

Supporting Information for

$\text{K}_2\text{SiF}_6:\text{Mn}^{4+}$ red-luminescent crystals with high external quantum efficiency (EQE_{max} of 78%) and high thermal quenching temperature ($T_{1/2} > 500$ K) enabling high brightness warm white LEDs

Wenrui Zhang,^a Yayun Zhou,^{*b} Ce Shi,^{*c} Jiajun Ren,^a Liying Zhang,^a Haipeng Ji^{*a}

a School of Materials Science and Engineering, Zhengzhou University, Zhengzhou 450001, China

b Guangdong-Hong Kong-Macao Joint Laboratory for Intelligent Micro-Nano Optoelectronic Technology, School of Physics and Optoelectronic Engineering, Foshan University, Foshan 528225, China

c Henan SoGoodTech Precision Manufacturing Co., Ltd., China

*Corresponding authors.

QE measurement and calculation. Excitation, reflection, and emission spectra for measuring the quantum efficiencies are shown in Figure S2. The internal quantum efficiency, absorption efficiency, and external quantum efficiency were calculated by the following equations:

$$IQE = \frac{\int \lambda P(\lambda) d\lambda}{\int \lambda [E(\lambda) - R(\lambda)] d\lambda} \#(1)$$

$$AE = \frac{\int \lambda [E(\lambda) - R(\lambda)] d\lambda}{\int \lambda E(\lambda) d\lambda} \#(2)$$

$$EQE = IQE \times AE = \frac{\int \lambda P(\lambda) d\lambda}{\int \lambda E(\lambda) d\lambda} \#(3)$$

where $E(\lambda)/h\nu$, $R(\lambda)/h\nu$, and $P(\lambda)/h\nu$ are the number of photons in the excitation, reflection, and emission spectra of the luminescent materials, respectively.

Table S1 Amount of each chemical used for $K_2Si_{1-x}F_6:xMn^{4+}$ crystal growth

x value	0.03	0.07	0.10	0.15	0.20	0.30	0.50	0.60	0.70
H_2SiF_6 (g)					1.7531				
KF (g)					0.4242				
K_2MnF_6 (g)	0.027	0.067	0.100	0.159	0.225	0.386	0.902	1.353	2.105
HF (mL)	9	9	2	2	6	6	1	1	3
	50	54	56	58	60	64	80	96	120

Preparation of the micro-sized powdery $K_2SiF_6:Mn^{4+}$ phosphor.

The powdery phosphor, with the nominal composition of $K_2Si_{1-x}F_6:xMn^{4+}$ ($x = 0.15$), was prepared by a co-precipitation process. The procedure was as follows. Raw

chemicals, KF, H_2SiF_6 solution, and K_2MnF_6 were weighed in accordance with the desired composition. Then, K_2MnF_6 was firstly dissolved in HF solution by magnetic stirring forming a transparent yellow solution. Next, KF was added under continuous stirring. After that, the H_2SiF_6 solution was added drop by drop to the mixture, resulting in the formation of yellow precipitates within the solution. The mixture was then subjected to centrifugation and washed with ethanol three times, and finally, the precipitate was dried at 70 °C for 4 hours to yield the powdery phosphor.

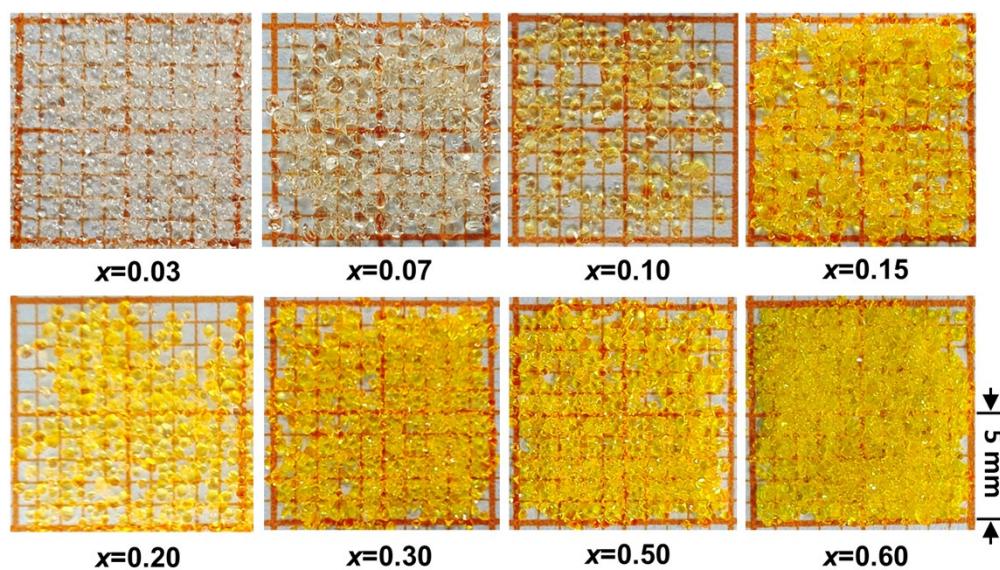


Figure S1 Digital photographs of the as-grown crystals with a nominal composition of $\text{K}_2\text{Si}_{1-x}\text{F}_6:x\text{Mn}^{4+}$, $x = 0.30-0.70$.

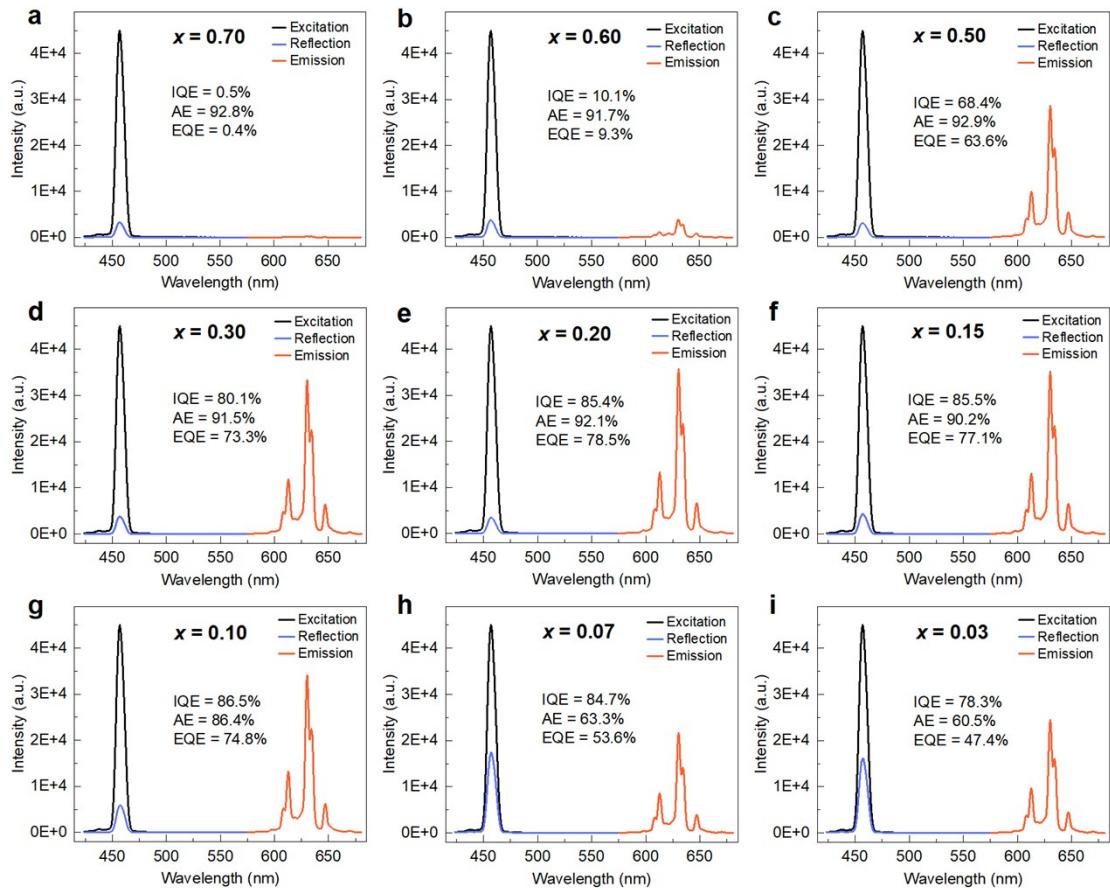


Figure S2 Quantum efficiency measurement for the $\text{K}_2\text{Si}_{1-x}\text{F}_6:\text{xMn}^{4+}$ crystals with nominal x values of 0.03, 0.07, 0.10, 0.15, 0.20, 0.30, 0.50, 0.60, and 0.70.

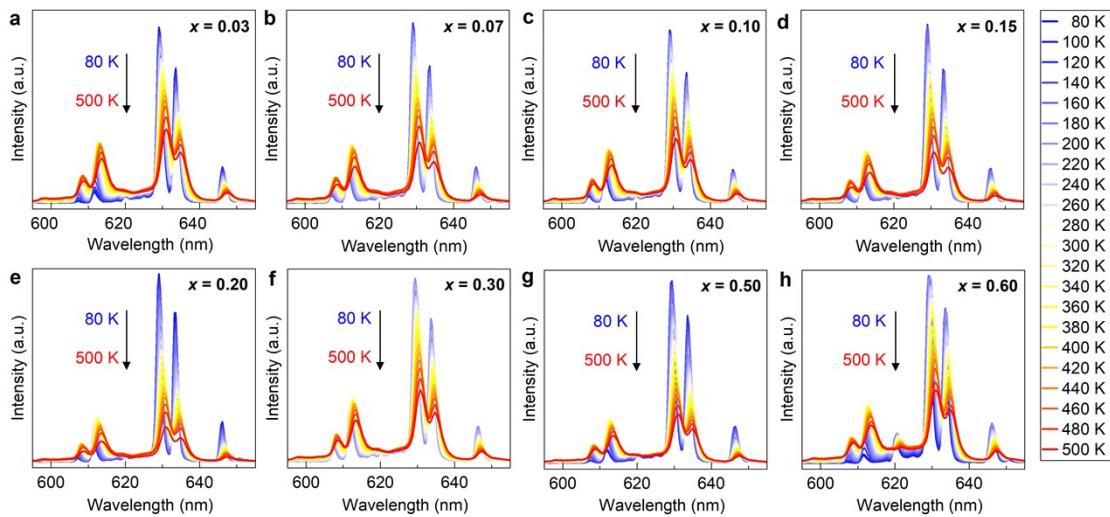


Figure S3 Temperature-dependent photoluminescence emission spectra ($\lambda_{\text{ex}} = 450 \text{ nm}$) of $\text{K}_2\text{Si}_{1-x}\text{F}_6:\text{Mn}^{4+}$ crystals with nominal x values of 0.03, 0.07, 0.10, 0.15, 0.20, 0.30, 0.50, and 0.60, measured at different temperatures (from 80 K to 500 K).

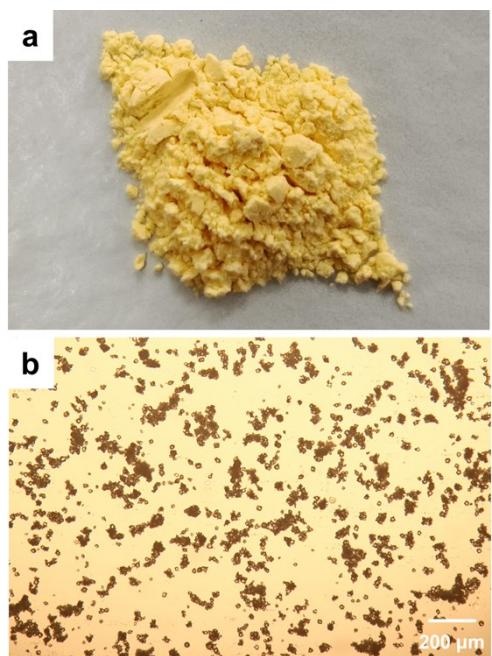


Figure S4 Optical image of the as-prepared $\text{K}_2\text{SiF}_6:\text{Mn}^{4+}$ powdery phosphor via a co-precipitation process.