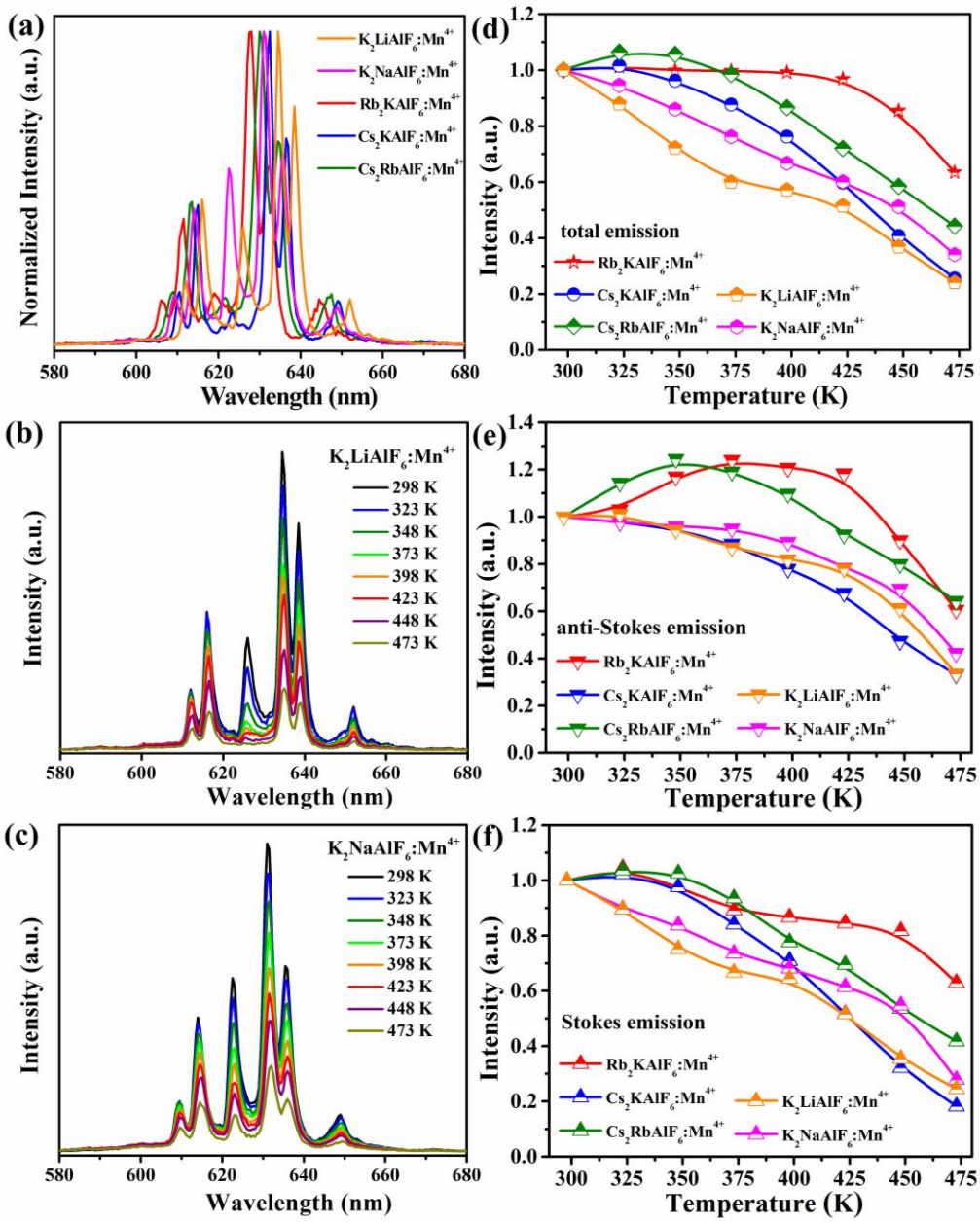

Fig. S6 Normalized PL spectra of $A_2BAlF_6:Mn^{4+}$ (A = K, Rb, Cs; B = Li, Na, K, Rb) (a). Temperature-dependent PL spectra of phosphors $K_2LiAlF_6:Mn^{4+}$ (b) and $K_2NaAlF_6:Mn^{4+}$ (c), respectively. The temperature-dependent integrated intensities of total red emission (d), anti-Stokes radiation (e), and Stokes transitions (f) of $A_2BAlF_6:Mn^{4+}$ (A = K, Rb, Cs; B = Li, Na, K, Rb).

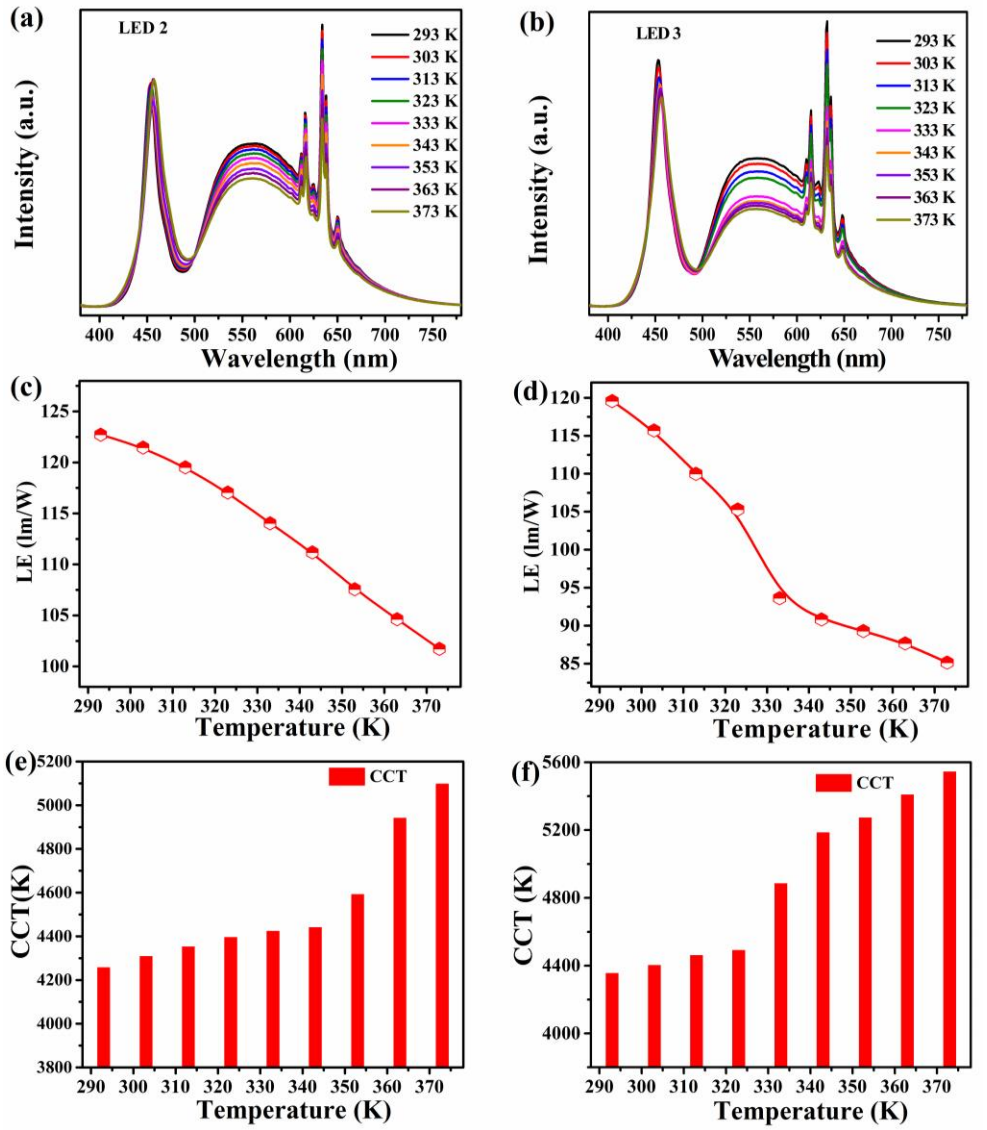

Fig. S7 Temperature-dependent photoelectric properties of warm LED 2 and LED 3 with temperature varying from 293 K to 373 K: EL spectra (a), LE (c) and CCT (e) for LED 2; EL spectra (b), LE (d) and CCT (f) for LED 3.

Table S1 Synthetic strategy of A_2BAlF_6 (A = Rb, Cs; B = K, Rb).

Phosphor	n $(Al(OH)_3)$/mmol	n(RF) /mmol	n(KF) /mmol	n(CsF) /mmol	V_{HF} /mL
$Rb_2KAlF_6:Mn^{4+}$	5	17.5	7.5	---	5.5
$Cs_2KAlF_6:Mn^{4+}$	5	---	5	17.5	5
$Cs_2RbAlF_6:Mn^{4+}$	5	9.5	---	17.5	5

Table S2 Tolerance factors, Al-F bond length and Mn^{4+} emission peak of $A_2BAlF_6:Mn^{4+}$ (A = K, Rb, Cs; B = Li, Na, K, Rb), respectively.

Compounds	r_A(CN= 12) /Å	r_B(CN = 6) /Å	r_{Al}(CN= 6)/Å	t	Bond length/Å (Al-F)	Mn^{4+} emission
$K_2LiAlF_6:Mn^{4+}$	1.64	0.76	0.54	1.06	1.812	634
$K_2NaAlF_6:Mn^{4+}$	1.64	1.02	0.54	1.00	1.806	631
$Rb_2KAlF_6:Mn^{4+}$	1.72	1.38	0.54	0.94	1.786	628
$Cs_2KAlF_6:Mn^{4+}$	1.88	1.38	0.54	0.99	1.806	632
$Cs_2RbAlF_6:Mn^{4+}$	1.88	1.52	0.54	0.96	1.792	630

Table S3 Photoelectric parameters of three warm WLED 1 to WLED 3 fabricated by using $Rb_2KAlF_6:Mn^{4+}$, $Cs_2KAlF_6:Mn^{4+}$, and $Cs_2RbAlF_6:Mn^{4+}$ as red light component, accordingly.

LED device	Chromaticity coordinates (*x*, *y*)	CCT (K)	Ra	LE (lm/W)
LED 1	(0.3728, 0.3770)	4217	80.1	80.6
LED 2	(0.3736, 0.3843)	4238	80.4	122.1
LED 3	(0.3687, 0.3658)	4263	85.6	121.8