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Electronic supplementary Information

Elimination of Surface Defects in Luminescent Crystals through Solid-Liquid Interface Friction

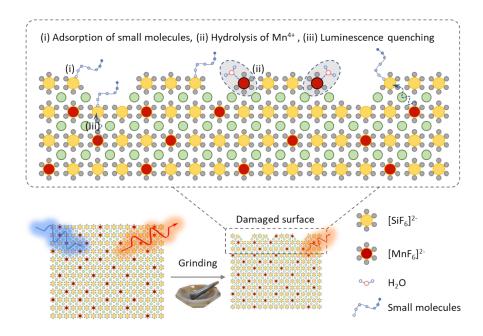
**Materials:** This study employed high-purity reagents, specifically ethanol (99.5%), hydrogen peroxide (30 wt%), potassium permanganate (99.9%) sourced from Sinopharm Chemical Reagent Co., alongside potassium fluosilicate (99%), potassium fluotitanate (99%), hydrofluoric acid (49 wt%), and potassium hydrofluoride (99%) acquired from Shanghai Aladdin Biochemical Technology Co.

Synthesis of K<sub>2</sub>MnF<sub>6</sub>: The synthesis entailed weighing precisely 45.00 g of KHF<sub>2</sub> and 2.25 g of KMnO<sub>4</sub> into a 250 mL plastic beaker, followed by the addition of 150 mL of HF via a measuring cylinder. The mixture was subjected to stirring under an ice-water bath for 60 minutes. Subsequently, hydrogen peroxide was carefully introduced dropwise until the solution's hue shifted from purple to light brown. Centrifugation the upper layer, and the residual yellow powder underwent repetitive washing with acetone and subsequent drying at 70°C for 120 minutes, yielding K<sub>2</sub>MnF<sub>6</sub> powder.

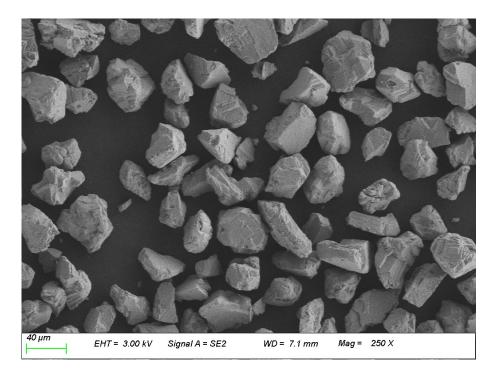
Synthesis of K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup>: The procedure initiated with dissolving 60 g of SiO<sub>2</sub> in 1400 mL of hydrofluoric acid under stirring for 40 minutes at ambient temperature to form a potassium fluosilicate solution. Separately, 125 g of potassium fluorohydride dissolved in 300 mL of hydrofluoric acid constituted the KHF<sub>2</sub> solution. A volume of 350 mL of the former solution combined with 6.90 g of K<sub>2</sub>MnF<sub>6</sub> in a 1000 mL beaker formed the mother liquor upon complete dissolution. Employing a peristaltic pump, 30 mL of the KHF<sub>2</sub> solution was gradually added, post which stirring ceased, and the precipitate was allowed to settle for 3 minutes before filtration. The solids underwent washing with tartaric acid solution, subsequent ethanol rinses, and final drying at 60°C for 12 hours, producing KSFM powder.

Grinding, Sorting, and Solvothermal Treatment of K<sub>2</sub>SiF<sub>6</sub>:Mn<sup>4+</sup>: Large KSFM particles exceeding 100 μm were sieved. Manual grinding for 5 minutes preceded sonication in ethanol for 15 minutes. The suspension was sieved, rinsed twice more with ethanol via centrifugation, and dried at 60°C for 12 hours to isolate particles of varying sizes. For solvothermal treatment, 1 g of the milled KSFM was introduced into a 50 mL reactor vessel along with 30 mL of ethanol, stirred, sealed, and placed in an oven set to predetermined conditions for temperature and duration.

Sample Characterization: Crystallographic analysis was conducted using a Bruker D8 ADVANCE X-ray powder diffractometer (XRD). Surface elemental composition was determined through EscaLab Xi+ X-ray photoelectron spectroscopy (XPS). Fluorescence properties, including excitation and emission spectra, were examined by an F-4500 fluorescence spectrophotometer, while luminescence decay dynamics and quantum yields were characterized by an FLS 1000 spectrometer. Morphological features and dimensions were assessed utilizing a Zeiss Sigma 300 microscope.



**Figure S1**. Schematic illustration of luminescence quenching through unsaturated coordination units on the damaged surface of fluoride crystal.



**Figure S2.** SEM image of the 40-KSFM-R sample.

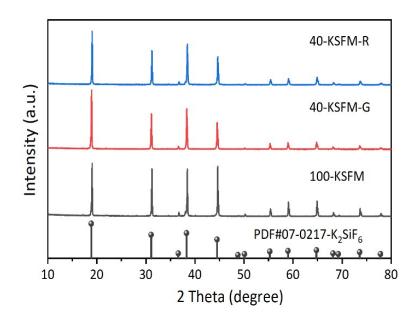
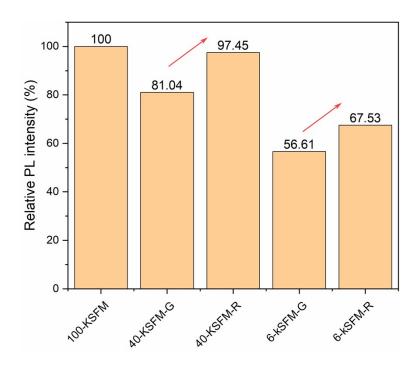


Figure S3. XRD patterns of the KSFM samples.



**Figure S4.** Relative PL intensity of the 40-KSFM and 6-KSFM samples.

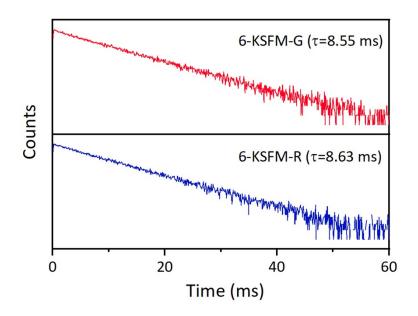


Figure S5. Decay curves of the 6-KSFM-G and 6-KSFM-R samples.

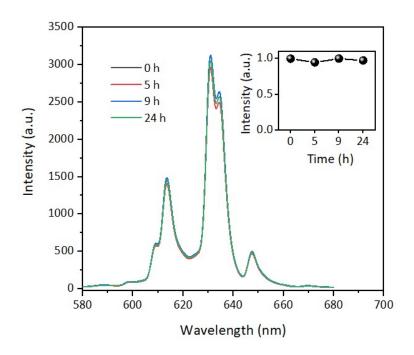
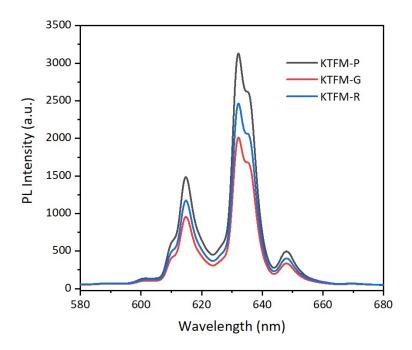


Figure S6. Emission spectra of 40  $\mu m$  unmilled KSFM samples treated with alcohol thermal method for different times.



**Figure S7.** Emission spectra of pristine KTFM (KTFM-P), milled KTFM (KTFM-G) and repaired KTFM (KTFM-R) samples.

**Table S1.** Molecule structure and boiling point of the selected solvents.

Name	Molecular	Boiling Point
Ethanol	ОН	78.3°C
Ethylene glycol	НО	197.3°C
Acetic acid	ОН	117.9°C
Propanoic acid	ОН	141.0°C
Salicylaldehyde	ОН	197.0°C
Ethylenediamine	$H_2N$ $NH_2$	117.3°C