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Dopant valence states control and crystal field influence in Mn-doped luminescent materials

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In this work we propose the study of Mn-doped materials as efficient alternatives to rare earth doped inorganic phosphors in solid-state lighting applications. Manganese ions can be stabilized in two oxidation states, namely Mn^{4+} and Mn^{2+}, both presenting different luminescence properties. In particular, when they are inserted in $SrMgAl_{10}O_{17}$ (SMA) or $BaMgAl_{10}O_{17}$ (BMA) host, and after excitation in the blue region, Mn^{4+} ions present a red luminescence ($^2E \rightarrow {}^4A_2$ transition), whereas Mn^{2+} ions exhibit a green luminescence ($^4T_1 \rightarrow {}^6A_1$ transition, figure 1a). In that context, we developed a new strategy based on the tuning of the valence states of dopant [1], which allows to modulate the Mn^{2+}/Mn^{4+} ratio and thus the luminescence properties of the phosphor. After solid state synthesis in oxidative atmosphere, a reductive thermal treatment is performed on powder to reduce Mn^{4+} ions in Mn^{2+}. For the second step, a thermal gradient oven is used. Multiple samples, which are spatially arranged, are post-treated along the temperature gradient in a single experiment. Thanks to this high throughput method, we demonstrate that the Mn^{4+}/Mn^{2+} ratio can be accurately controlled according to the temperature of the reductive treatment. Hence, the resulting photoemission properties can be optimized as shown on figure 1b. Through this strategy, the CIE coordinates of the phosphor can be tuned. The luminescence thermal sensitivity between Mn^{2+} and Mn^{4+} in isostructural SMA and BMA phosphors can also be explored. The influence of the crystal field on luminescence properties can then be highlighted. This promising approach based on the oxidation state control of manganese activators can be considered for various applications such as white LEDs or luminescent thermal sensor.

[1] Yuan et al., J. of Solid State Chemistry, 288 (2020)

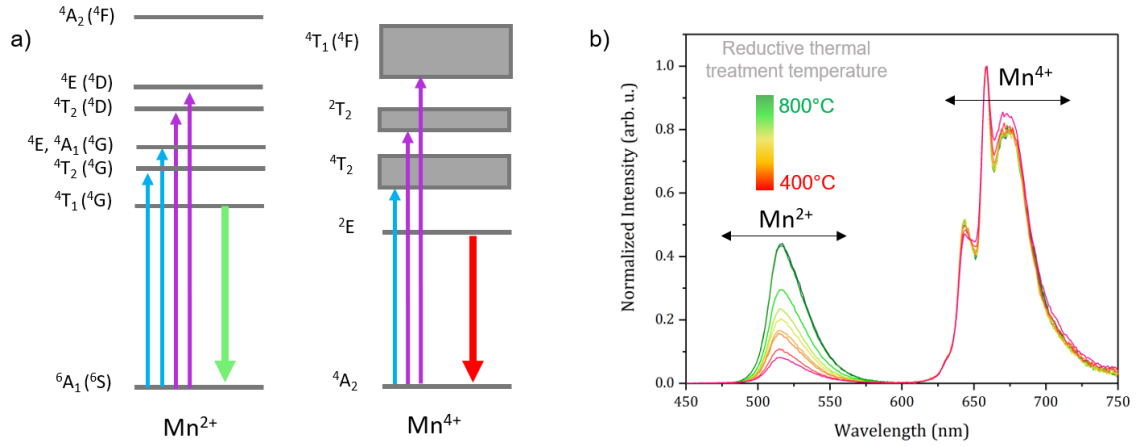

Figure 1 : a) Simplified scheme of energetic levels of Mn^{2+} and Mn^{4+} ions b) Selected photoluminescence spectra of SMA:Mn^{4+},Mn^{2+} after reductive thermal treatment at different temperatures