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Utilizing low-melting-point oxides for high-temperature solid state reaction to enhance the upconversion luminescence intensity

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Abstract

In the traditional solid-state sintering method, the addition of low-melting-point (LMP) oxides (MoO_3 , Sb_2O_3 , TeO_2) promotes tantalate crystal growth and significantly enhances the upconversion luminescence (UCL) intensity. Compared with the samples (S23 sample) without the addition of LMP oxides, the green UCL integral strength of S19 sample with Sb_2O_3 has increased by 20 times. The experiment proved that LMP oxides are prone to erode the corundum crucible at high temperatures, causing the aluminum element to enter the samples. Therefore, a series of samples by changing $\text{Er}^{3+}/\text{Yb}^{3+}$ doping concentrations and the holding time were synthesized using platinum crucibles. The experiments showed that the $\text{Sb-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ sample (S26 sample) with the optimal $0.01\text{Er}^{3+}/0.06\text{Yb}^{3+}$ concentration had the optimal UCL intensity. The main phase of S26 sample is $\text{L-Ta}_2\text{O}_5$, along with some $(\text{Er}/\text{Yb})\text{Ta}_7\text{O}_{19}$ phases and $(\text{Er}/\text{Yb})\text{TaO}_4$ phases. The morphology of this sample is characterized by a porous structure, which is caused by the volatilization of cations in LMP oxides. The morphological results can indicate that the aforementioned three phases have a symbiotic characteristic. The application implementation examples demonstrate that the $\text{Sb-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ phosphor can be applied in the fields of temperature sensing and UCL display.

Keywords: Tantalate, Upconversion luminescence, Sensing, Solid state reaction, Low-melting-point oxides

1. Introduction

Upconversion luminescence (UCL) materials, which convert low-energy near-infrared photons into high-energy visible photons through a multi-step excitation process, have attracted extensive attention owing to their unique anti-Stokes luminescence properties and promising applications in optical temperature sensing [1,2], biological imaging [3,4], anti-counterfeiting labeling [5,6], and solar energy conversion [7,8], *etc.* Among various host matrices, tantalate-based compounds stand out due to their low phonon energy [9,10], excellent chemical and thermal stability [11,12], and tunable crystal structures [13], which are beneficial for achieving efficient UCL emission. However, the practical application of tantalate UCL phosphors is severely limited by their low UCL intensity, which mainly originates from the imperfect crystal quality, uneven distribution of $\text{Er}^{3+}/\text{Yb}^{3+}$ and

abundant surface defects generated during the high-temperature solid state reaction (SSR) synthesis process. Therefore, it is a challenge to further enhance the UCL intensity of tantalate compounds by changing the preparation methods.

Flux-assisted solid-state reaction has been widely adopted to promote crystal growth and improve crystallinity, and the flux is used as Na_2CO_3 [14], H_3BO_3 [15,16], and Li_2CO_3 [17], *etc.* Some aluminates luminescence powders with high firing temperatures often use flux to enhance the luminescence intensity [18,19]. Our previous research has shown that by using SSR and adding Na_2CO_3 , and conducting repeated sintering, the red UCL intensity of $\text{GdLaO}_3:\text{Er}^{3+}/\text{Yb}^{3+}/\text{Sc}^{3+}$ can be enhanced [20]. Component tests were conducted on the sample with the best performance, and it was found that almost all the Na element had evaporated. Therefore, Na_2CO_3 played a role in promoting growth and increasing crystallinity, and did not occupy lattice positions to form inclusions, ultimately enhancing the strength of UCL. However, for tantalates, adding Na_2CO_3 can easily form the NaTaO_3 phase [21,22]. The NaTaO_3 structure is not conducive to achieving high UCL intensity. Therefore, we need to look for other oxides that can form a liquid at high temperatures, are not likely to enter the crystal lattice or only a small amount can enter the crystal lattice, and are easy to volatilize, in order to enhance the UCL brightness.

In this work, we introduce low-melting-point (LMP) oxides MoO_3 (melting point: 795 °C), Sb_2O_3 (melting point: 655 °C) and TeO_2 (melting point: 733 °C) as flux agents, and $\text{Er}^{3+}/\text{Yb}^{3+}$ co-doped tantalate phosphors were prepared by SSR. Compared with the samples without LMP oxides, the UCL intensity of the samples with LMP oxides has been significantly enhanced, which can be increased by up to 20 times. It is demonstrated that the samples with different types of LMP oxides are affected by oxygen in the atmosphere during the SSR process. Corundum crucibles are prone to corrosion by LMP oxides. Therefore, we used platinum crucibles to conduct experiments. In $\text{Sb-Ta-O}:\text{Er}^{3+}/\text{Yb}^{3+}$ system, this optimal UCL intensity (S26 sample) was obtained. The UCL color is pure green. The absolute and relative temperature sensitivities reached 0.01083 K^{-1} and $0.80 \% \text{ K}^{-1}$, respectively. In phosphor-converted 980 nm LEDs fabricated based on the samples, the CIE color coordinate can reach (0.21, 0.75), which exceeds the NTSC standard.

2. Experimental

2.1 Samples synthesis

Powder raw materials including Er_2O_3 (99.99%), Yb_2O_3 (99.99%), MoO_3 (99.99%), TeO_2 (99.99%), Sb_2O_3 (99.0%) and Ta_2O_5 (99.99%) were purchased from Sinopharm Chemical Reagent Co., Ltd., China. All the samples were synthesized via SSR.

For the $\text{Mo-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ samples, corundum crucibles and platinum crucibles were used. The raw materials are weighed according to the molar ratio. The molar ratio of Mo:Ta was 1.1:1, and the molar ratio of Er/Ta and Yb/Ta is 0.01:1 and 0.02:1, respectively. These raw materials were mixed evenly in the mortar and the grinding time was 25 min. The evenly mixed ingredients are placed in an alumina crucible or a platinum crucible with lids, and then put into a muffle furnace. Subsequently, the preparation process of SSR is shown in Fig. 1. The heating temperature ranges from 775 to 1475 °C, with a temperature interval of 100 °C. The temperature is maintained at 775 °C for 12 h, and other temperatures are maintained for 4 h. After the furnace cooling each time, the samples are ground. Then a certain amount of the samples is taken out and numbered. S1 to S8 samples were prepared by using corundum crucibles, while S9 to S16 samples are prepared by using platinum crucibles, as shown in Fig. 1.

After conducting experiments with LMP oxide MoO_3 , we selected a suitable temperature of 1300 °C and a holding time of 4 h to carry out the addition experiments for other LMP oxides such as Sb_2O_3 and TeO_2 . All sample preparations were carried out using platinum crucibles with and without lids. The specific experimental parameters and sample numbers are shown in Table 1.

The above experiments revealed that the $\text{Sb-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ sample exhibited the highest UCL intensity. Therefore, we conducted experiments varying the doping concentration of $\text{Er}^{3+}/\text{Yb}^{3+}$ and the holding time, aiming to further enhance the UCL strength. All sample preparations were carried out using platinum crucibles with lids. The firing temperature is 1300 °C and the holding time is 4 h. The experimental parameters in detail and sample numbers are shown in Table 2.

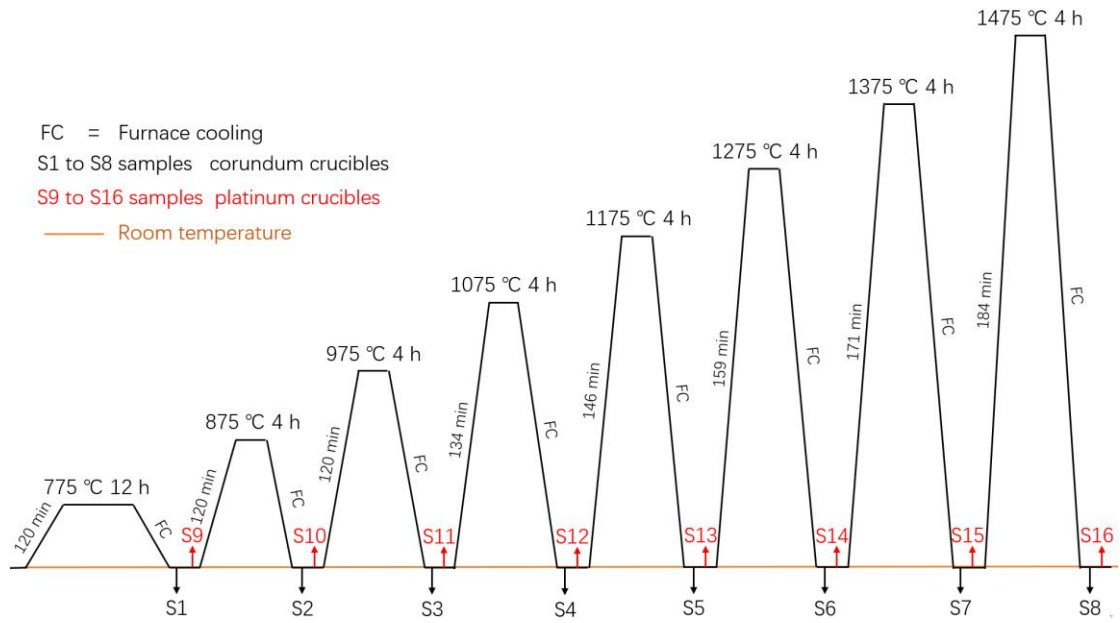


Fig. 1. Mo-Ta-O: Er/Yb samples were prepared using corundum crucibles and platinum crucibles with lids. After being heated at different continuous temperatures, the samples were taken out and numbered. S1 to S8 samples are prepared by using corundum crucibles, while S9 to S16 samples are prepared by using platinum crucibles.

Table 1 Synthesis conditions of Mo-Ta-O:Er³⁺/Yb³⁺, Sb-Ta-O:Er³⁺/Yb³⁺ and Te-Ta-O:Er³⁺/Yb³⁺ obtained from SSR with platinum crucibles with and without lids, with the molar ratio of Mo to Ta being 1.1:1, the molar ratios of Sb to Ta and Te to Ta are both 1:1. The molar ratio of Er/Ta and Yb/Ta is 0.01:1 and 0.02:1, respectively.

Run No.	MoO ₃ /g	Sb ₂ O ₃ /g	TeO ₂ /g	Ta ₂ O ₅ /g	Er ₂ O ₃ /g	Yb ₂ O ₃ /g	Holding temperature/°C	Holding time/h	lid
S17	1.4332			2	0.0173	0.0357	1300	4	√
S18	1.4332			2	0.0173	0.0357	1300	4	×
S19		1.3194		2	0.0173	0.0357	1300	4	√
S20		1.3194		2	0.0173	0.0357	1300	4	×
S21			1.4447	2	0.0173	0.0357	1300	4	√
S22			1.4447	2	0.0173	0.0357	1300	4	×
S23				2	0.0173	0.0357	1300	4	√
S24				2	0.0173	0.0357	1300	4	×

Table 2 Synthesis conditions of Sb-Ta-O:Er³⁺/Yb³⁺ obtained from SSR by using platinum crucibles with lids, with the molar ratio of Sb to Ta being 1:1.

Run No.	Sb ₂ O ₃ /g	Ta ₂ O ₅ /g	Er ₂ O ₃ /g	Yb ₂ O ₃ /g	Molar ratio Er/Ta	Molar ratio Yb/Ta	Holding temperature/°C	Holding time/h
S25	1.3194	2	0.0173	0.0714	0.01	0.04	1300	4
S26	1.3194	2	0.0173	0.1070	0.01	0.06	1300	4
S27	1.3194	2	0.0173	0.1427	0.01	0.08	1300	4
S28	1.3194	2	0.0173	0.1784	0.01	0.10	1300	4
S29	1.3194	2	0.0086	0.1070	0.005	0.06	1300	4
S30	1.3194	2	0.0433	0.1070	0.025	0.06	1300	4
S31	1.3194	2	0.0692	0.1070	0.04	0.06	1300	4
S32	1.3194	2	0.0173	0.1070	0.01	0.06	1300	2
S33	1.3194	2	0.0173	0.1070	0.01	0.06	1300	3
S34	1.3194	2	0.0173	0.1070	0.01	0.06	1300	5

2.2 Characterizations

Powder X-ray diffraction (XRD) patterns were recorded using an X-ray diffractometer (Rigaku D/MAXRE) with Cu K α radiation to analyze the crystal structure. The morphology and component distribution of the samples was examined using scanning electron microscopy (SEM) with a ZEISS Sigma 300 instrument (Germany). UCL spectra at room temperature were acquired by using a Hitachi F-4600 fluorescence spectrophotometer equipped with an external 980 nm laser. Temperature-dependent UCL spectra in the range from 303 to 633 K were systematically measured using a temperature-controlled device (DMU-TC450). The synthesized Sb-Ta-O:Er³⁺/Yb³⁺ powders were thoroughly mixed with silicone rubber and subsequently deposited onto a 980 nm chip to fabricate the LED device. A Wanptek (GPS305D) adjustable regulated DC power supply was used to provide the driving current, and the electroluminescence (EL) spectra of the resulting LEDs were measured by using a Hitachi F-4600 fluorescence spectrophotometer.

3. Results and discussion

Fig. 1(a) presents the powder X-ray diffraction (XRD) patterns of S1 to S8 samples calcined in corundum crucibles with lids at temperatures ranging from 775 to 1475 °C. By comparing with the standard cards, the experimental results show that at 775 °C, $\text{Mo}_2\text{Ta}_2\text{O}_{11}$ (S1) phase forms. Subsequently, as the temperature increases, this phase transition process is: $\text{Mo}_2\text{Ta}_2\text{O}_{11}$ (S1, S2) $\rightarrow$ $\text{MoTa}_{12}\text{O}_{33}$ (S3, S4) $\rightarrow$ $\text{L-Ta}_2\text{O}_5 + \text{MoTa}_{12}\text{O}_{33}$ (S5) $\rightarrow$ $\text{L-Ta}_2\text{O}_5$ (S6) $\rightarrow$ $\text{L-Ta}_2\text{O}_5 + \text{AlTaO}_4$ (S7) $\rightarrow$ $\text{CaTa}_4\text{O}_{11} + \text{L-Ta}_2\text{O}_5$ (S8). This result indicates that as the temperature rises, Mo undergoes severe volatilization. In the end, there was no element of Mo, which is verified by the EDS mapping result of S8 sample in Fig. S1 (Supplementary data). At 1375 °C, the Al element from the corundum crucible entered the sample and formed the AlTaO_4 phase. At 1475 °C, a large amount of the $\text{CaTa}_4\text{O}_{11}$ phase was formed, which might be related to the entry of the Al element. The entry of the Al element into the sample was confirmed through EDS mapping, as shown in Fig. S1. However, the formation mechanism of the $\text{CaTa}_4\text{O}_{11}$ phase remains unclear and will be further investigated in future studies. Fig. 1(b) shows the corresponding colors for different phases in the samples. Fig. 1(c) shows the UCL spectra of S1 to S8 samples under 980 nm laser excitation. All samples show green emissions centered at approximately 523, 532, 543 and 556 nm, originating from the $^2\text{H}_{11/2}/^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$ transitions of Er^{3+} ions. The red emission corresponding to the $^4\text{F}_{9/2} \rightarrow ^4\text{I}_{15/2}$ transition is extremely weak, as clearly demonstrated in the inset image which is enlarged by 50 times in the 500-700 nm range. As Mo evaporates, after $\text{L-Ta}_2\text{O}_5$ forms, the UCL strength significantly increases. At 1475 °C, the $\text{CaTa}_4\text{O}_{11}$ phase structure is formed, and the S8 sample exhibits the highest UCL strength. Fig. 1(d) presents the SEM image of S1 sample calcined at 775 °C. The grains are somewhat aggregated, and the enlarged picture in Fig. 1(d) shows that the grains are in the form of sheets. When the temperature reached 975 °C, Fig. 1(e) indicated that the grains (S3) significantly grew and took on an irregular shape, which was a typical SSR morphology. The temperature was raised to 1475 °C, and the grains (S8) further aggregated. However, no significant increase in the grain size was observed.

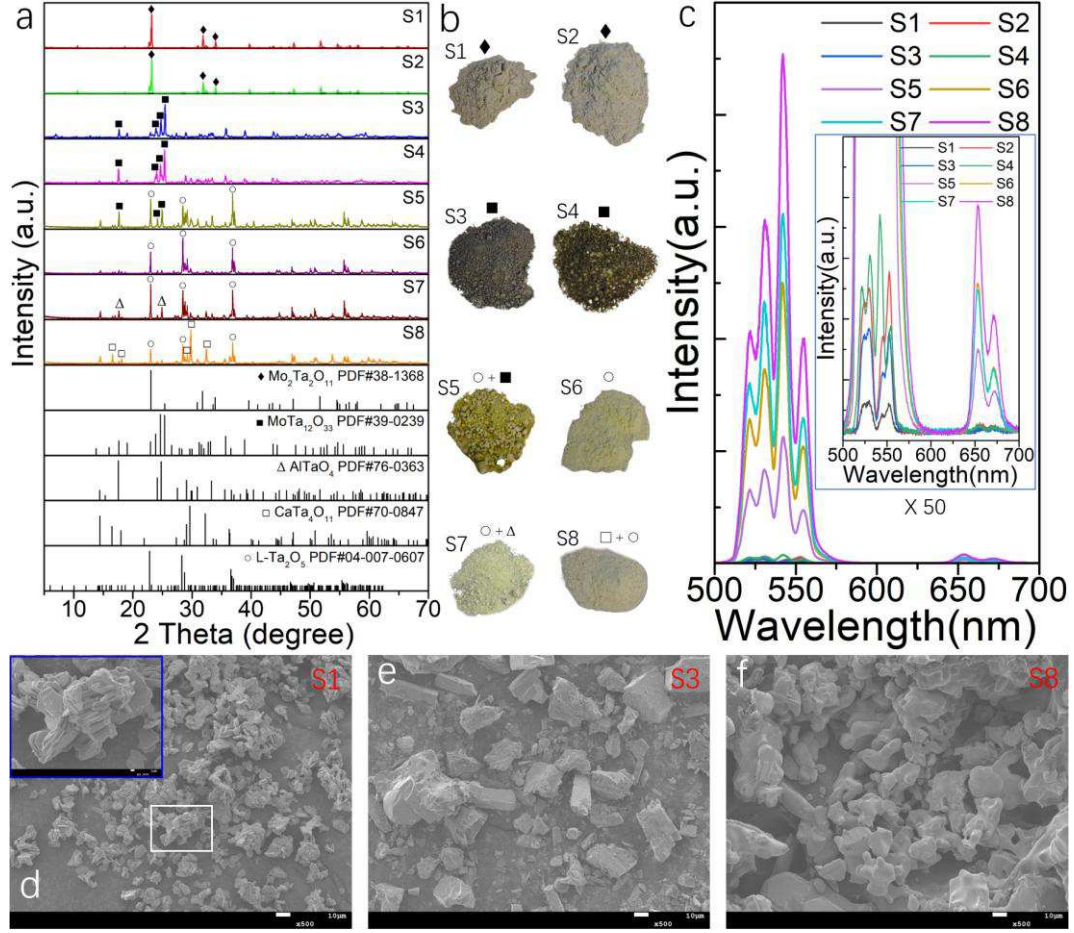


Fig. 1. (a) Powder XRD results of S1 to S8 samples with different temperature calcined in a corundum crucible with lids. (b) Digital photographs of samples S1 to S8. Different crystalline phases are denoted by distinct symbols, with the dominant phase listed first. (c) UCL spectra of S1 to S8 samples under 980 nm laser excitation. The inset image is enlarged by 50 times from 500 to 700 nm. (d)-(f) are SEM images of S1, S3 and S8 samples, respectively. The illustration in (d) is an enlarged version.

Due to the above experiments, the addition of MoO_3 caused the corundum crucible to corrode at high temperatures, and the Al element entered the sample, making the analysis of the phase transformation process extremely complicated. Therefore, we conducted the same experiment using a platinum crucible. Fig. 2(a) shows the powder XRD patterns of S9 to S16 samples calcined in platinum crucibles with lids at different temperatures. From 775 to 1475 °C, this phase transition process is: $\text{Mo}_2\text{Ta}_2\text{O}_{11}$ (S9, S10) $\rightarrow$ $\text{MoTa}_{12}\text{O}_{33}$ (S11, S12, S13) $\rightarrow$ $\text{MoTa}_{12}\text{O}_{33}$ + $\text{L-Ta}_2\text{O}_5$ (S14) $\rightarrow$ $\text{L-Ta}_2\text{O}_5$ (S15) $\rightarrow$ $\text{H-Ta}_2\text{O}_5$ + $\text{L-Ta}_2\text{O}_5$ (S16). The situation of the platinum crucible is different from that of the corundum crucible. No AlTaO_4 and $\text{CaTa}_4\text{O}_{11}$ phases are formed in the platinum crucible, which was because there was no pollution from Al. At 1475 °C, the high-temperature $\text{H-Ta}_2\text{O}_5$ phase was observed to form. Fig. 2(b) is the corresponding colors for different phases. Fig. 2(c) is the UCL spectra of S9 to S16 samples under 980 nm laser excitation. S15 sample has the highest UCL

intensity. S15 sample is L-Ta₂O₅ phase, and S16 sample is H-Ta₂O₅+ L-Ta₂O₅, and it shows L-Ta₂O₅ as host structure is much better than H-Ta₂O₅ structure. Fig. 2(d) to (g) are the SEM results of S9, S11, S15 and S16 samples. The morphology of S9 sample is irregular and differs from that of S1 sample, and the grain size is larger than that of S1 sample. This indicates that the morphologies of samples fired in different crucibles are different. S11, S15 and S16 samples exhibits a long and narrow shape. This minor change in the morphology should be based on the morphology of S11 sample as a template, and it is solely caused by the Mo volatilization.

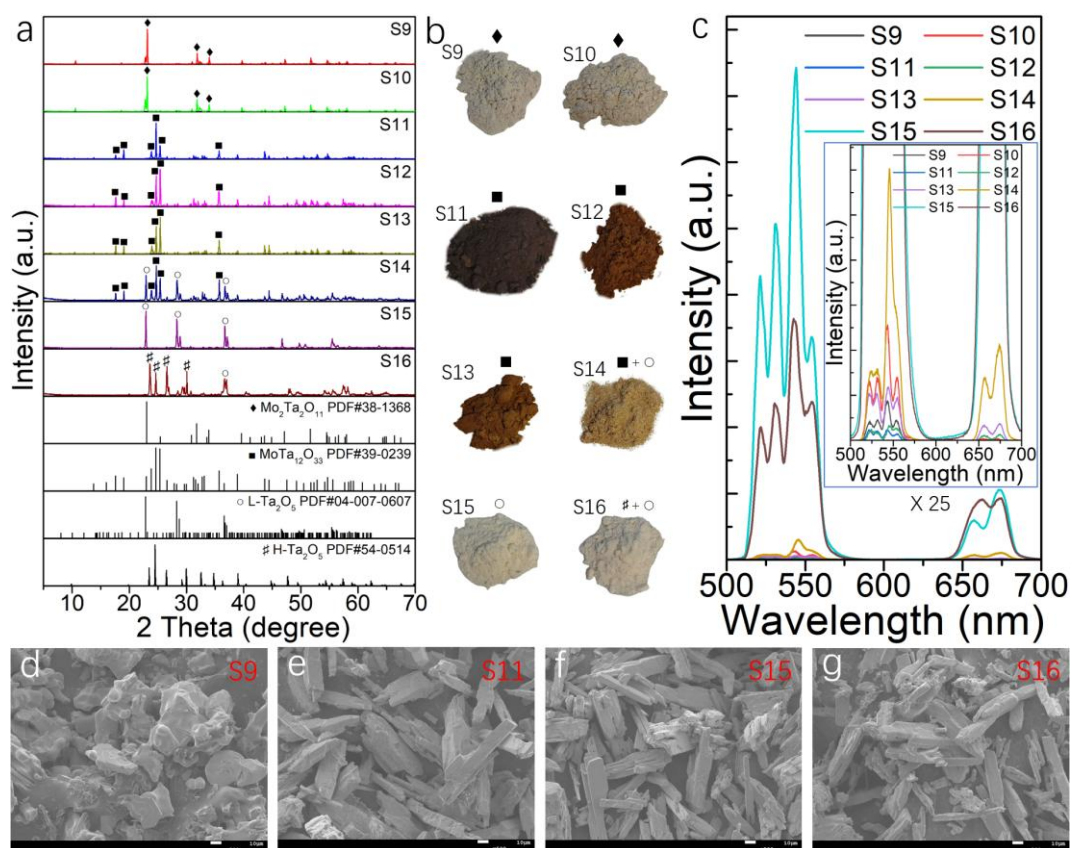


Fig. 2. (a) Powder XRD results of S9 to S16 samples with different temperature calcined in a platinum crucible with lids. (b) Digital photographs of S9 to S16 samples. Different crystalline phases are denoted by distinct symbols, with the dominant phase listed first. (c) UCL spectra of S9 to S16 samples under 980 nm laser excitation. The inset image is enlarged by 25 times from 500 to 700 nm range. (d)-(g) are SEM images of S9, S11, S15 and S16 samples.

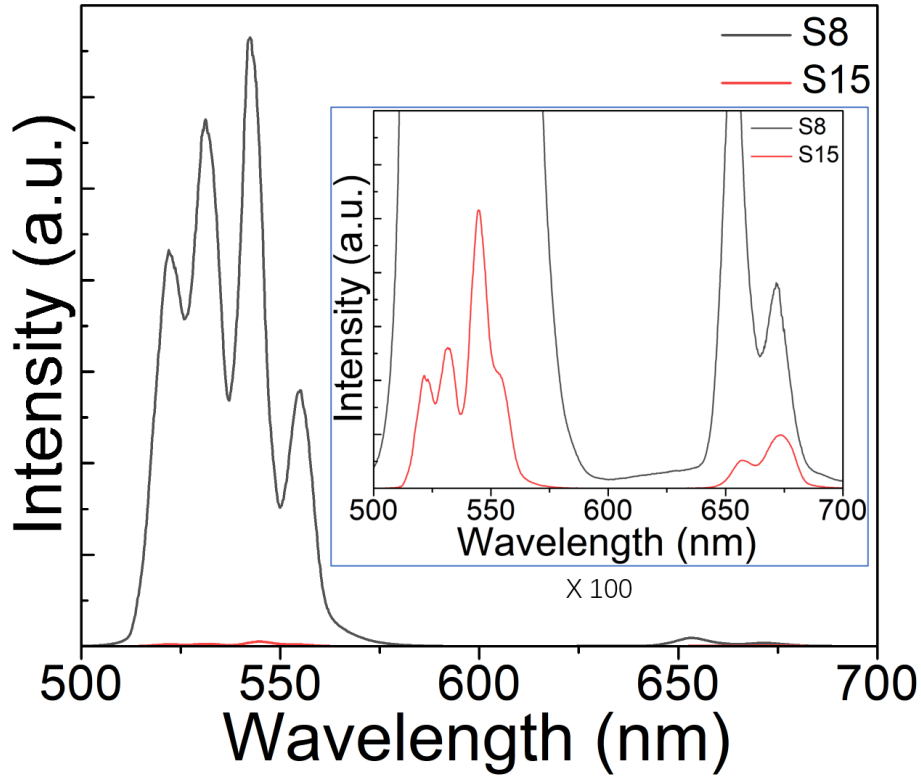


Fig. 3. UCL spectra of S8 and S15 phosphors under 980 nm laser excitation. The inset image is enlarged by 100 times from 500 to 700 nm range.

Fig. 3 compares the UCL intensities of the S8 and S15 samples using corundum crucibles and platinum crucibles, respectively. The result shows the UCL intensity of S8 sample is much higher than that of S15 sample. This might be due to the fact that S8 sample contains a large amount of the $\text{CaTa}_4\text{O}_{11}$ phase structure. This will continue to be studied in the future as described above. However, the sample structure by using corundum crucibles is too complex. Therefore, in the following step, we plan to use LMP oxides such as Sb_2O_3 and TeO_2 for the research on platinum crucibles.

Fig. 4(a) presents the powder X-ray diffraction (XRD) patterns of S17 to S24 samples in platinum crucibles with and without lids. The synthesis of tantalate is greatly influenced by the atmosphere. Therefore, we chose to cover the container and also chose not to cover it. The results show that the main phase of all samples is $\text{L-Ta}_2\text{O}_5$, along with some $(\text{Er/Yb})\text{Ta}_7\text{O}_{19}$ and $(\text{Er/Yb})\text{TaO}_4$ phases. Fig. 4(b)-(e) are UCL spectra of S17 to S24 samples under 980 nm laser excitation. The results show that for $\text{Mo-Ta-O:Er}^{3+}/\text{Yb}^{3+}$, $\text{Sb-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ samples, and the samples (S23, S24) without LMP oxide added, the UCL strength of the samples with lids was higher than that of the samples without lids. However, for $\text{Te-Ta-O:Er}^{3+}/\text{Yb}^{3+}$ sample (S21, S22), the situation is the opposite. Fig. 4(g)-(j) shows SEM images with different magnification levels of S21 and S22 samples. S21 samples is

composed of many small crystals connected in a strip-like form, and the clear boundaries between the crystals can be observed in Fig. 4(h). In the S22 sample, the grain boundaries disappeared and the grains fused together. This should be the reason why the UCL strength of S22 is higher than that of S21. Fig. 4(f) is UCL spectra of S17, S19, S22 and S23 samples under 980 nm laser excitation. Sb-Ta-O:Er³⁺/Yb³⁺ (S19 sample) has the highest UCL intensity. Compared with the samples (S23 sample) without the addition of LMP oxides, the green UCL integral strength of S19 sample with Sb₂O₃ has increased by 20 times. Fig. 5 shows the morphology of S23 and S24 samples. The two samples are in an irregular shape. There is no difference in appearance whether the crucible lid is used or not. However, the spectral comparison results shown in Fig. 4(e) indicate that the UCL intensity of the S23 sample is higher than that of the S24 sample. This suggests that the UCL performance is significantly influenced by oxygen in the atmosphere. This is similar with previous other studies [23]. Next, for Sb-Ta-O:Er³⁺/Yb³⁺ system, the concentrations of Er³⁺/Yb³⁺ and the holding time were changed to further enhance the UCL strength.

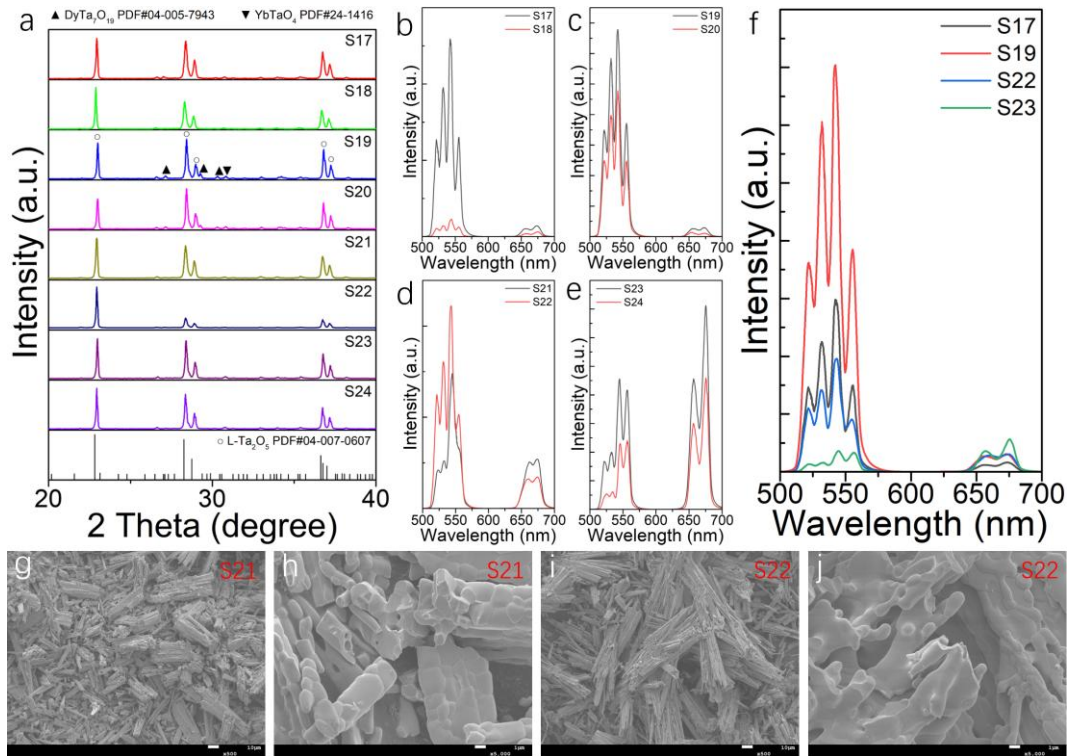


Fig. 4. (a) Powder XRD results of S17 to S24 samples calcined in a platinum crucible with and without lids. (b)-(e) UCL spectra of S17 to S24 samples under 980 nm laser excitation. (f) UCL spectra of S17, S19, S22 and S23 samples under 980 nm laser excitation. (g)-(j) SEM images with different magnification levels of S21 and S22 samples.

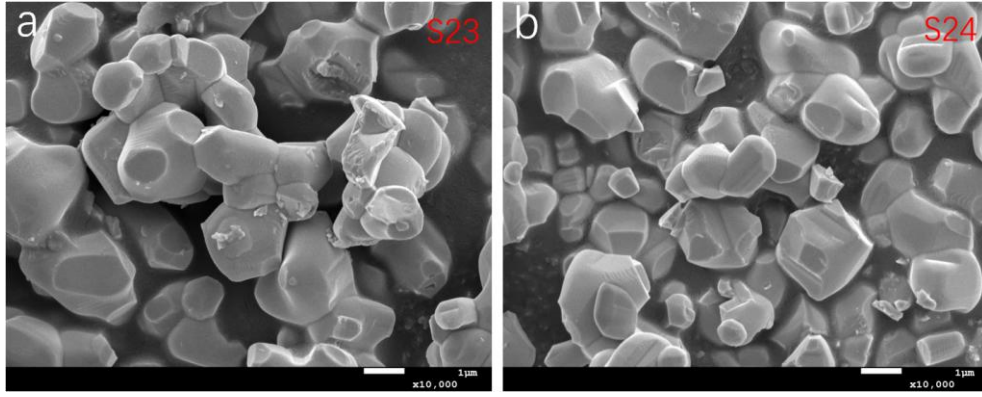


Fig. 5. (a) and (b) SEM images with S23 and S24 samples.

Fig. 6(a) shows the powder X-ray diffraction (XRD) patterns of $\text{Sb-Ta-O:0.01Er}^{3+}/\text{xYb}^{3+}$ ($x = 0.01, 0.04, 0.06, 0.08, 0.10$) samples calcined in platinum crucibles with lids, corresponding to S19, S25, S26, S27 and S28 samples, respectively. Here, the concentration marking of $\text{Er}^{3+}/\text{Yb}^{3+}$ here is the same as the molar ratio of Er/Yb to Ta. This result indicates that the main phase of all samples is $\text{L-Ta}_2\text{O}_5$. As the concentration of Yb^{3+} increases, the $(\text{Er}/\text{Yb})\text{Ta}_7\text{O}_{19}$ phase gradually increases to S26 sample and then decreases. The YbTaO_4 phase has been continuously increasing. Fig. 6(b) is the UCL spectra of S19 and S25 to S28 samples under 980 nm laser excitation. S26 sample has the highest UCL intensity, which should be related to an increase in $(\text{Er}/\text{Yb})\text{Ta}_7\text{O}_{19}$ phase. Our previous research also found that when $\text{L-Ta}_2\text{O}_5$ and $\text{YTa}_7\text{O}_{19}$ coexist, the UCL strength is the highest [24], which is consistent with the results of this experiment.

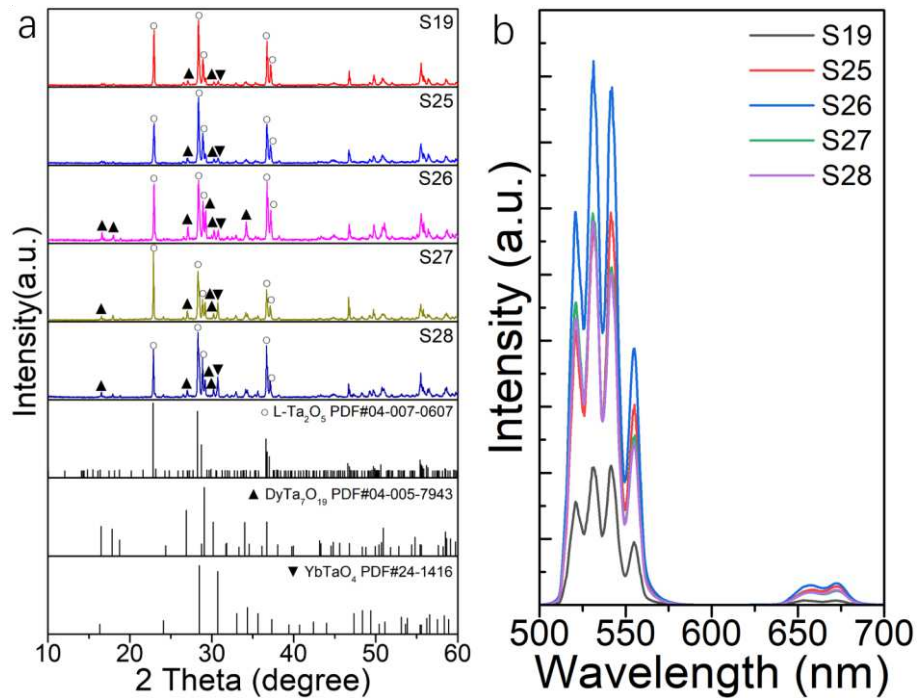


Fig. 6. (a) Powder XRD results of S19 and S25 to S28 samples with different Yb concentrations. (b) UCL spectra of S19 and S25 to S28 samples under 980 nm laser excitation.

Based on the optimal 0.06Yb^{3+} concentration mentioned above, we adjusted the concentration of Er^{3+} . Fig. 7(a) presents the powder X-ray diffraction (XRD) patterns of $\text{Sb-Ta-O:xEr}^{3+}/0.06\text{Yb}^{3+}$ ($x = 0.005, 0.01, 0.025, 0.04$) phosphors calcined in platinum crucibles with lids, corresponding to S29, S26, S30 and S31 samples, respectively. This result indicates that as the concentration of Er^{3+} increases, the amounts of $(\text{Er/Yb})\text{Ta}_7\text{O}_{19}$ and $(\text{Er/Yb})\text{TaO}_4$ phases gradually increase. This corresponding UCL spectra are shown in Fig. 7(b). S26 sample has the highest UCL intensity.

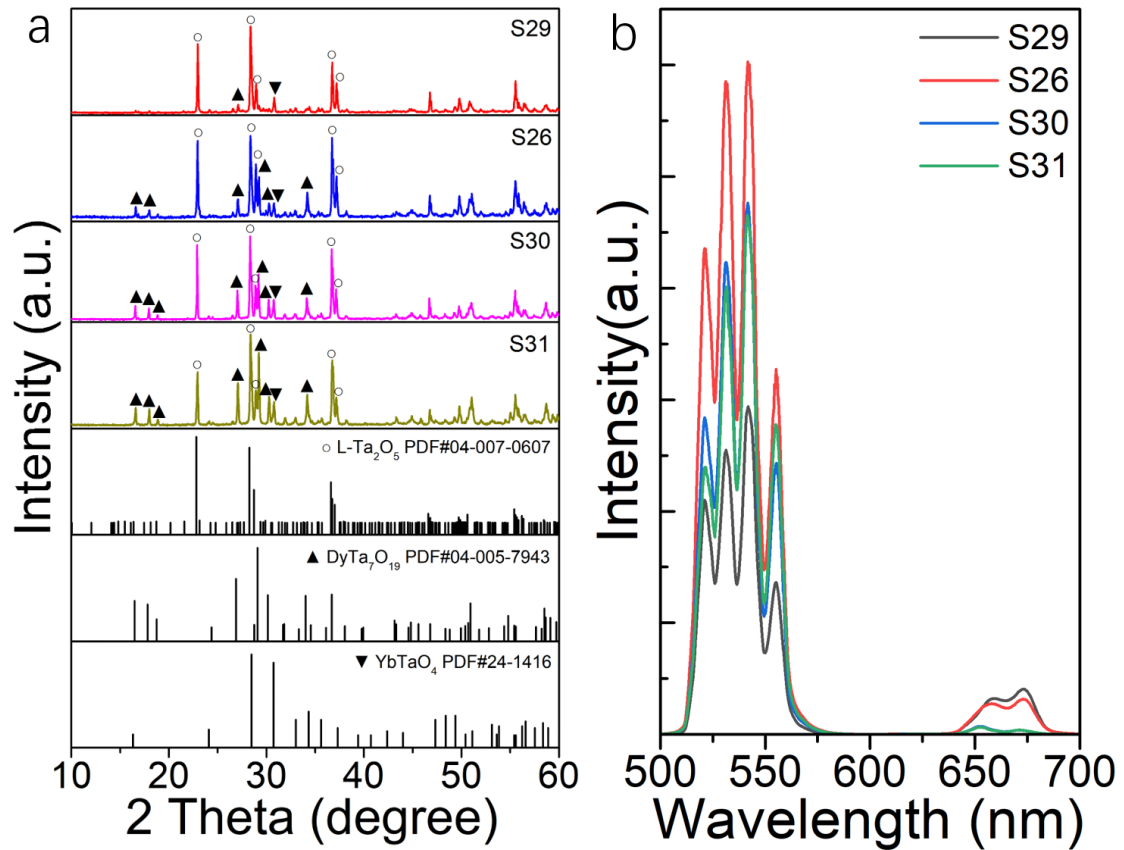


Fig. 7. (a) Powder XRD results of S26 and S29 to S31 samples with different Er^{3+} concentrations. (b) UCL spectra of S26 and S29 to S31 samples under 980 nm laser excitation.

Based on the optimal $0.01\text{Er}^{3+}/0.06\text{Yb}^{3+}$ concentration mentioned above, we adjusted the holding time, hoping to further enhance the UCL strength. Fig. 8(a) presents the powder X-ray diffraction patterns of S26 and S32 to S34 samples at $1300\text{ }^\circ\text{C}$ with different holding times from 2 to 5 h, respectively. This result indicates that as the holding time increases, the $(\text{Er/Yb})\text{TaO}_4$ phase shows a decreasing trend, while the $(\text{Er/Yb})\text{Ta}_7\text{O}_{19}$ phase first increases to the S26 sample and then

decreases. Fig. 8(b) is the corresponding UCL spectra. S26 sample has the highest UCL intensity, which is related to the relatively high proportion of (Er/Yb)Ta₇O₁₉ among all the samples.

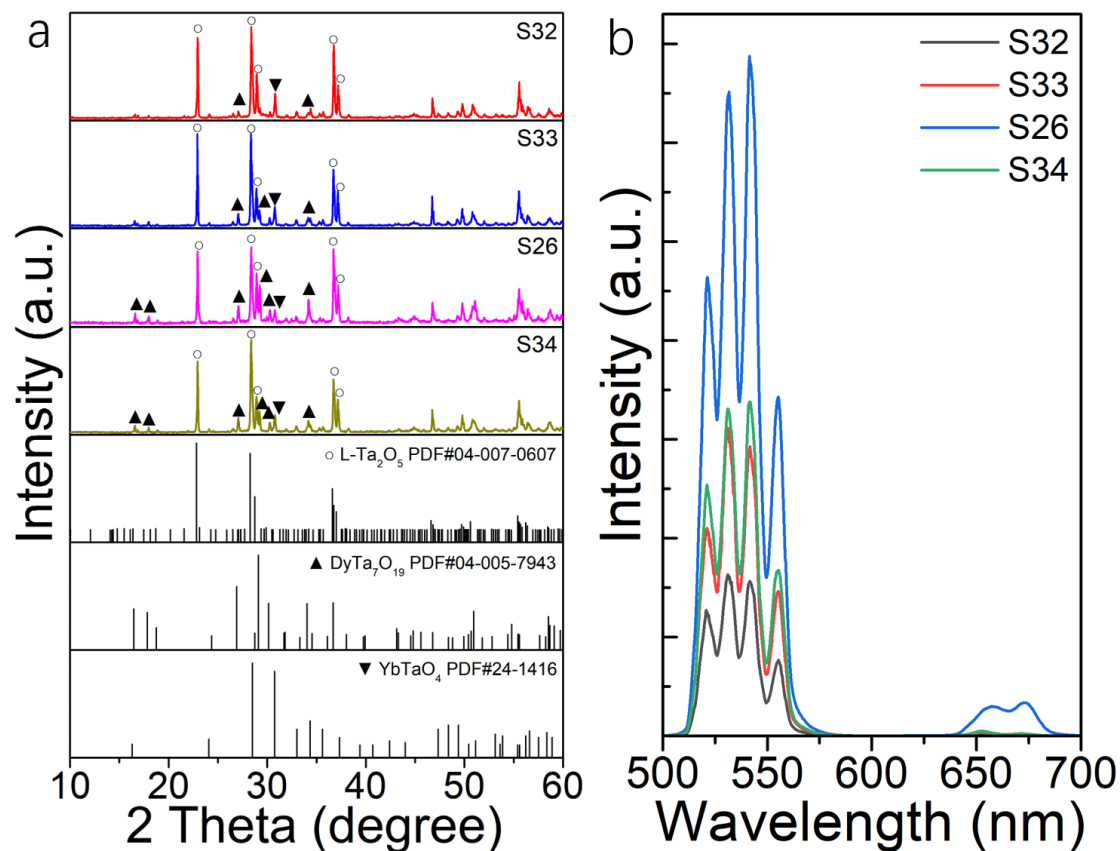


Fig. 8. (a) Powder XRD results of S26 and S32 to S34 samples with different holding times. (b) UCL spectra of S26 and S32 to S34 samples under 980 nm laser excitation.

Fig. 9(a)-(c) are SEM images of S26 sample with different magnification levels. The sample exhibits a porous column-like structure. This void should be caused by the volatilization of the Sb element. In Fig. 9(c), small columnar structures phase grows within the L-Ta₂O₅ host. In Fig. S2, the point composition test was conducted using SEM. The results showed that the small columnar position contained more Sb, Er and Yb elements. From the perspective of composition, it was a different phase. Based on the XRD results, it was speculated to be (Er/Yb)TaO₄ or (Er/Yb)Ta₇O₁₉ phases. These various phases are combined together in a symbiotic manner.

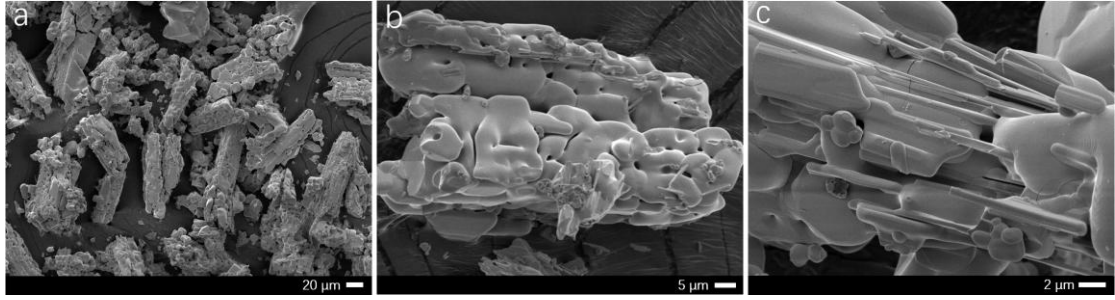


Fig. 9. (a)-(c) SEM images of S26 sample with different magnification levels.

Fig. 10 presents the comparison of UCL spectra between S26 samples from Sb-Ta-O systems and the S8 sample from Mo-Ta-O systems under 980 nm laser excitation. This result indicates that the UCL strength of the S26 sample is higher than that of the S8 sample, reaching 1.8 times.

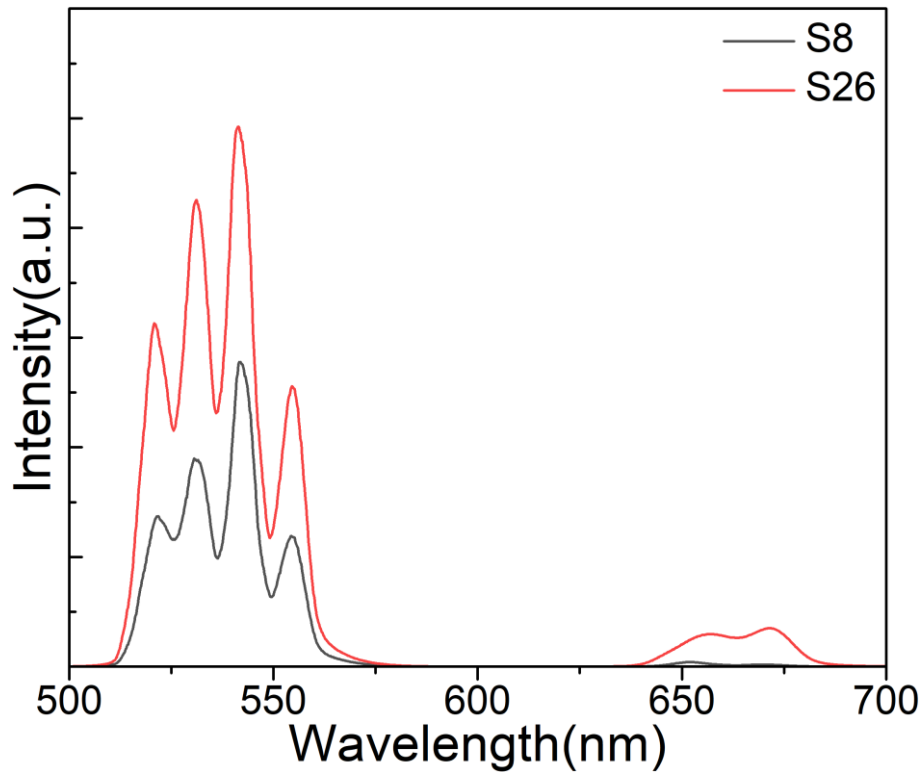


Fig. 10. UCL spectra of S8 and S26 samples phosphors under 980 nm laser excitation.

Fig. 11(a) presents the temperature-dependent green UCL spectra of S26 sample measured in the range of 303-633 K under 980 nm laser excitation. The intensities of all characteristic green emission peaks decrease gradually with increasing temperature, which is ascribed to the enhanced non-radiative relaxation processes caused by thermal activation at elevated temperatures. Since the $^2H_{11/2}$ and $^4S_{3/2}$ energy levels of Er^{3+} ions are thermally coupled, the luminescence intensity ratio (LIR) follows the Boltzmann distribution law, as expressed by the following equation:

$$\text{LIR} = \frac{I_H}{I_S} = B \exp\left(\frac{-\Delta E}{k_B T}\right) \quad (1)$$

Where B is the coefficient, ΔE is the band gap width of the thermal coupling level, k_B represents the Boltzmann constant, and T represents the absolute temperature. Additionally, I_H and I_S denote the UCL integral intensities obtained via Gaussian peak deconvolution from the $^2\text{H}_{11/2}$ and $^4\text{S}_{3/2}$ energy levels, respectively. The variation of LIR with temperature is plotted in Fig. 11(b), and the experimental data are well fitted by the exponential function with a high correlation coefficient $R^2 = 0.9980$. The absolute sensitivity S_A and relative sensitivity S_R can then be calculated using the following equations:

$$S_A = \frac{d\text{LIR}}{dT} = B \frac{\Delta E}{k_B T^2} \exp\left(\frac{-\Delta E}{k_B T}\right) \quad (2)$$

$$S_R = \frac{1}{\text{LIR}} \frac{d\text{LIR}}{dT} = \frac{\Delta E}{k_B T^2} \quad (3)$$

The S_A value increases first and then decreases with rising temperature, reaching a maximum value of 0.01083 K^{-1} at 371 K in Fig. 11(c). The S_R value decreases monotonically with increasing temperature, and the maximum S_R is obtained as $0.80 \% \text{ K}^{-1}$ at 303 K . These results confirm that S26 sample possesses favorable optical temperature sensing properties, making it suitable for high-temperature optical sensing applications.

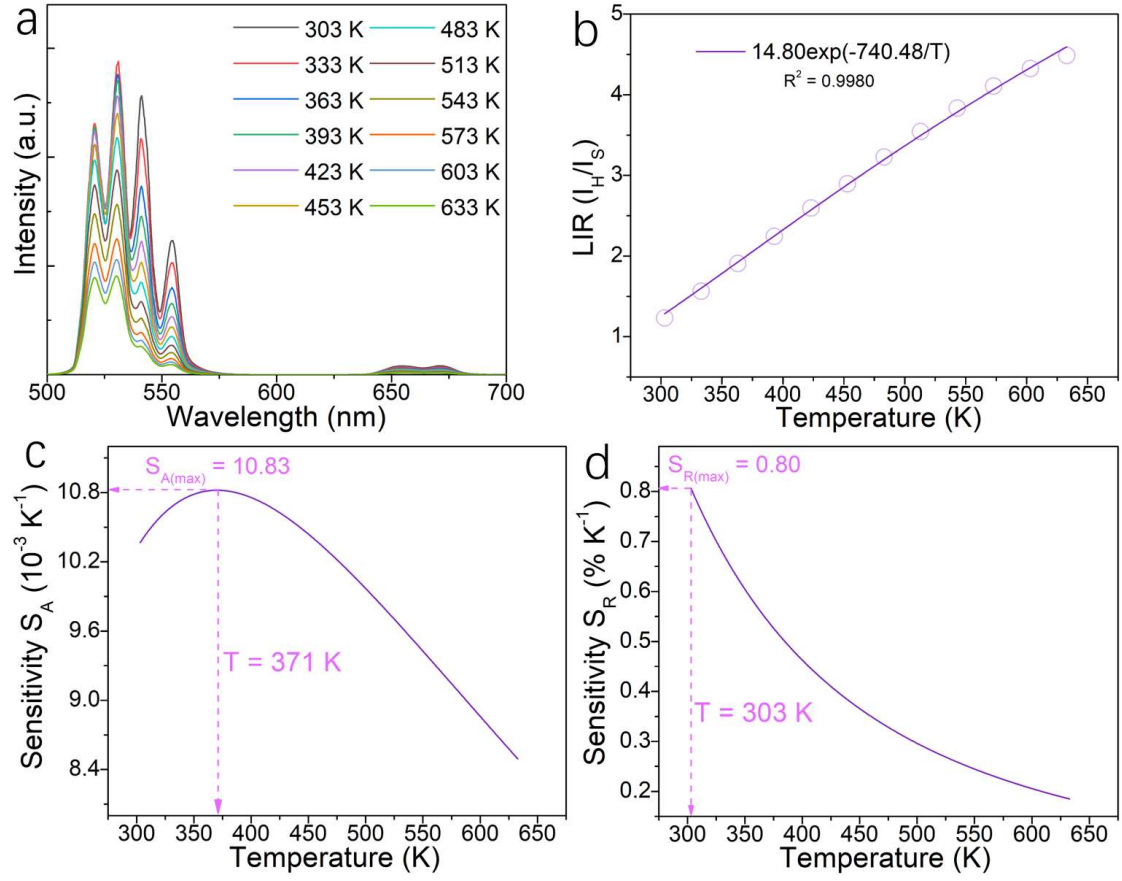


Fig. 11. (a) Green UCL spectra of S26 sample at 303-633 K under 980 nm laser excitation. (b) The relationship between LIR and temperature. The absolute sensitivity S_A curve (c) and the relative sensitivity S_R curve (d).

Table 3 Absolute sensitivity S_A and relativity values S_R of various upconversion materials under 980 nm laser excitation.

Upconversion materials	S_A ($10^{-3} K^{-1}$)	S_R (% K^{-1})	Ref.
$Ca_2ScNbO_6:Er^{3+}/Yb^{3+}$ phosphor	4.4 (298 K)	0.93 (298 K)	[25]
$LuNbO_4:Er^{3+}/Yb^{3+}/Tm^{3+}$ phosphor	69.1 (650 K)	4.0 (300 K)	[26]
$Cs_3Bi_2Br_9:Er^{3+}/Yb^{3+}$ single crystal	0.87 (410 K)	0.879 (303 K)	[27]
$La_2MoO_6:Er^{3+}/Yb^{3+}$ phosphor	7 (453 K)	1.04 (303 K)	[28]
$Ca_2GdNbO_6:Er^{3+}/Yb^{3+}$ phosphor	2.4 (523 K)	1.24 (298 K)	[29]
$BiTa_7O_{19}:Er^{3+}/Yb^{3+}$ single crystal	8.51 (377 K)	0.83 (303 K)	[30]
$Sb-Ta-O:Er^{3+}/Yb^{3+}$ phosphor	10.83 (371 K)	0.80 (303K)	This study

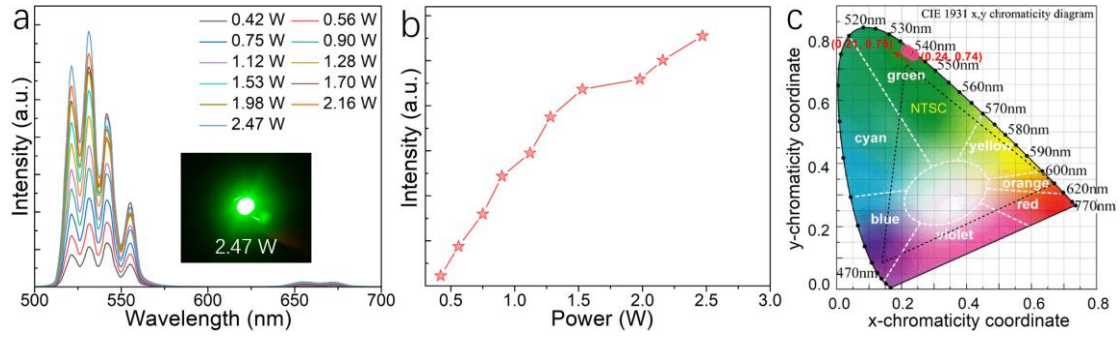


Fig. 12. (a) The electroluminescence spectra of LED chips fabricated with S26 sample. The inset is a digital photograph taken under 2.47 W. (b) The corresponding green UCL integral intensity. (c) CIE color coordinates as compared with the NTSC standard (dashed line).

Fig. 12(a) presents the electroluminescence spectra of LED chips fabricated with S26 sample under various 980 nm LED powers. All samples exhibit intense characteristic pure green emissions, and the emission intensity increases remarkably with rising LED power. The inset in Fig. 12(a) displays the digital photograph under 2.47 W, clearly showing a bright and uniform green UCL. As the power increases from 0.42 to 2.47 W, the green UCL intensity gradually increases, as shown in Fig. 12(b). The Commission Internationale de l'Eclairage (CIE) chromaticity coordinates under different LED powers are calculated and presented in Fig. 12(c). The CIE coordinates change slightly from (0.24, 0.74) to (0.21, 0.75) as the LED power increases, progressing along the edge of the chromaticity diagram toward a greener region and extending beyond the NTSC color gamut. The result demonstrates that the LED device can be regarded as a pure green UCL display.

4. Conclusion

During SSR process, the addition of LMP oxide enhanced the strength of UCL. The experiment revealed that the LMP oxide is prone to eroding the crucible at high temperatures. For Mo-Ta-O:Er³⁺/Yb³⁺, the phase transition process is determined as the temperature increases. Sb₂O₃ and TeO₂ are also utilized for the synthesis of samples. Compared with the sample without the addition of LMP oxides, the green UCL integral strength of S19 sample with Sb₂O₃ has increased by 20 times. For Sb-Ta-O systems, the optimal Er³⁺ and Yb³⁺ concentrations are 0.01 and 0.06, respectively. The main phase of optimal S26 sample is L-Ta₂O₅, along with some (Er/Yb)Ta₇O₁₉ and (Er/Yb)TaO₄ phases. The morphological is porous structure and the aforementioned three phases have a symbiotic characteristic. The S_A and S_R can reach 0.01083 K⁻¹ and 0.80 % K⁻¹, respectively. The CIE

coordinates of LED chips fabricated with S26 sample are beyond the NTSC color gamut under 980 nm LED excitation. All the results indicate that Sb-Ta-O:Er³⁺/Yb³⁺ phosphors can be utilized in the fields of temperature sensing and UCL display.

Conflicts of interest

There are no conflicts of interest to declare

Acknowledgements

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