

Thermometric properties of $\text{Na}_2\text{Y}_2\text{TeB}_2\text{O}_{10}:\text{Tb}^{3+}$ green phosphor based on fluorescence/excitation intensity ratio

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Abstract: For noncontact optical thermometry, in contrast with fluorescence intensity ratio (FIR) technology, excitation intensity ratio (EIR) technology has been seriously limited due to low sensitivity. Moreover, by exploring all possible temperature-dependent response, developing multimode optical thermometry is of great importance. In this work, a new $\text{Na}_2\text{Y}_2\text{TeB}_2\text{O}_{10}$ (NYTB): Tb^{3+} phosphor is obtained by a solid-state reaction. Based on FIR and EIR models of Tb^{3+} , thermometric properties are studied thoroughly. Excellent relative and absolute sensitivity (S_R and S_A) are acquired due to the significant difference in emission/excitation lines in response to temperature. Meanwhile, Tb^{3+} content-dependent luminescence quenching mechanism is discussed. This study shows a feasible route for exploiting well-performing FIR-/EIR-based thermometric materials.

Keywords: $\text{Na}_2\text{Y}_2\text{TeB}_2\text{O}_{10}$ (NYTB); Tb^{3+} ion; photoluminescence (PL); thermometric properties

1 Introduction

Temperature is a vital parameter in industrial and civil areas, which makes developing temperature-detecting technology significant [1–3]. The shortcomings of conventional contact thermometers include environmental restrictions and long response time, which gradually cannot meet the actual usage requirements [4–6]. Hereby, Ghahrizjani *et al.* [7] and Cheng *et al.* [8] have paid much research to noncontact temperature-detecting technologies. Among these technologies, the optical thermometry method based on temperature-dependent

luminescent properties of a phosphor is a fascinating issue owing to the advantages of noncontact model, high resolution, and fast response [8,9].

For optical temperature measurements, diverse strategies have been developed such as fluorescence intensity ratio (FIR), excitation intensity ratio (EIR), fluorescence lifetime, and emission intensity/position/bandwidth [10–14]. The mechanism of the FIR/EIR for optical thermometry focuses on studying the relationship between the FIR/EIR and the temperature [15]. The FIR approach can be achieved by monitoring the difference between the two emission peaks in response to the temperature. For instance, a fluorescence thermometer has been extensively investigated based on thermally-coupled energy levels of Er^{3+} , Pr^{3+} , and Nd^{3+} [16–18]. Recently, an EIR model has been increasingly

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developed [10]. Compared with an FIR model, the EIR model provides more information about exciting levels of luminescence centers that cannot be detected in a emission spectrum [19]. The EIR model provides an efficient method for constructing a new high-performance thermometer.

Among rare-earth ions, the ${}^5D_4 \rightarrow ({}^7F_6, {}^7F_5, {}^7F_4, \text{ and } {}^7F_3)$ emissions of Tb^{3+} ions present opposite temperature-dependent behaviors, which are suitable for FIR thermometry with high relative sensitivity (S_R). Besides, 5D_3 and 5D_4 ground levels of Tb^{3+} can be used to realize the EIR thermometry due to the strong temperature-sensitive difference. This arouses our interest in developing multimode optical thermometry in Tb^{3+} -activated phosphors. Recently, low-lying thermally-coupled 7F_5 and 7F_6 principle of Tb^{3+} was employed to investigate temperature sensing performances of glass-ceramic materials of $Ba_2LaF_7:Tb^{3+}$ [20] and $Na_2GdF_7:Tb^{3+}$ [21], which presented the maximum S_R (S_{R-max}) of $2.88 \% \cdot K^{-1}$ at 300 K and $2.33 \% \cdot K^{-1}$ at 310 K. Drabik *et al.* [22,23] developed a single-band-ratiometric method to utilize opposite thermal dependence of a characteristic emission for Tb^{3+} excited by the two different wavelengths (λ). For example, based on the opposite temperature-dependent behaviors of the ${}^5D_4 \rightarrow {}^7F_3$ emission under the excitations of the ${}^7F_6 \rightarrow {}^5D_4$ and ${}^7F_5 \rightarrow {}^5D_4$ transitions, it is reported that $KLaP_4O_{12}:0.1Tb^{3+}$ exhibited the S_{R-max} value of $5.5 \% \cdot K^{-1}$ [23]. Wei *et al.* [24] researched the thermometric properties of $CaNb_2O_6:Tb^{3+}$ in terms of the descending difference of temperature-dependent emissions from a host and a Tb^{3+} activator. The thermometric strategy associated with the host was also reported in $Sr_3MgCe(PO_4)_7:Tb^{3+}$ [25], $Y_2O_3:Tb^{3+}$ [26], and $Lu_2O_3:Tb^{3+}$ [26] phosphors. Especially, Li *et al.* [27] reported the EIR in $La_2ZnTiO_6:Tb^{3+}$. According to the different thermal dependence of 381 nm (4f–4f of Tb^{3+}), 285 nm (4f–5d of Tb^{3+}), and 313 nm (intervalence charge transfer) excitation bands, the EIR models ($EIR(I_{381}/I_{285})$ and $EIR(I_{381}/I_{313})$) were established to obtain the maximum absolute sensitivity (S_{A-max}) ($4.46 \times 10^{-2} K^{-1}$) and S_R ($0.727 \% \cdot K^{-1}$). To our knowledge, there are no reports for Tb^{3+} -activated phosphors with FIR/EIR-based thermometric properties.

Considering our early work on the yellow $Na_2Y_2TeB_2O_{10}$ (NYTB)-based phosphor activated with a Dy^{3+} ion [28], we prepared a Tb^{3+} -activated NYTB phosphor via a solid-state reaction in this work. Characteristic green-emitting, luminescent quenching

mechanism, and thermometric properties were systematically investigated. Moreover, according to distinct temperature dependence of the characteristic emissions and excitations of Tb^{3+} , both FIR and EIR models were proposed to measure temperature signals.

2 Experimental

In the solid-state preparation route of a Tb^{3+} -incorporated NYTB phosphor, the employed reagents were Na_2CO_3 (analytical reagent (A.R.)), H_3BO_3 (A.R.), Y_2O_3 (99.99%), TeO_2 (99.99%), and Tb_2O_3 (99.99%). Based on the formula of $Na_2Y_{2-x}Tb_xTeO_4(BO_3)_2$ ($x = 0-90$ mol%), they were weighed, mixed, and ground for 30 min. Subsequently, the obtained mixture was placed in an open muffle furnace (M1210, Henan Chengyi Laboratory Equipment Co., Ltd.) and preheated at $450^\circ C$ for 10 h. After preheating, the reactant was ground and sintered at $780^\circ C$ for 84 h with the grinding at 10 h intervals. The heating rate was $2.5^\circ C \cdot min^{-1}$. Finally, a white powder sample was prepared after cooling down.

X-ray diffraction (XRD) patterns were characterized by an X-ray diffractometer (DX-2700BH, Bruker; $Cu K\alpha$ X-ray source with $\lambda = 1.54056 \text{ \AA}$). Fourier transform infrared spectroscopy (FTIR) data were collected by an infrared spectrometer (Nicolet iS10, Thermo Scientific). A transmission electron microscope (TEM; JEM 2100, JEOL) was employed to observe morphological features. An elemental investigation was carried out via an energy-dispersive X-ray spectrometer (EscaLab Xi+, FEI) in a scanning electron microscope (SEM; Nova NanoSEM 450, FEI). X-ray photoelectron spectroscopy (XPS) data were recorded by an X-ray photoelectron spectrometer (K-Alpha+, Thermo Fisher Scientific). Diffuse reflectance data were recorded by an ultraviolet/visible (UV/Vis) spectrophotometer (UV2600, SHIMADZU). Luminescent properties were measured by a fluorescence spectrophotometer (F-7100, HITACHI). Electroluminescence (EL) behaviors of a fabricated white light emitting diode (w-LED) device was characterized by an LED optoelectronic analyzer (HP 9000, Hongpu Photoelectric Technology Co., Ltd.).

3 Results and discussion

3.1 Phase identification

Figure 1(a) displays that the detected XRD profiles of

NYTB: $x\text{Tb}^{3+}$ ($x = 0\text{--}90$ mol%) coincide well with the standard result of the NYTB (JCPDS No. 66-799). It is suggested that the monoclinic $P2_1/c$ structure of the NYTB is unchanged with the incorporation of the Tb^{3+} ions. Additionally, the \blacklozenge symbol denotes a weak impurity peak coming from $\text{Na}_2\text{B}_8\text{O}_{13}$ (JCPDS No. 71-2163) owing to the secondary reaction. In an NYTB lattice, a coordination number (CN) of the Y^{3+} ion is seven (Fig. 1(b)). The activator Tb^{3+} (CN = 7, radius (r) = 0.98 Å) keeps a similar size as Y^{3+} (CN = 7, r = 0.96 Å) [29], because they belong to a lanthanide group. Thus, the activator Tb^{3+} is inclined to substitute Y^{3+} . Moreover, the magnified XRD peak shifts to a smaller diffraction angle with a Tb^{3+} concentration rising (Fig. 1(a)), which is also the proof that Y^{3+} is substituted by Tb^{3+} . In view of this fact, the following luminescence investigation is uninfluenced by a small amount of $\text{Na}_2\text{B}_8\text{O}_{13}$. Figure 1(c) depicts a Rietveld refinement pattern for a representative sample of the NYTB:70% Tb^{3+} . Residual factors of R_{wp} , R_{p} , and χ^2 (R_{wp} , R_{p} , and χ^2 are the refinement results of the weighted profile R -factor, profile residual factor, and goodness-of-fit, respectively) are converged to be 5.02%, 3.58%, and 2.29, respectively, indicating reliability of a

monoclinic structure for the prepared sample. Other refined structural data are summarized in Table S1 in the Electronic Supplementary Material (ESM). To explore the impact of incorporating Tb^{3+} on the NYTB lattice, Table 1 summarizes the comparison of the lattice constants for the NYTB and NYTB:70% Tb^{3+} . Clearly, owing to a slightly larger size of Tb^{3+} than that of Y^{3+} , the lattice constants for the NYTB:0.7 Tb^{3+} are a little greater than those for the NYTB [28]. On the other hand, FTIR measurements further verify the monoclinic structure of Tb^{3+} -doped samples (Fig. S1 in the ESM). Positions and shapes of the observed vibrations coincide well with those of the NYTB, deriving from $\nu_{\text{as}}(\text{B-O})$, $\nu_{\text{s}}(\text{B-O})$, $\nu_{\text{s}}(\text{Te-O})$, $\nu_{\text{b}}(\text{B-O})$, and $\nu(\text{Te-O})$ in a host lattice [30,31] wherein $\nu_{\text{as}}(\text{B-O})$ is defined as the asymmetric stretch of the BO_3 groups, $\nu_{\text{s}}(\text{B-O})$ is defined as the symmetric stretching vibration of the BO_3 groups, $\nu_{\text{s}}(\text{Te-O})$ is defined as the symmetric stretching vibration of the TeO_6 groups, $\nu_{\text{b}}(\text{B-O})$ is defined as the bending vibration of the BO_3 groups, and $\nu(\text{Te-O})$ is defined as the Te-O antistretching and Te-O-Te vibration. The results mean that structural groups of the NYTB are unbroken even by doping the Tb^{3+} ions.

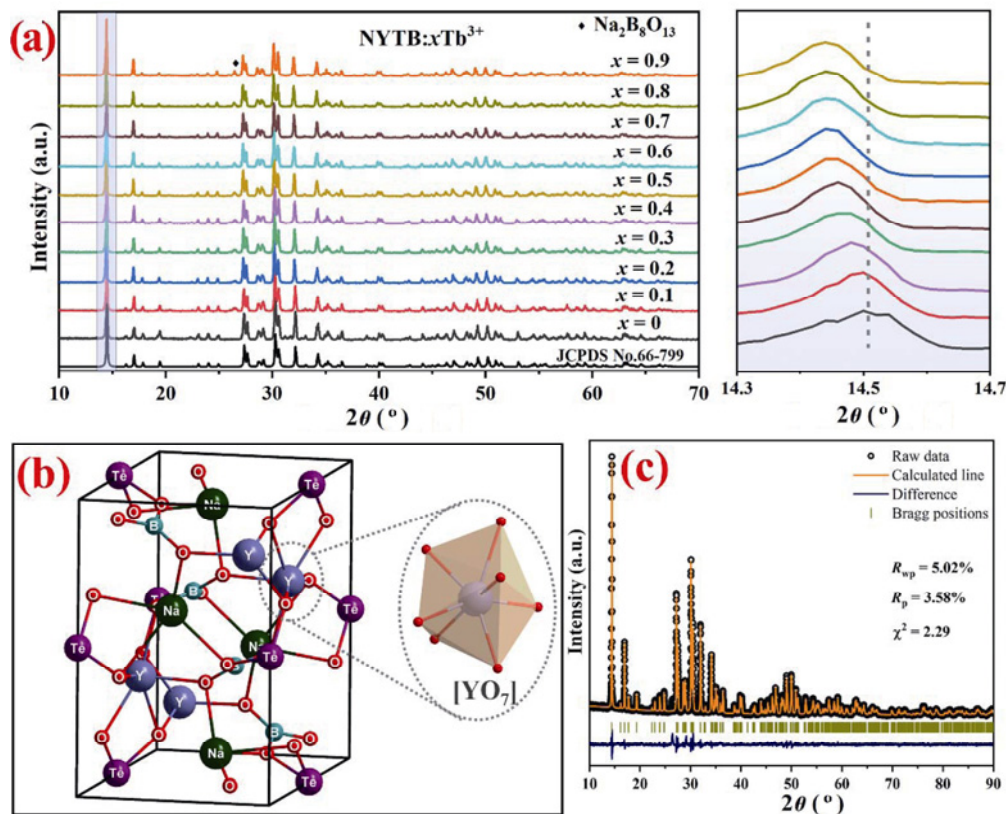


Fig. 1 (a) XRD patterns of NYTB: $x\text{Tb}^{3+}$ ($x = 0\text{--}90$ mol%) and magnified view of dominant diffraction peak. (b) Crystal structure of NYTB and coordination of Y^{3+} . (c) Rietveld refinement pattern for NYTB:70% Tb^{3+} .

Table 1 Rietveld refinement data for NYTB:70%Tb³⁺

Sample	NYTB [28]	NYTB:70%Tb ³⁺ (this work)
System	Monoclinic	Monoclinic
Space group	<i>P</i> 2 ₁ / <i>c</i> (14)	<i>P</i> 2 ₁ / <i>c</i> (14)
<i>a</i> (Å)	6.3101	6.3227
<i>b</i> (Å)	9.9363	9.9750
<i>c</i> (Å)	6.7343	6.7547
<i>V</i> (Å ³)	409.286	412.66
<i>Z</i>	2	2
2θ (°)	10–90	10–90
<i>R</i> _{wp} (%)	6.52	5.02
<i>R</i> _p (%)	4.31	3.58
χ ²	3.852	2.29

Note: *Z* is the number of molecules in a unit cell.

3.2 Morphological and elemental analysis

The morphology, particle size, and crystallinity of

phosphor particles have a significant impact on photoluminescent behaviors. Hereby, the morphological discussion for the studied phosphor is indispensable. According to Fig. 2(a) (scale bar = 30 μm), particle morphology for NYTB:70%Tb³⁺ powder is unevenly shaped. Combined with Fig. 2(b) (scale bar = 3 μm), most of the particles are micron-sized within the range of 3–15 μm, which coincides well with the size requirements of commercial w-LED phosphor particles [32]. Meanwhile, the TEM image discloses a smooth surface of the measured particle (Fig. 2(c)). Figure 2(d) presents the corresponding high-resolution TEM (HRTEM) image with clearly visible lattice fringes. The *d*-spacing of 3.02 Å corresponds to the (031) crystal plane of the diffraction peak (2θ = 30.12°), revealing that the studied phosphor maintains good crystallization. According to the elemental investigation (Fig. 2(e)), the NYTB:70%Tb³⁺ powder indeed consists of the

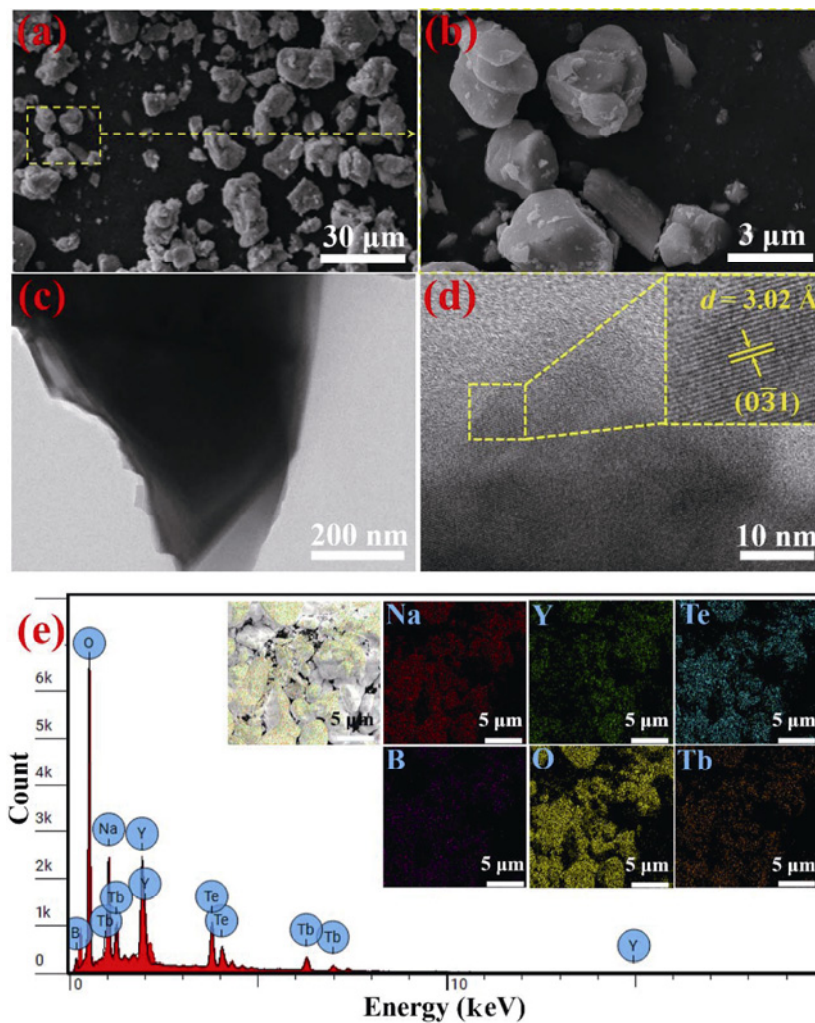


Fig. 2 (a, b) SEM images. (c) TEM image. (d) HRTEM image. (e) Energy-dispersive X-ray spectroscopy (EDS) data for NYTB:70%Tb³⁺ (the inset shows elemental mappings).

elements of Na, Y, Te, B, O, and Tb, which present an even distribution without any additional element.

To accurately determine elemental oxidation states of the representative NYTb:70%Tb³⁺ powder, the corresponding XPS results are illustrated in Fig. 3. A survey scan spectrum shows typical signals of Tb 3d, O 1s, B 1s, Te 3d, Y 3d, and Na 1s (Fig. 3(a)), supporting the obtained EDS data (Fig. 2(e)). For Tb 3d (Fig. 3(b)), the XPS spectrum clearly presents Tb 3d_{5/2} (1241.24 eV) and Tb 3d_{3/2} (1275.91 eV) peaks with a reasonable binding energy difference (Δ) of 34.67 eV, revealing a +3 oxidation state of terbium [33]. The XPS spectra for Na 1s, B 1s, O 1s, Te 3d, and Y 3d are presented in Fig. S2 in the ESM. The Na 1s peak at 1070.64 eV confirms a +1 valence of sodium [32]. The B 1s peak at 191.47 eV supports a +3 valence of boron [28]. An O 1s spectrum is well-fitted into the three peaks at 530.01, 531.27, and 535.13 eV. The peak with higher binding energy comes from the adsorbed oxygen [32], and the other peaks result from the lattice oxygen bonding with sodium, yttrium, tellurium, boron, and terbium [34]. For Te 3d, the Te 3d_{5/2} (576.08 eV) and Te 3d_{3/2} (586.46 eV) peaks with $\Delta = 10.38$ eV demonstrate a +6 valence of tellurium [31]. For Y 3d, the Y 3d_{5/2} (157.29 eV) and Y 3d_{3/2} (159.35 eV) peaks have the Δ value of 2.06 eV, manifesting a +3 valence of yttrium [28].

3.3 Photoluminescence (PL) properties

Figure 4(a) exhibits diffuse reflectance curves of the NYTb:*x*Tb³⁺ (*x* = 0–90 mol%). Charge transitions of a NYTb host cause the strong absorption below 300 nm,

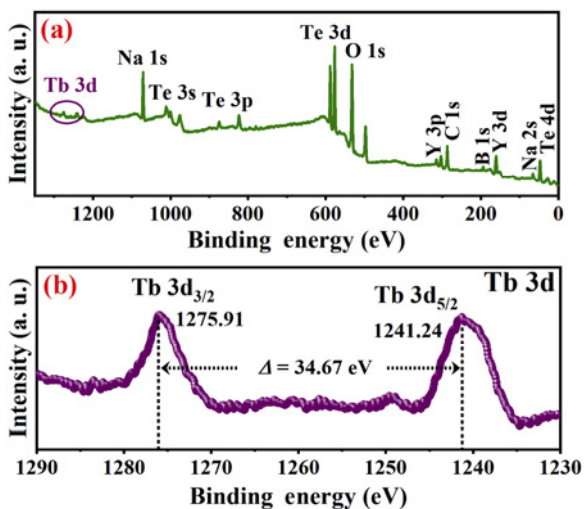


Fig. 3 (a) XPS survey scan spectrum. (b) Core-level spectrum of Tb 3d for NYTb:70%Tb³⁺.

including the two absorption band peaks at 217 and 284 nm. For the pure NYTb host, apart from a deep ultraviolet absorption band, there is no other absorption. For the Tb³⁺-doped samples, the obtained absorption peaks at 378 and 487 nm are due to the ⁷F₆→⁵D₃ and ⁷F₆→⁵D₄ transitions of Tb³⁺, respectively (the inset of Fig. 4(a)) [35]. Moreover, their intensity is gradually enhanced with the doping Tb³⁺ ions, which would further support the truth that the Tb³⁺ ions are successfully introduced into the NYTb lattice. According to the diffuse reflectance data, the Tb³⁺ content-dependent band gap (E_g) was determined. The E_g values are 3.54, 3.71, 3.67, 3.65, 3.67, 3.66, 3.67, 3.66, 3.66, and 3.70 eV, corresponding to *x* = 0, 10, 20, 30, 40, 50, 60, 70, 80, and 90 mol%, respectively (Fig. 4(b)), which are similar to those reported by Feng *et al.* [30]. Varying the Tb³⁺ contents have almost no impact on E_g of the Tb³⁺-activated NYTb phase, indicating that the prepared samples have good crystallization and few defects.

We used $\lambda = 378$ nm (⁷F₆→⁵D₃ of Tb³⁺) to irradiate the NYTb:*x*Tb³⁺ (*x* = 10–90 mol%) powder samples, according to PL excitation (PLE) curves (Fig. S3 in the ESM). The obtained PL spectra are illustrated in Fig. 5(a). Five PL components (497, 547, 585, 624, and 648 nm) come from the electronic transitions of Tb³⁺ between an excited state (⁵D₄) and ground states (⁷F₆, ⁷F₅, ⁷F₄, ⁷F₃, and ⁷F₂, respectively) [36]. Even though the Tb³⁺ contents varied, the shapes and positions of the emissions are unchanged besides the intensity. Integral PL intensity is dependent on the Tb³⁺ content, as given in Fig. 5(b). The intensity presents an optimum value, corresponding to *x* = 70 mol%. Subsequently, when the Tb³⁺ content exceeds 70 mol%, the intensity drops due to content-dependent luminescence quenching [37]. Moreover, a variation trend for the integral PL intensity excited at 487 nm (Fig. S4 in the ESM) is consistent with the results in Fig. 5(b). On the other hand, the PLE measurement also shows that the 70 mol% Tb³⁺ content is the optimal (Fig. S3 in the ESM). The results support reliability of luminescent data. Such high Tb³⁺ content added into host materials has been also reported, such as Ca₉Y(PO₄)₇:Tb³⁺ [38] and Sr₃Gd(PO₄)₃:Tb³⁺ [39].

Considering that there are generally re-absorption, exchange interaction, and multipolar interaction-induced concentration quenching [40], first of all, we compared the PL with the PLE of the representative NYTb:70% Tb³⁺ sample. The overlap with the range of 475–500 nm is observed (Fig. S5 in the ESM), revealing the probability of re-absorption. Subsequently, Blasse theory [41] was

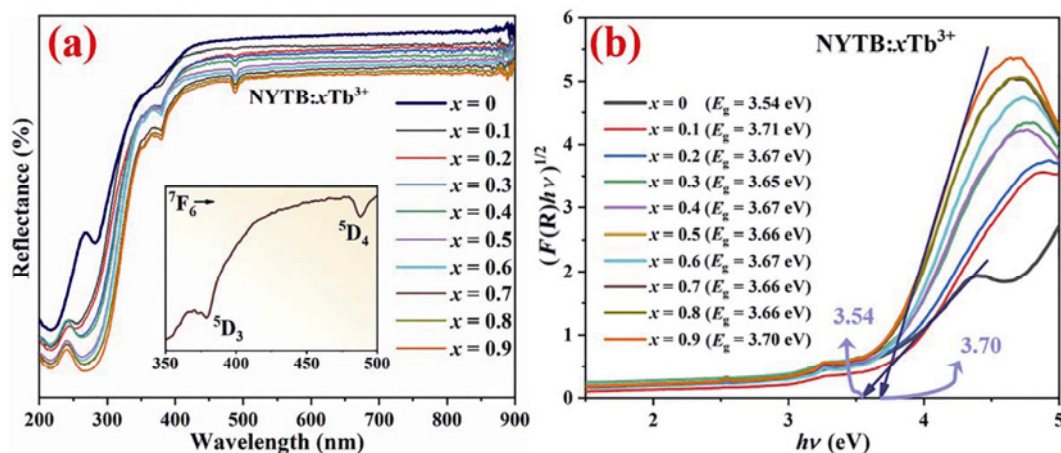


Fig. 4 (a) Diffuse reflectance spectra (the inset shows a magnified part in 350–500 nm for representative NYTb:70%Tb³⁺ sample). (b) E_g for NYTb: x Tb³⁺ ($x = 0$ –90 mol%).

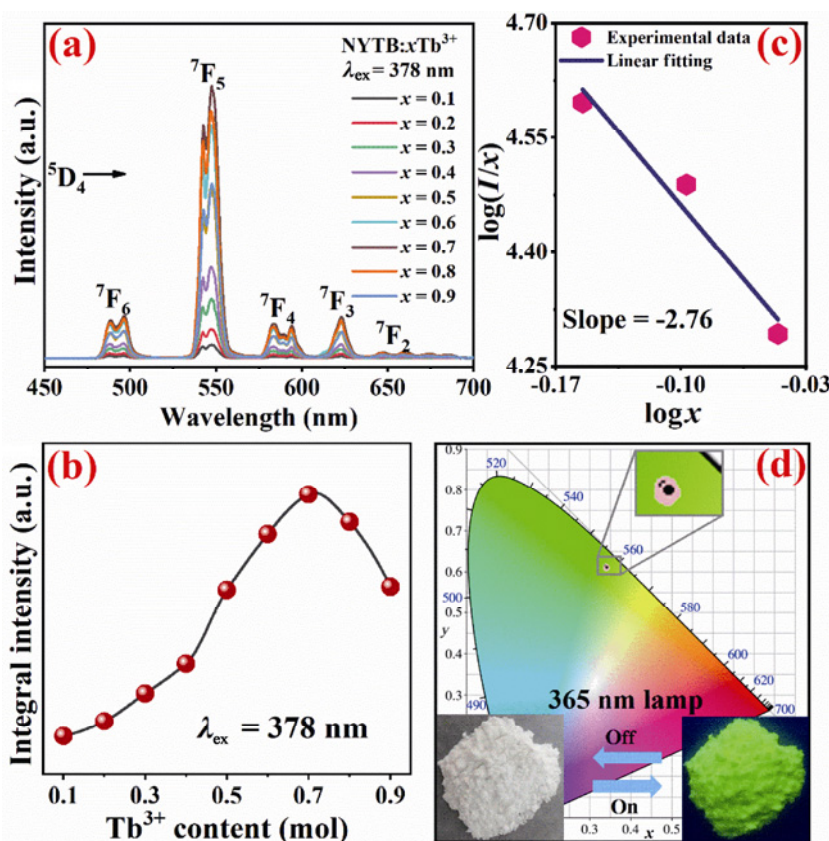


Fig. 5 (a) PL spectra of NYTb: x Tb³⁺ ($x = 10$ –90 mol%) excited at 378 nm. (b) Tb³⁺ content-dependent PL intensity. (c) Plot of $\log(I/x)$ vs. $\log x$. (d) CIE chromaticity diagram (the insets illustrate digital photos of respective NYTb:70%Tb³⁺ sample excited by 365 nm lamp). Note: λ_{ex} represents the excitation wavelength.

employed to discuss the critical distance (R_c) among the Tb³⁺ ions. In this system, the R_c value (6.55 Å) is larger than 5 Å, indicating that the non-radiative energy transfer is ascribed to the multipolar interaction rather than the exchange interaction. Finally, the $\log x$ dependence of $\log(I/x)$ was explored by a Dexter model (Eq. (S1) in the ESM) [42], as depicted in Fig. 5(c). The slope

(−2.76) of the fitting line means that $\theta = 8.28$, demonstrating that the concentration quenching is caused by the dipole–quadrupole interaction. As analyzed above, both re-absorption and dipole–quadrupole interaction contribute to the luminescent concentration quenching.

Based on the PL data excited at 378 nm, chromatic coordinates of the representative NYTb:70%Tb³⁺

phosphor were obtained, which are located in the green region (Fig. 5(d)). The inset of Fig. 5(d) shows an image of a green-emitting sample excited by a portable 365 nm lamp. The Tb^{3+} concentration-dependent chromatic coordinates are summarized in Table 2. The coordinates are slightly changed with the increasing Tb^{3+} ions, falling into the green region. Moreover, color purity (CP) is calculated by Eq. (S2) in the ESM. The CP values are above 73% (Table 2), which is well comparable to the data of the Tb^{3+} -activated phosphors reported such as 73% for $\text{Bi}_3\text{TeBO}_9:0.11\text{Tb}^{3+}$ [31], 19.8% for $\text{Sr}_3\text{Gd}(\text{PO}_4)_3:0.01\text{Tb}^{3+}$ [39], 68% for $\text{Ba}_3(\text{ZnB}_5\text{O}_{10})\text{PO}_4:0.07\text{Tb}^{3+}$ [43], 64.13% for $\text{CaLa}_4\text{Si}_3\text{O}_{13}:0.07\text{Tb}^{3+}$ [44], and 86% for $\text{Na}_3\text{La}_{0.92}(\text{VO}_4)_2:0.08\text{Tb}^{3+}$ [45].

Figure 6(a) shows decay curves ($\lambda_{\text{ex}} = 378$ nm and the emission wavelength ($\lambda_{\text{em}} = 547$ nm) of the NYTB: $x\text{Tb}^{3+}$ ($x = 10\text{--}90$ mol%) phosphors, which could be fitted by a two-exponential model by Eq. (S3) in the ESM. The results indicate the presence of two Tb^{3+} emitting centers. Tb^{3+} activators have a seven-fold coordination by replacing Y^{3+} ions. Besides, a part of Tb^{3+} activators probably substitute Na^+ ions or appear at a surface of the powder particles due to the high Tb^{3+} concentration [46,47]. Rapid, slow, and average

decay time (marked as τ_1 , τ_2 , and τ_{ave} , respectively) are listed in Table S2 in the ESM. The τ_{ave} value gradually decreases with the increasing Tb^{3+} concentration (Fig. 6(b)), originating from the increased non-radiative transition among Tb^{3+} ions. Besides, quantum efficiency is a significant parameter for the phosphor materials. Hereby, internal quantum efficiency (IQE) and external quantum efficiency (EQE) of the NYTB:70% Tb^{3+} are measured and estimated by Eq. (S4) in the ESM. The IQE and EQE values are determined to be 46.4%, and 6.7%, respectively (Fig. 6(c)). Finally, we constructed

Table 2 Chromaticity coordinates and CP of NYTB: $x\text{Tb}^{3+}$ ($x = 10\text{--}90$ mol%)

x (mol%)	Coordinate (x, y)	CP (%)
10	(0.3396, 0.6079)	73.5
20	(0.3386, 0.6095)	74.0
30	(0.3390, 0.6108)	74.5
40	(0.3391, 0.6116)	74.8
50	(0.3405, 0.6117)	74.8
60	(0.3422, 0.6110)	74.7
70	(0.3419, 0.6131)	75.7
80	(0.3392, 0.6146)	75.6
90	(0.3419, 0.6120)	75.5

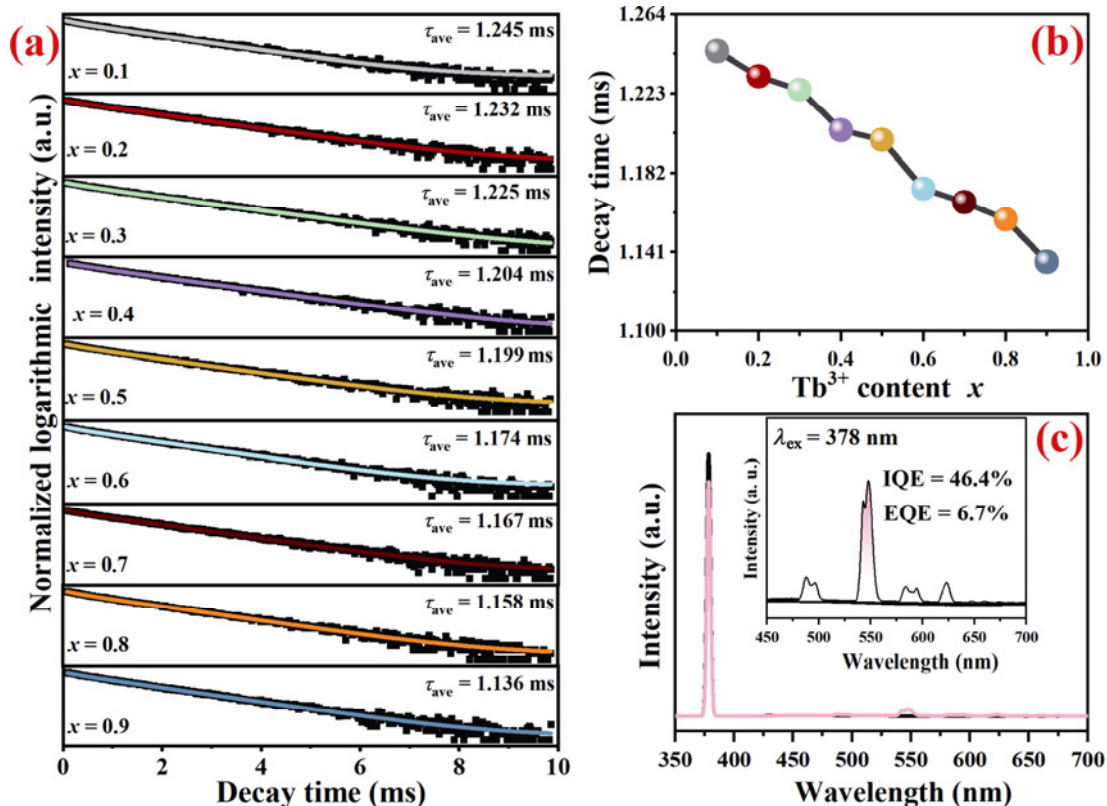


Fig. 6 (a) Decay curves of NYTB: $x\text{Tb}^{3+}$ ($x = 10\text{--}90$ mol%). (b) Decay time in response to Tb^{3+} content. (c) IQE and EQE of NYTB:70% Tb^{3+} .

a w-LED device (Fig. S6 in the ESM), in which the studied NYTb:70%Tb³⁺ phosphor is combined with a near-UV (NUV) chip (380 nm) and commercial phosphors (BaMgAl₁₀O₁₇ (BAM):Eu²⁺ (blue) and (Ca,Sr)AlSiN₃:Eu²⁺ (red)). Driven by a current of 100 mA, the w-LED device emits bright white light with $R_a = 85$, CCT = 5125 K, and the CIE = (0.3413, 0.3427) where R_a is the color render index, and CCT is the correlated color temperature. However, the temperature dependence of the PL spectra for the NYTb:70%Tb³⁺ reveals weak luminescent thermostability (Fig. S7 in the ESM). The results also indicate a potential optical temperature sensing behavior for the studied phosphor.

3.4 Temperature sensing properties

The temperature dependence of the PL spectra for the NYTb:70%Tb³⁺ shows that the integral intensity descends dramatically with the rising temperature (Fig. S7 in the ESM). Notably, in the range of 300–475 K, the decreasing trends of 497 nm (⁵D₄→⁷F₆), 547 nm (⁵D₄→⁷F₅), and 585 nm (⁵D₄→⁷F₄) peaks are various, as given in Fig. S10 in the ESM. The PL intensity at 547 nm decreases markedly, whereas those at 497 and 585 nm descend slowly. The results reveal the

feasibility of detecting the temperature signals based on the FIR among these peaks. Figures 7(a) and 7(c) illustrate the FIR(I_{497}/I_{547}) and FIR(I_{585}/I_{547}) as a function of the temperature, respectively, which are fitted via a semi-exponential model (Eq. (S5) in the ESM). The high R^2 values ($R^2 = 0.997$ for the FIR(I_{497}/I_{547}) and 0.999 for the FIR(I_{585}/I_{547})) mean the reliability of the fitting results. Subsequently, to further evaluate the temperature sensing properties, S_A and S_R are obtained by Eqs. (S6) and (S7) in the ESM, respectively. The S_R and S_A data for the FIR(I_{497}/I_{547}) and FIR(I_{585}/I_{547}) in response to the temperature are given in Figs. 7(b) and 7(d), respectively. Both S_R and S_A values at 300 K reach the maximum ($S_{R-max} = 5.23 \% \cdot K^{-1}$ and $S_{A-max} = 1.01 \times 10^{-2} K^{-1}$ for the FIR(I_{497}/I_{547}); $S_{R-max} = 5.74 \% \cdot K^{-1}$ and $S_{A-max} = 1.02 \times 10^{-2} K^{-1}$ for the FIR(I_{585}/I_{547})).

The temperature resolution (δT) is a key parameter for the luminescent thermometry, which is determined by Eq. (S8) in the ESM. To obtain an accurate $\delta FIR/FIR$, the FIR(I_{497}/I_{547}) value was measured about thirty times, as shown in Fig. 8(a). Based on the standard deviation, the $\delta FIR/FIR$ for the FIR(I_{497}/I_{547}) is defined as 0.1%. The δT values are 0.019 and

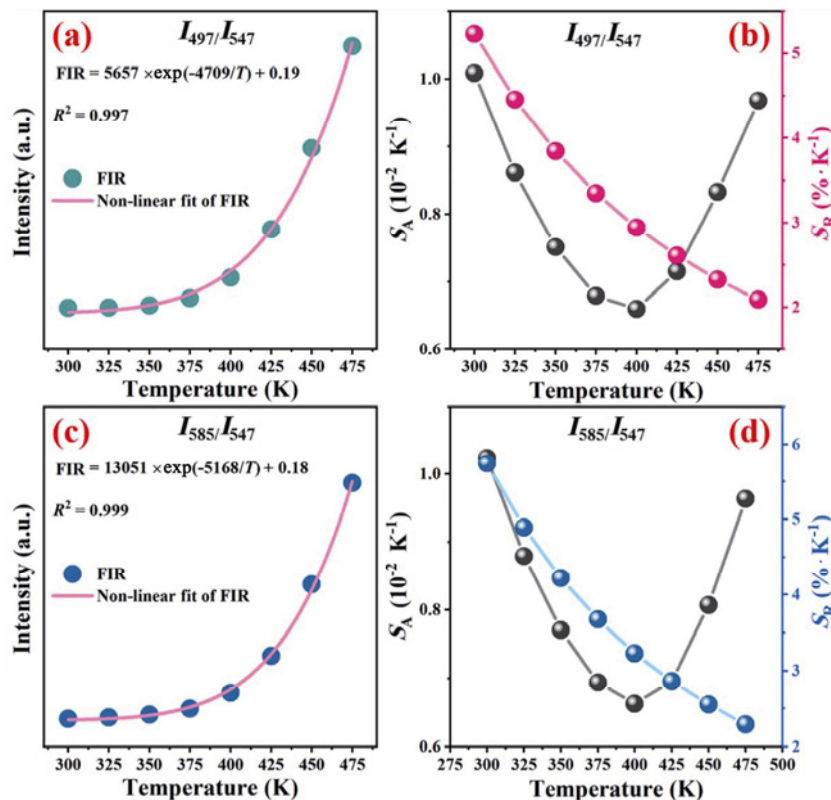


Fig. 7 (a) Variation of FIR(I_{497}/I_{547}). (b) Changes of S_A and S_R for FIR(I_{497}/I_{547}). (c) Variation of FIR(I_{585}/I_{547}). (d) Changes of S_A and S_R for FIR(I_{585}/I_{547}) in response to temperature.

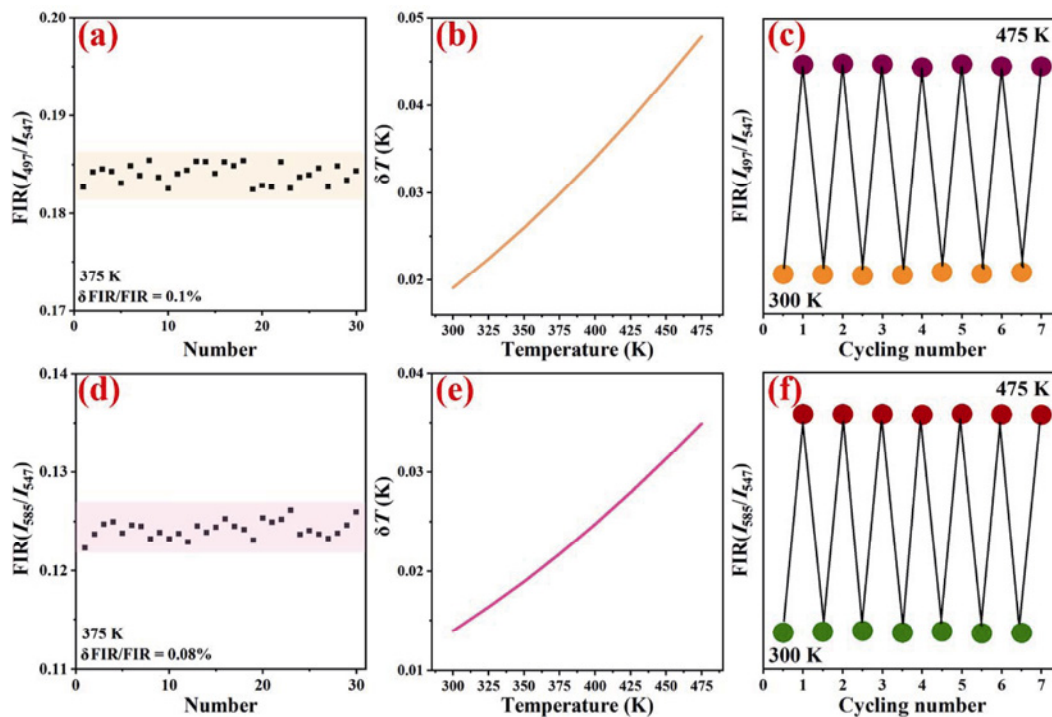


Fig. 8 (a) Thirty measurements of $\text{FIR}(I_{497}/I_{547})$. (b) δT of $\text{FIR}(I_{497}/I_{547})$. (c) Thermal repeatability of $\text{FIR}(I_{497}/I_{547})$ between 300 and 475 K. (d) Thirty measurements of $\text{FIR}(I_{585}/I_{547})$. (e) δT of $\text{FIR}(I_{585}/I_{547})$. (f) Thermal repeatability of $\text{FIR}(I_{585}/I_{547})$ between 300 and 475 K.

0.048 K at 300 and 475 K, respectively (Fig. 8(b)). Meanwhile, the thermal repeatability (R) is performed by temperature cycle measurements under the same conditions, which show excellent repeatability and reversibility after seven cycling experiments, as displayed in Fig. 8(c). Figure S9(a) in the ESM shows the one temperature cycle measurement PL spectra. The PL spectra of heating match well with the PL spectra of cooling. In the thermal cycle, the $\text{FIR}(I_{497}/I_{547})$ values return to the original strength (Fig. S9(b) in the ESM). Additionally, the $\delta\text{FIR}/\text{FIR}$ for the $\text{FIR}(I_{585}/I_{547})$ is defined as 0.08% (Fig. 8(d)). The δT value is changed from 0.014 to 0.035 K at the rising temperatures from 300 to 475 K (Fig. 8(e)). Figure 8(f) shows a temperature cycle process, which exhibits good repeatability for a $\text{FIR}(I_{585}/I_{547})$ model. Meanwhile, the one cycle of the $\text{FIR}(I_{585}/I_{547})$ model is presented in Fig. S9(c) in the ESM, indicating excellent repeatability for luminescent thermometric applications.

In addition, based on the PLE data of the NYTB:70% Tb^{3+} phosphor in response to the temperature (Fig. S10 in the ESM), the thermometric properties were also discussed. The EIR between the peaks at 378 nm (${}^7\text{F}_6 \rightarrow {}^5\text{D}_3$) and 487 nm (${}^7\text{F}_6 \rightarrow {}^5\text{D}_4$) is applied to the excited state thermometry. Figure 9(a) shows a variation of the EIR in response to the temperature,

which was fitted by Eq. (S5) in the ESM. The fitting results are reasonable due to $R^2 = 0.999$. Equations (S6) and (S7) in the ESM are employed to evaluate S_A and S_R for the $\text{EIR}(I_{378}/I_{487})$, respectively. According to Fig. 9(b), the largest S_R and S_A values are $2.49 \text{ \%}\cdot\text{K}^{-1}$ and $2.16 \times 10^{-2} \text{ K}^{-1}$, respectively.

Similarly, the $\delta\text{EIR}/\text{EIR}$ and δT values for the EIR model are evaluated, as shown in Figs. 10(a) and 10(b), respectively. The $\delta\text{EIR}/\text{EIR}$ is determined as 0.38%. The δT value is changed from 0.15 to 0.38 K upon the rising temperatures from 300 to 475 K. The temperature cycle process is carried out to assess the repeatability and reversibility. Figure 10(c) displays excellent temperature repeatability of the EIR model. The PLE spectra in the one temperature cycle measurement show a similar trend, revealing good repeatability and reversibility for the EIR model (Fig. S11 in the ESM).

Based on the FIR/EIR model, in contrast to some reported thermometric materials, the NYTB:70% Tb^{3+} phosphor demonstrates satisfactory S_R and S_A parameters, as listed in Table 3. Compared with the FIR model, the EIR-based temperature sensing technology for rare-earth-ion-activated phosphors is insufficiently investigated. Moreover, there are few thermometric materials reported, which can present the thermometric properties based on both FIR and EIR technologies.

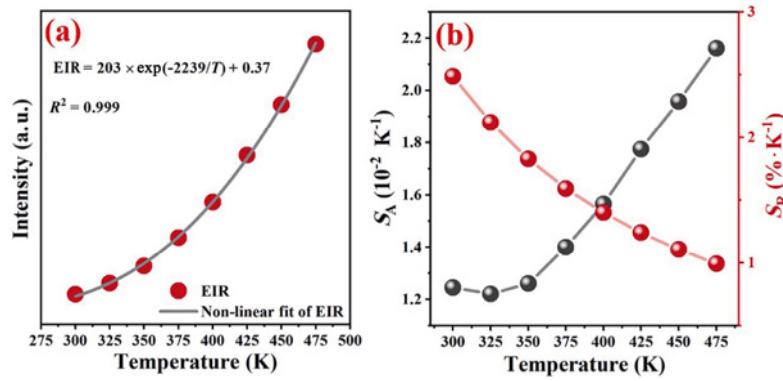


Fig. 9 (a) Variation of EIR. (b) Changes of S_A and S_R in response to temperature.

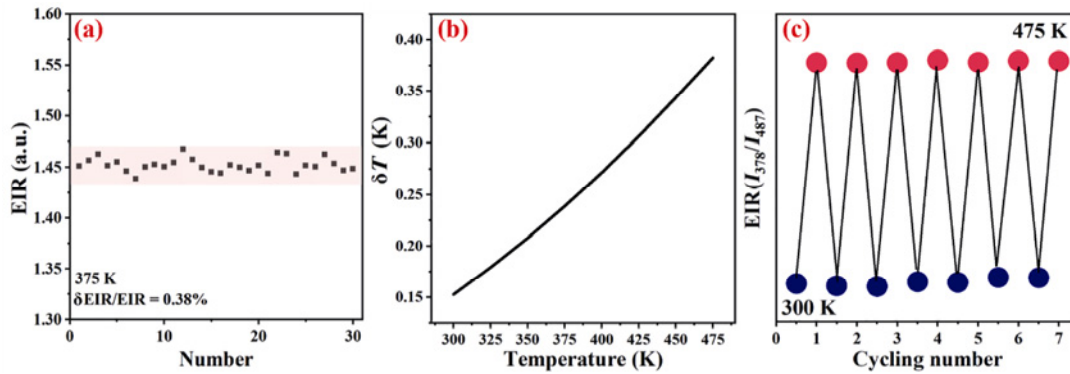


Fig. 10 (a) Thirty measurements of EIR. (b) δT of EIR(I_{378}/I_{487}). (c) Thermal repeatability between 300 and 475 K.

Table 3 S_{R-max} and S_{A-max} parameters of reported thermometric materials

Material	Model	S_{R-max} (%·K ⁻¹)	S_{A-max} (10 ⁻² K ⁻¹)	T (K)	Ref.
YVO ₄ :Eu ³⁺	EIR	1.19	0.038	298–473	[10]
KLaP ₄ O ₁₂ :Tb ³⁺	EIR	5.5	—	273–473	[23]
La ₂ ZnTiO ₆ :Tb ³⁺	EIR	0.734	4.46	293–573	[27]
Ca ₂ LaNbO ₆ :Sm ³⁺	EIR	0.28	—	313–573	[48]
CaWO ₄ :Nd ³⁺	EIR	0.96	0.939	298–573	[49]
K ₃ LuSi ₂ O ₇ :Eu ³⁺	EIR	0.539	2.03	313–573	[50]
NYTB:Tb ³⁺	EIR	2.49	2.16	300–475	This work
YVO ₄ :Eu ³⁺	FIR	1.28	0.581	298–466	[10]
Ca ₂ LaNbO ₆ :Sm ³⁺	FIR	0.23	—	313–573	[48]
LaMg _{0.402} Nb _{0.598} O ₃ :Pr ³⁺	FIR	0.83	6	298–523	[51]
NaLuF ₄ :Eu ³⁺ @g-C ₃ N ₄	FIR	0.455	5.7	303–503	[52]
CaWO ₄ :Tb ³⁺	FIR	1.21	—	343–783	[53]
LuNbO ₄ :Pr ³⁺ /Tb ³⁺	FIR	1.26	2.4	283–493	[54]
Ba _{1.5} Sr _{0.5} Ga ₄ O ₈ :Bi ³⁺	FIR	1.295	0.5	7–400	[55]
CaBaZn ₂ Ga ₂ O ₇ :Bi ³⁺	FIR	1.453	15.4	298–473	[56]
Sr ₂ LaF ₇ :Er ³⁺	FIR	0.639	40.9	303–433	[57]
LaTiSbO ₆ :Mn ⁴⁺ /YAG:Ce ³⁺ -PiG	FIR	1.61	3.58	273–418	[58]
Y ₂ MgTiO ₆ :Mn ⁴⁺	FIR	0.142	—	10–513	[59]
MgTiO ₃ :Mn ⁴⁺	FIR	1.2	—	133–363	[60]
SrAl ₁₂ O ₁₉ :Mn ⁴⁺	FIR	0.27	0.4	273–393	[61]
CaNb ₂ O ₆ :Bi ³⁺ /Eu ³⁺	FIR	3.793	12.2	298–523	[62]
Gd ₂ ZnTiO ₆ :Bi ³⁺ ,Mn ⁴⁺	FIR	2.4	16.8	313–473	[63]
NYTB:Tb ³⁺	FIR	5.74	1.02	300–475	This work

Note: YAG and PiG represent Y₃Al₅O₁₂ and phosphor-in-glass, respectively.

4 Conclusions

In summary, NYTB: $x\text{Tb}^{3+}$ ($x = 10\text{--}90$ mol%) phosphors were developed by a solid-state reaction. The luminescent content quenching mechanism is determined. Under the excitation at 378 nm, the representative NYTB:70% Tb^{3+} phosphor shows green light with the coordinates of (0.3419, 0.6131) and an IQE of 46.4%. Importantly, the excellent thermometric properties for the NYTB:70% Tb^{3+} are illustrated based on the proposed FIR/EIR model. The obtained $S_{R\text{-max}}/S_{A\text{-max}}$ for the FIR(I_{497}/I_{547}), FIR(I_{585}/I_{547}), and EIR(I_{487}/I_{378}) signals are ($5.23\% \cdot \text{K}^{-1}/1.01 \times 10^{-2} \text{K}^{-1}$), ($5.74\% \cdot \text{K}^{-1}/1.02 \times 10^{-2} \text{K}^{-1}$), and ($2.49\% \cdot \text{K}^{-1}/2.16 \times 10^{-2} \text{K}^{-1}$), respectively. The good δT and repeatability are confirmed. The results suggest that the studied phosphor is a well-performing FIR-/EIR-based thermometric material. This work provides a new perspective on the noncontact optical thermometry based on the FIR/EIR technologies.

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Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material

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