

Heterophase engineering enables efficient tin-based perovskite red light-emitting diodes for cardiac pulse detection

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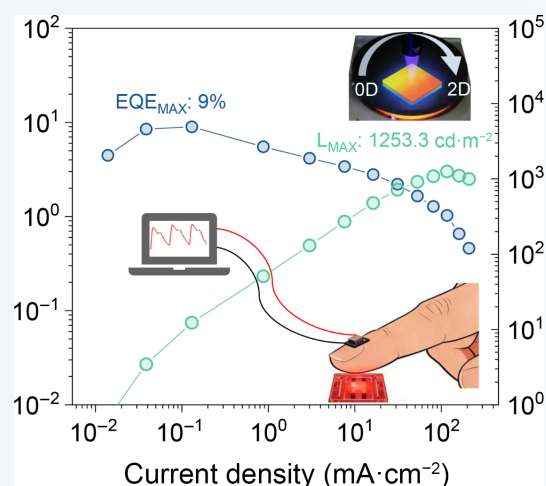
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ABSTRACT: Tin (Sn)-based perovskite light-emitting diodes (LEDs), featuring environmental friendliness and low toxicity, have attracted growing interest as light sources for high-definition displays, medical and health monitoring applications. However, the widespread existing defects arising from the rapid crystallization and the undesirable oxidation of Sn^{2+} markedly limit the efficiency and brightness of current Sn-based perovskite LEDs. Herein, we develop an zero-dimensional (0D)–two-dimensional (2D) heterophasic transition strategy to promote the performance of PEA_2SnI_4 perovskite films. The results demonstrate that the formation of heterophase extends the processing time window of perovskites, thereby enabling effective modulation and optimization of films crystallization process by regulating initial nucleation. Finally, the fabricated device based on the optimized PEA_2SnI_4 films achieves a maximum external quantum efficiency (EQE) of 9% and a luminance of $1253.3 \text{ cd}\cdot\text{m}^{-2}$, representing one of the highest performances reported to date for PEA_2SnI_4 -based perovskite LEDs. Furthermore, the fabricated device was successfully employed as light sources for human cardiac pulse detection, achieving performances comparable to that of commercial detectors, representing the first demonstration of such functionality in lead-free perovskite systems.

KEYWORDS: PEA_2SnI_4 , heterophase engineering, light-emitting diodes, cardiac pulse detection



1 Introduction

Recently, halide perovskite light-emitting diodes (LEDs) due to their excellent optoelectronic properties like high efficiency, narrowband emission, wide color gamut, and low-cost solution-processing, have emerged as one of the most promising semiconductor families for next-generation light source technologies [1–4]. Over the past few years, electrically driven perovskite LEDs technology has advanced rapidly. Although lead (Pb)-based perovskite LEDs have made a vigorous development, and their external quantum efficiencies (EQEs) have exceeded

30% [5–11], the toxicity of Pb has raised concerns regarding environmental and health-related issues [12]. Thus, the development of high-performance lead-free perovskite LEDs has become imperative. Currently, numerous theoretical and experimental efforts have been devoted to screen suitable elements to replace toxic Pb, and many promising candidates have been identified. Among them, substitution of Pb^{2+} by homovalent group-14 element Sn^{2+} is regarded as the most promising approach because of their similar ionic radii and electron structure [12–14]. The newly explored two-dimensional (2D) perovskite PEA_2SnI_4 represents an ideal pure-red emitter, characterized by a narrowband emission centered at 628 nm, which holds broad application potential in medical detection and fully complies with the Rec. 2020 standard [15, 16]. However, the rapid crystallization rate of tin-based perovskites often results in poor film morphology and coverage [17, 18]; besides, Sn^{2+} is highly prone to oxidation into Sn^{4+} , leading to the formation of Sn vacancy defects [19]. These issues

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collectively degrade the electroluminescent (EL) performance of tin-based perovskite LEDs significantly. To this end, extensive efforts have been devoted to address this issues by introducing various reducing agents and additives, such as Sn powder, SnF_2 , and amino derivatives to suppress the oxidation of Sn^{2+} and enhance the quality of PEA_2SnI_4 films [20–23]. Despite this, the device performance based on PEA_2SnI_4 emitter remains highly unsatisfactory; in particular, efficiency and luminance still cannot be simultaneously achieved.

In the optimization of lead-based perovskite LEDs, the construction of heterophase structures to enhance EL performance has been widely adopted. For instance, Yuan et al. improved the stability and charge transport properties of CsPbI_3 quantum dots through a heteroepitaxy approach [8]. Similarly, Tang et al. regulated the ratio of components in the precursor solution to manipulate the heterophase distribution of CsPbBr_3 nanocrystals, thereby enhancing the luminescent performance of the films [24]. However, the utilization of heterophase to boost the performance of tin-based perovskite LEDs has rarely been reported. Therefore, investigating heterophase strategies aimed at improving the luminescent performance and stability of tin-based perovskite films is both pioneering and significant.

Herein, we propose an zero-dimensional (0D)–2D heterophasic transition strategy to fabricate high-performance PEA_2SnI_4 perovskite LEDs by modulating the phenethylammonium iodide (PEAI) component in the precursor solution. The results reveal that the addition of excess PEAi leads to the formation of PEA_6SnI_8 phase during the spin-coating process. The presence of PEA_6SnI_8 phase significantly slows down the crystallization rate of perovskite films, which facilitates the growth of high-quality films. Meanwhile, the formed PEA_6SnI_8 phase converts to the PEA_2SnI_4 phase and PEAi during the annealing process [25], where the residual PEAi molecules passivate defects and suppress the non-radiative recombination losses [26, 27]. Consequently, we fabricated an efficient pure-red tin-based perovskite LED with PEA_2SnI_4 films, achieving a maximum EQE of 9% and a luminance of $1253.3 \text{ cd}\cdot\text{m}^{-2}$, ranking among the highest-performing PEA_2SnI_4 -based LEDs reported to date [28]. Inspired by the device performances, we integrated it with a silicon-based detector to create a sensor capable of accurately monitoring human cardiac pulse [29–32], demonstrating its promising potential for applications in wearable medical devices.

2 Results and discussion

The composition of the precursor solution is of crucial importance in the preparation of perovskite films. Typically, a molar ratio of 2:1 for PEAi to SnI_2 is adopted to prepare the PEA_2SnI_4 precursor solution. However, our investigation into the effect of precursor solution composition revealed that varying the amount of PEAi significantly affects the resulting perovskite films. To assess the influence of PEAi ratio on the films quality, we characterized the perovskite films with varying PEAi ratios by photoluminescence (PL). As shown in Fig. S1 in the Electronic Supplementary Material (ESM), the PL intensity initially increased and then declined with increasing PEAi content, reaching their maximum at a molar ratio of 2.4:1 for PEAi: SnI_2 . Correspondingly, a similar trend was observed in the PL lifetime measurements (Fig. S2 and Table S1 in the ESM). Based on the above results, we therefore selected the samples with a PEAi: SnI_2 molar ratio of 2.4:1 for subsequent

studies. The PEA_2SnI_4 films were fabricated by the solution spin-coating method, with the detailed spin-coating procedures illustrated in Fig. 1(a). During the spin-coating process, 100 μL of chlorobenzene (CB) was added as the antisolvent at 40 s to assist crystallization. The films were then transferred to a hotplate for annealing at 75 °C (see Experimental section for full details). Notably, films with excess PEAi exhibited orange-yellow emission when excited by a 365 nm UV lamp during spin-coating. Upon the subsequent annealing process, the luminescence color of the films rapidly changed to pure-red and remained stable afterward (Movie S1 in the ESM). To directly probe the effect of excess PEAi on perovskite crystallization, *in-situ* PL measurements were performed during the spin-coating process (Figs. 1(b) and 1(c)). Films prepared with excess PEAi and stoichiometric ratio were denoted as target and control samples, respectively.

Given that the annealing process significantly influences the luminescence color of the target sample, we first compared the PL characteristics of the control and target samples prior to annealing. Therefore, for the target films, we defined an intermediate state denoted as target (w/o annealing) sample. As shown in Fig. S3 in the ESM, the target (w/o annealing) sample exhibits a broad emission band ranging from 460 to 800 nm, which can be deconvoluted into three sub-peaks located at approximately 625, 580, and 655 nm, respectively. The emission component located at 625 nm is consistent with the PL characteristics of the control sample, corresponding to the intrinsic PL of PEA_2SnI_4 ; whereas the emission components at 580 and 655 nm most likely originate from the PEA_6SnI_8 crystals [33, 34]. To further verify the formation of PEA_6SnI_8 phase in the target (w/o annealing) films, time-resolved PL (TRPL) spectroscopy and wavelength-dependent TRPL spectroscopy were carried out. At the wavelength of 580 nm, corresponding to the characteristic emission of PEA_6SnI_8 , the target (w/o annealing) films exhibit a microsecond-scale lifetime (0.32 μs), which is comparable to the PL lifetime of pure PEA_6SnI_8 but is absent in the control sample (Fig. S4 in the ESM) [25]. Additionally, in the wavelength-dependent TRPL measurements (Fig. S5 in the ESM), the target (w/o annealing) films exhibited a PL lifetime profile similar to that of the pure PEA_6SnI_8 . These results are consistent with our initial hypothesis that the PEA_6SnI_8 phase is indeed formed in the target films during the spin-coating process.

To further analyze the growth process of the perovskite films, we extracted PL intensity and full width half maximum (FWHM) data from the *in-situ* PL measurements and plotted them as a function of time (Fig. 1(d) and Fig. S6 in the ESM). During the antisolvent addition stage, PL is generated and collected immediately, with the emission intensity increasing sharply (Fig. S6 in the ESM). This behavior can be attributed to the antisolvent triggering rapid perovskite nucleation and promoting crystal growth. The control films exhibited narrow-band emission consistent with the reported luminescence characteristics of PEA_2SnI_4 [15], while the target (w/o annealing) films showed a FWHM of approximately 150 nm throughout the spin-coating process. During annealing, the FWHM of the control films broadened to 31 nm, accompanied by an increase in emission intensity, and subsequently stabilized at this value. In contrast, the FWHM of the target films narrowed rapidly from 150 nm to a final value of 29 nm. The slight broadening observed for the control films during annealing is attributed to the increased defect states induced by rapid grain growth. Conversely, the rapid narrowing of FWHM of the target films is due to the annealing process facilitating the formation of the PEA_2SnI_4 phase

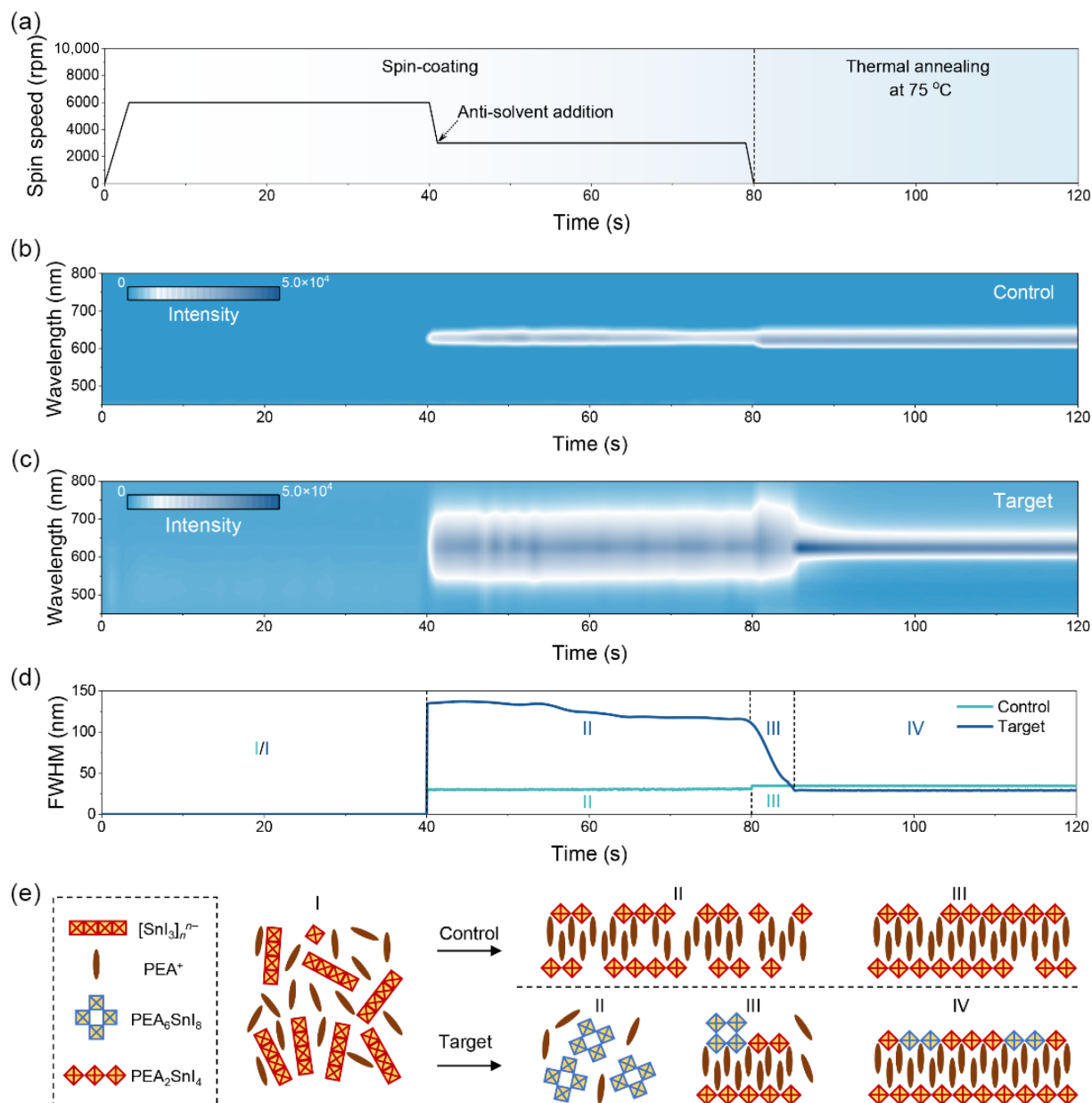


Figure 1 *In-situ* PL measurements of Sn-based perovskites during the fabrication process. (a) Spin-coating speed-time profile for preparing PEA₂SnI₄ films. (b) and (c) Contour plots of PL spectra during the spin-coating and annealing processes of the control and target PEA₂SnI₄ films. (d) Evolution of the FWHM for control and target films extracted from the *in-situ* PL spectra. (e) Schematic crystallization pathways of the control and target PEA₂SnI₄ films.

[25]. Concurrently, the unstable PEA₆SnI₈ phase decomposes rapidly into the PEA₂SnI₄ phase and PEA₂I. This process ultimately yields a heterophase system comprising a predominant amount of high-quality PEA₂SnI₄ alongside a minor residual fraction of PEA₆SnI₈ phase. Throughout both the spin-coating and annealing processes, the target sample sustained substantially higher PL intensity compared to the control one. This enhanced emission suggests that excess PEA₂I effectively passivates defects generated during films growth, leading to observed improvement in PL intensity.

Based on the above experimental results, the proposed crystallization mechanisms of the control and target samples are illustrated in Fig. 1(e). During spin-coating, the removal of solvent molecules by the anti-solvent generates numerous nucleation sites, triggering the rapid aggregation of perovskite clusters. For the

control samples, PEA⁺ ions crystallize rapidly under van der Waals forces, resulting in a disordered distribution of 2D perovskites in the final films. This structural disorder not only introduces additional defects but also hinders energy transfer. In contrast, for the target sample, excess PEA₂I induces the initial formation of both PEA₆SnI₈ and PEA₂SnI₄, thereby slowing down the overall crystallization kinetics. Residual free PEA₂I further passivates films defects and enhances the luminescence intensity. During the subsequent annealing process, the metastable PEA₆SnI₈ phase decomposes thermally into the PEA₂SnI₄ phase. This transformation not only remedies the defects at grain boundaries but also improves the purity and intensity of the pure-red emission.

To trace the evolution of the PEA₆SnI₈ phase in the films preparation, we further performed the grazing-incidence wide-angle X-ray scattering (GIWAXS) measurements on three types of

samples: the control films, the target (w/o annealing) films, and the target (annealed) films (Figs. 2(a)–2(c)). The GIWAXS results showed that the control films exhibited no characteristic peaks corresponding to the PEA_6SnI_8 phase. In contrast, characteristic peaks of the PEA_6SnI_8 phase were detected in both target films. Notably, compared to the target (w/o annealing) films, the target (annealed) films show attenuated PEA_6SnI_8 peaks alongside significantly intensified peaks of the PEA_2SnI_4 phase, consistent with our earlier hypothesis. Beyond the differences in characteristic peaks, two target films also displayed stronger diffraction ring intensities than the control films, indicating that the formation of the PEA_6SnI_8 phase enhances the crystallinity of the perovskite films. To provide a clearer view of the heterophase distribution, out-of-plane integration of the GIWAXS patterns (Figs. 2(d)–2(f)) was further conducted, and the results aligned well with the aforementioned observations, providing additional evidence that the introduction of excess PEAi effectively improves films crystallinity. The structural characteristics of three types of films were further identified by X-ray diffraction (XRD) (Figs. 2(g) and 2(h)). The control films showed typical (00*l*, *l* = 2, 4, 6, 8, 10, 12) diffraction peaks corresponding to the 2D perovskites. In contrast, both the target films (w/o annealing, annealed) exhibited not only the characteristic peaks of 2D perovskites but also an additional

peak at 5.04° belonging to the 0D PEA_6SnI_8 phase, which is consistent with previous characterization results [25]. In addition, the XRD patterns of two target films showed significantly higher diffraction intensities than those of the control films, further corroborating the enhanced crystallinity achieved with the addition of excess PEAi.

To directly visualize the PEA_6SnI_8 phase in the resulting films, we further performed the transmission electron microscopy (TEM) measurements. As shown in Fig. S7 in the ESM, the target (annealed) films showed reduced grain sizes, averaging approximately 100 nm, compared to the control films. High-resolution TEM was further utilized to examine the atomic structure. For the control films, only an interplanar spacing of 0.33 nm was observed, corresponding to the (0010) plane of the PEA_2SnI_4 phase. While, the target (annealed) films exhibited not only the 0.33 nm interplanar spacing but also a wider interplanar spacing of 0.73 nm, matching the (112) plane of the PEA_6SnI_8 phase. These results provide direct evidence confirming the presence of the PEA_6SnI_8 phase in the target films.

To evaluate the effects of excess PEAi on the optical properties of perovskite films, steady-state and transient PL spectroscopy were further conducted. Meanwhile, we characterized the films by using the PL spectroscopy. As seen in Fig. S8 in the ESM, both the control

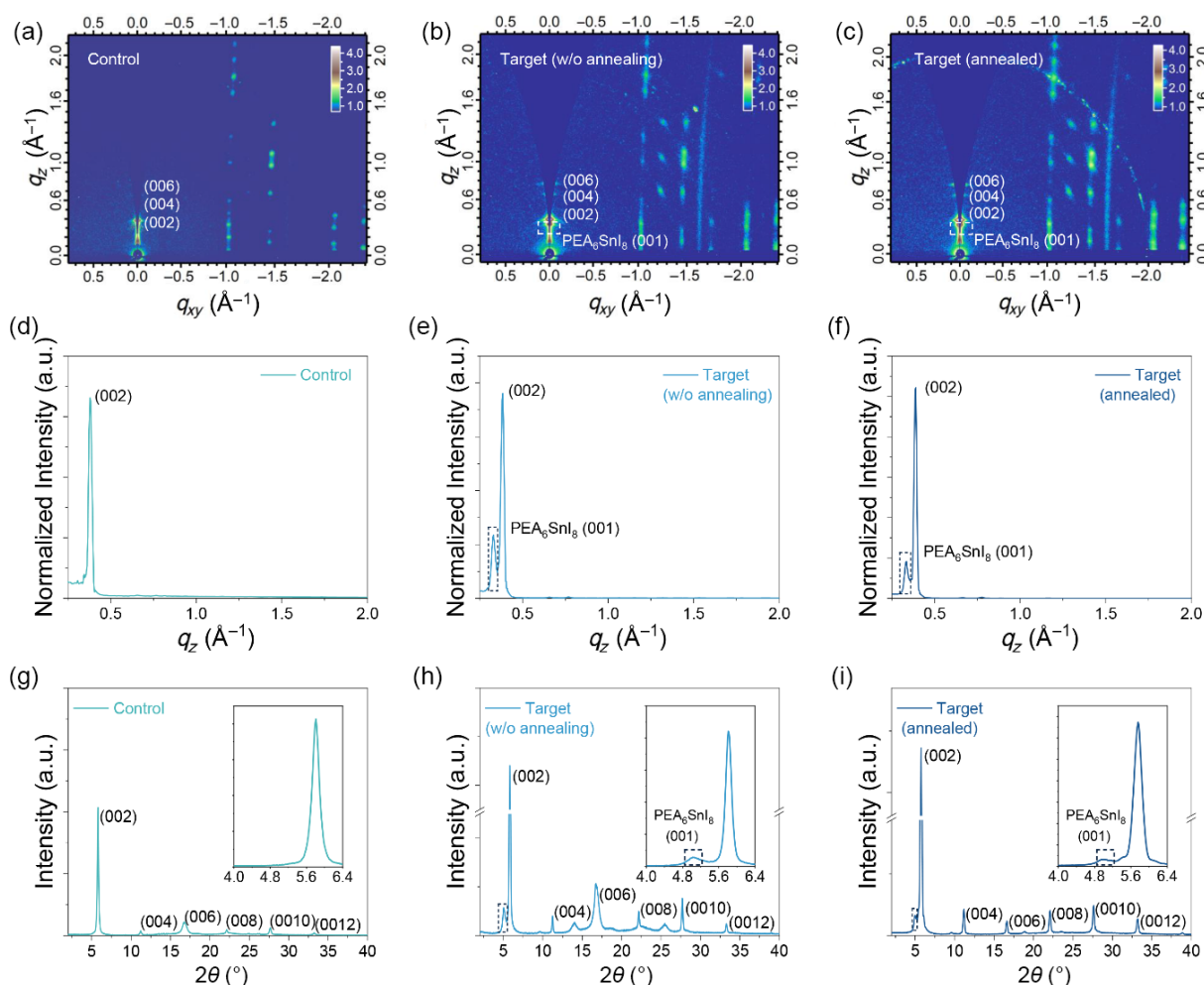


Figure 2 Verification of the existence of heterophase. GIWAXS patterns of the (a) control, (b) target (w/o annealing), and (c) target (annealed) perovskite films. Radially integrated GIWAXS patterns of the (d) control, (e) target (w/o annealing), and (f) target (annealed) perovskite films. XRD patterns of the (g) control, (h) target (w/o annealing), and (i) target (annealed) perovskite films.

and target films exhibited pure-red emission at 625 nm. However, compared to the control films, the targets film displayed a narrowed FWHM of 29 nm and a significantly enhanced luminescence intensity (Fig. S8(a) in the ESM). Correspondingly, its photoluminescence quantum yield (PLQY) increased from 6.1% to 31% (Fig. 3(a)). To gain more insights into the exciton recombination dynamics of PEA_2SnI_4 films, we performed time-resolved PL spectroscopy and fitted the obtained data using a biexponential decay model (Fig. S8(b) in the ESM). One can see that the target films exhibited an extended PL lifetime of 2.33 ns compared with the control one (1.02 ns). Concomitantly, the non-radiative recombination rate of the target films decreased from 9.21×10^8 to $2.96 \times 10^8 \text{ s}^{-1}$ (Table S2 in the ESM). These results demonstrate that the heterophase strategy effectively suppresses the non-radiative recombination in PEA_2SnI_4 films. Additionally, wavelength-dependent time-resolved PL spectroscopy with nanosecond-scale resolution was performed. As shown in Fig. S9 in the ESM, across the measured wavelength range of 600–650 nm, the target films exhibited an extended PL lifetime compared to the control films, indicating a significant suppressed non-radiative recombination. The surface morphologies of the PEA_2SnI_4 films were characterized by scanning electron microscopy (SEM) and atomic force microscopy (AFM) (Figs. 3(b) and 3(c)). Compared with the control films, the target films had a smoother and denser surface, which is conducive to reducing the leakage current

pathways. Correspondingly, the root-mean-square (RMS) roughness of the films decreased from 16.02 to 8.29 nm with the addition of excess PEA.

Additionally, we conducted temperature-dependent PL spectroscopy on both films. As shown in Fig. 3(d), the target films had significantly superior thermal quenching resistance compared to the control films. The Arrhenius plots of their integrated PL intensity were shown in Fig. 3(e), and the temperature quenching behavior can be well fitted by the following Eq. (1) [35]

$$I(T) = I_0 / [1 + A \cdot \exp(-E_b/k_B T)] \quad (1)$$

where I_0 denotes the integrated PL intensity at 0 K, and k_B is the Boltzmann constant, E_b is the exciton binding energy. Consequently, the E_b values were calculated to be 57.6 and 76.6 meV for the control and the target films, respectively. The increased E_b reduces the probability of exciton dissociation into free carriers and mitigates trap-mediated non-radiative recombination.

To evaluate the films stability, both films were exposed to ambient conditions, and their PL properties were monitored over time (Fig. S10 in the ESM). After aging in air for 90 min, the control films showed almost quenched luminescence under 365 nm UV irradiation, whereas the target films still exhibited strong PL emission, indicating a significantly improved stability. The degradation of PEA_2SnI_4 films in ambient conditions is primarily attributed to the oxidation of Sn^{2+} to Sn^{4+} . To quantify this stability,

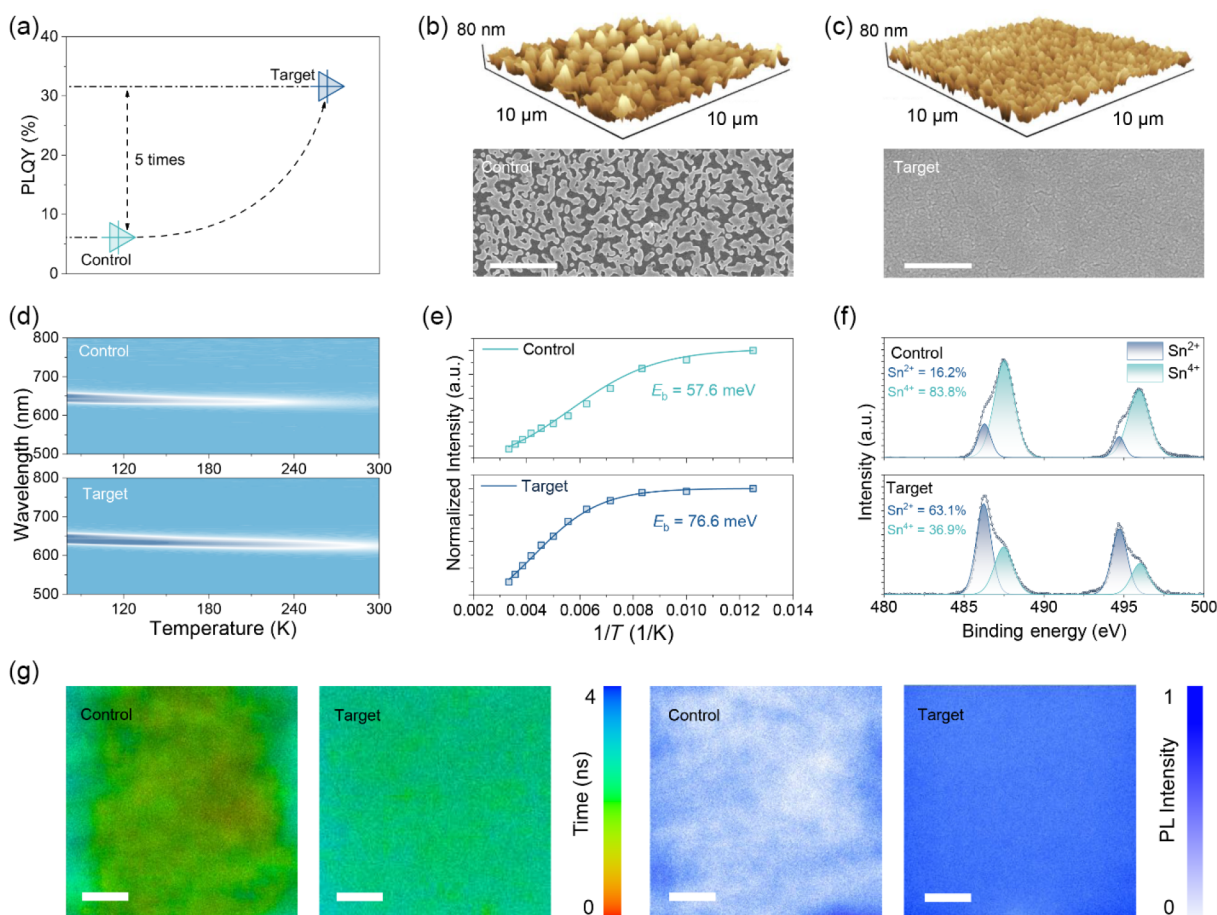


Figure 3 Optical properties of perovskite films. (a) PLQY comparison of the control and target perovskite films. SEM and AFM images of (b) control and (c) target perovskite films. Scale bar, 1 μm . (d) Pseudocolor map of temperature-dependent PL spectra of the control and target perovskite films. (e) Temperature-dependent PL intensity of the control and target perovskite films and fitting curves for E_b . (f) XPS spectra of Sn in control and target perovskite films. (g) PL mapping and time-resolved PL mapping of the control and target perovskite thin films. Scale bar, 5 μm .

we performed X-ray photoelectron spectroscopy (XPS) on both films after 90 min of air exposure. As shown in Fig. 3(f), the Sn^{4+} content in the control films was determined to be 83.8%, while that in the target films was only 36.9%, confirming the enhanced stability of the latter [36–38].

Since the oxidation of Sn^{2+} to Sn^{4+} is known to cause browning of the precursor solution, we exposed both precursor solutions to air and monitored their color changes [39]. After 16 h of air exposure, the precursor solution for the control films turned brown, whereas that for the target films remained relatively transparent (Fig. S11 in the ESM). Micro-area PL spectroscopy was used to evaluate the luminescence uniformity of two films. As shown in Fig. 3(g), the target films exhibited enhanced luminescence uniformity and intensity, along with an increased PL lifetime compared to the control films. To quantitatively compare the trap state density (N_t) of PEA_2SnI_4 films, we fabricated hole-only device with the structure of indium tin oxide (ITO)/poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate) (PEDOT:PSS)/ $\text{PEA}_2\text{SnI}_4/\text{MoO}_3/\text{LiF}/\text{Ag}$ and electron-only device with the structure of ITO/ $\text{ZnO}/\text{PEA}_2\text{SnI}_4/\text{TPBi}/\text{LiF}/\text{Al}$ by using space charge limit current (SCLC) method. Thus N_t could be determined by the trap-filled limit voltage (V_{TFL}) according to the following Eq. (2) [40]

$$N_t = 2\varepsilon\varepsilon_0 V_{\text{TFL}}/eL^2 \quad (2)$$

where ε (4.8) and ε_0 are relative permittivity of the PEA_2SnI_4 films and vacuum permittivity, L (50 nm) is the thickness of the PEA_2SnI_4 films, and e is the elementary charge. As shown in Fig. S12 in the ESM, after the introduction of excess PEA, the V_{TFL} decreased from 1.64 to 1.32 V; correspondingly, the trap state density drops from 1.64×10^{17} to $4.21 \times 10^{17} \text{ cm}^{-3}$. This significant improvement underscores the effectiveness of the 0D–2D heterophase strategy in suppressing defect-mediated recombination losses. Meanwhile, the target device exhibits more balanced carrier injection compared to the control device, and the more balanced carrier injection can be attributed to the improved film morphology, which reduces the formation of local over-injection of carriers [41, 42], as shown in Fig. S13 in the ESM.

To evaluate the EL performance of the optimized films, electrically driven LEDs with PEA_2SnI_4 as the emitters were fabricated. The device structure is ITO/PEDOT:PSS/LiF/perovskite/TPBi/LiF/Al. Figure 4(a) presents the energy band diagrams of all functional layers relative to the vacuum level, in which the energy levels of control and target films were determined via UV photoelectron spectroscopy (UPS) combined with UV/vis absorption spectroscopy (Fig. S14 in the ESM). Figure 4(b) shows the corresponding cross-sectional SEM image of the target device, displaying a compact framework and well-defined heterointerfaces. Figure 4(c) shows the EL spectra of the target LEDs under various applied biases. One can see that the EL spectra all centered at 628 nm with a FWHM of 30 nm. The EL intensity monotonously rises with the applied bias, suggesting that the undesirable nonradiative recombination or series resistance in the device would not increase proportionally. The inset of Fig. 4(e) displays the corresponding emission photo of the operated LED with the Commission International de l'Eclairage (CIE) color coordinates of (0.71, 0.30), which matches the Rec. 2020 specification for red emitters (Fig. S15(a) in the ESM). Under different bias voltages, the CIE coordinates of the target device hardly change, as seen in Fig. S15(b) in the ESM, confirming its stable operation.

In addition, we compared the transient EL behaviors of two

devices based on the control and target films. As shown in Fig. S16 in the ESM, under 3 V electrical pulses with a period of 200 μs , the target device exhibited a faster turn-on speed. When the pulse period was adjusted to 5 ms, it also demonstrated stable EL emission. This enhanced dynamic performance confirms the efficacy of the 0D–2D heterophase structure transition strategy in reducing the non-radiative recombination. Specifically, owing to the defect repair during the 0D–2D heterophasic structure transition and effective surface passivation by PEA, the performance of the target device has been significantly improved. Moreover, the maximum EQE and luminance of the target device reach 9% and 1253.3 cd/m^2 , representing a 12-fold and a 6-fold enhancement over the control device, respectively (Figs. 4(d) and 4(e)). Note that these results set a new benchmark for reported PEA_2SnI_4 -based LEDs (EQE > 8% and luminance > 1000 cd/m^2) (Fig. 4(f) and Table S3 in the ESM). Meanwhile, the target LED also show excellent reproducibility. The average EQE of 6.18% substantially surpasses that of the control device (0.9%) (Fig. S17 in the ESM). Furthermore, we measured the half-lifetime (T_{50}) of both devices. Under an initial luminance of 100 cd/m^2 , the T_{50} of the target device is 185 min, far exceeding that of the control device (32 min), as shown in Fig. S18(a) in the ESM. Subsequently, we captured the thermal mapping images of both devices during operation, as shown in Fig. S18(b) in the ESM. After 30 min of operation, the target device exhibits less heat generation compared to the control device, confirming the effectiveness of the heterophase strategy. Besides, we fabricated a large-area LED with a size of 1 cm^2 (Fig. S19 in the ESM), which exhibit bright and uniform emission when driven at 3.0 V. The large-area device achieves a maximum EQE of $\sim 4.1\%$ and a maximum luminance of $\sim 570 \text{ cd/m}^2$.

Benefiting from the low-voltage driving capability and large-area emission potential of our devices, we further explored their application as light sources in wearable medical devices (Fig. 4(g)). Currently, the mainstream principle of heart pulse detection involves transmitting light through the finger: by leveraging the absorption difference of hemoglobin at specific wavelengths, changes in the oxygen-carrying capacity of hemoglobin during cardiac pumping alter the light transmittance, which is captured by a sensor and converted into a pulse signal. Following the schematic in Fig. 4(g), when the encapsulated device was driven at 2.4 V, a stable electrical signal was detected by a commercial silicon detector (Fig. 4(h)). A corresponding demonstration video was provided in Movie S2 in the ESM. Through signal analysis, the diastolic and systolic phases of the pulse can be clearly distinguished (Fig. S20 in the ESM). A direct comparison was made with a measurement using a commercial LED under identical conditions. As shown in Fig. 4(i), the data acquired with our PEA_2SnI_4 LED show excellent agreement with the commercial LED reference, confirming the promising prospects of our devices in wearable medical applications.

3 Conclusions

In conclusion, we have demonstrated an 0D–2D heterophasic structure transition strategy for constructing high-quality PEA_2SnI_4 films enabling efficient pure-red LEDs. Experimental results reveal that excess PEA induces the formation of 0D PEA_6SnI_8 phase in the films, which slows down the crystallization rate, passivates defects, and enhances the PL properties of the resulting PEA_2SnI_4 films. Consequently, the obtained films not only exhibit a dense and uniform morphology but also possess excellent luminescent

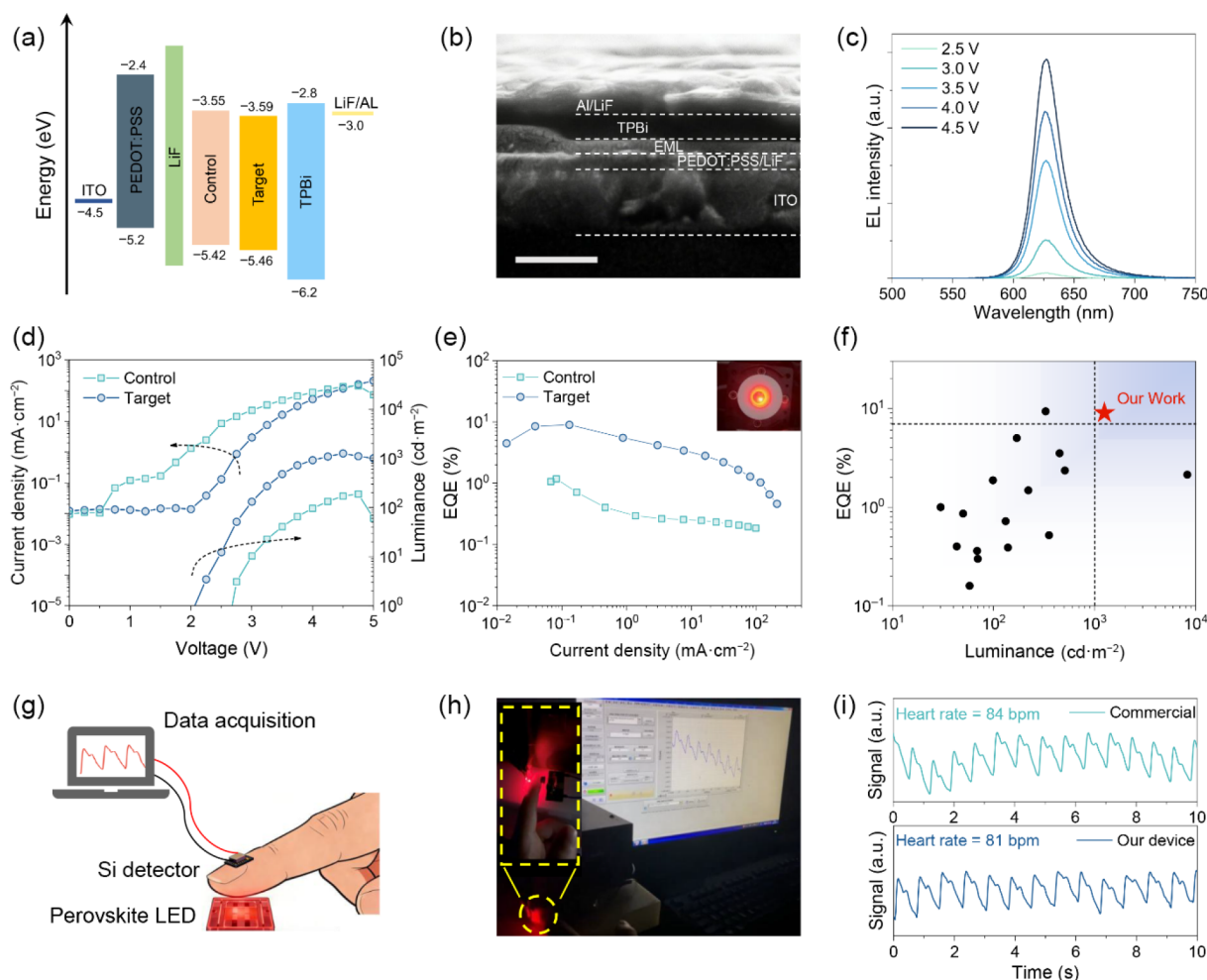


Figure 4 Performances of the target perovskite LEDs and their applications in human cardiac pulse detection. (a) Energy level diagram of the device. (b) Cross-sectional SEM image of the device. Scale bar, 200 nm. (c) EL spectra of the target device driven at different bias voltages. (d) Current density–voltage–luminance (J – V – L) curves of two devices. (e) EQE-current density curves of two devices. (f) Summary of the EQEs and luminance of the PEA_2SnI_4 LEDs reported to date. (g) Schematic diagram of a cardiac pulse detector based on perovskite LEDs. (h) Physical working demonstration of the proposed cardiac pulse detector. (i) Comparison of the cardiac pulse detection signals between commercial light sources and our perovskite LEDs.

performance and stability. Our results show that the tin-based perovskite LEDs fabricated via the 0D–2D heterophasic structure transition strategy achieve an EQE of 9% and a luminance of $1253.3 \text{ cd}\cdot\text{m}^{-2}$, ranking among the highest-performing PEA_2SnI_4 -based LEDs. Furthermore, this light source was successfully employed for human cardiac pulse detection by integrating with a commercial silicon detector, highlighting the future prospects of such LEDs for applications in wearable medical devices.

4 Experimental

4.1 Materials and preparation of perovskite precursor solution

LiF (99.99%), PEA₁ ($\geq 99.5\%$), PEDOT:PSS (1.3 wt. %–1.7 wt. % solution in water), and TPBi ($> 99\%$) were purchased from Xi'an Yuri Solar Co. Indium tin oxide (ITO) glass was purchased from Advanced Technology Co., Ltd. Chlorobenzene (CB, 99.8%) and dimethylsulfoxide (DMSO, 99.7%, SpcDry) were purchased from Sigma-Aldrich. SnI_2 (99.999%) was purchased from Shanghai Bide Pharmatech Ltd. Technology Co. All materials were used as received without further purification.

For control PEA_2SnI_4 precursor, PEA₁ and SnI_2 with a molar ratio of 2:1 were dissolved in pure DMSO. For target PEA_2SnI_4 precursor, PEA₁ and SnI_2 with a molar ratio of 2.4:1 were dissolved in pure DMSO. The concentration of SnI_2 was fixed at $0.1 \text{ mmol}\cdot\text{mL}^{-1}$. $10 \text{ mg}\cdot\text{mL}^{-1}$ tin powder was selectively added into the precursor solution to inhibit the oxidation of Sn^{2+} ions. The mixture solution was stirred at room temperature overnight in a nitrogen-filled glove box and filtered with $0.45 \mu\text{m}$ polytetrafluoroethylene (PTFE) filters before use.

4.2 Device fabrication and characterization

ITO-coated glass substrates were used as the anode and sequentially ultrasonically cleaned in deionized water, acetone, and ethanol for 15 min each. The cleaned substrates were treated with ultraviolet-ozone for 15 min to enhance the surface hydrophilic. Then, the PEDOT:PSS solution was spin-coated onto the substrates at 4000 rpm for 50 s, followed by annealing at 150°C in air for 20 min. The substrates were transferred to a vacuum evaporation chamber for the deposition of a 1 nm-thick LiF layer. Subsequently, the substrates were moved into a nitrogen-filled glovebox for the fabrication of the emissive layer. For the PEA_2SnI_4 emissive layer, the precursor solution was first spin-coated at 6000 rpm for 40 s. At

the 40th second, the spin speed was reduced to 3000 rpm, and 100 μL of CB was rapidly dropped onto the surface as an anti-solvent; spin-coating was continued until a total duration of 80 seconds was reached. The perovskite films were then annealed at 75 $^{\circ}\text{C}$ for 10 min. Finally, in the vacuum evaporation chamber, TPBi (40 nm), LiF (1 nm), and Al electrodes (100 nm) were deposited layer-by-layer through a shadow mask to complete the device fabrication. The effective area of the device was 4 mm^2 , defined by the overlapping area of the ITO anode and Al cathode.

The surface roughness and morphologies of the films were tested by AFM (Dimension Icon, Bruker Corporation) and SEM (Jeol-7500F, 15 keV), respectively. The XRD patterns were recorded by X'Pert Pro diffractometer. GIWAXs was measured by the Anhui GKINST, covering a Q-value range from 0.05 to 2.5 $\text{\AA}^{-1}$. For the preparation of TEM samples, prior to transferring the mixed-dimensional perovskites onto the TEM grid, a poly (methyl methacrylate) (PMMA) solution was spin-coated at 5000 rpm for 60 s, followed by baking at 80 $^{\circ}\text{C}$ for 10 min. Subsequently, the perovskite layer was deposited on the PMMA-coated substrate. To transfer the perovskite layer onto the TEM grid, chlorobenzene (CB) was drop-cast to dissolve the PMMA layer, thereby detaching the perovskite layer from the substrate. The resulting CB suspension containing the perovskite layer was then dropped onto a TEM grid (ultra-thin carbon film supported on a 200-mesh copper grid). Afterward, the TEM grid was kept under vacuum to eliminate residual solvent, and then it was ready for TEM imaging characterization. HRTEM imaging was performed using a TEM system (Tecnai G20) at an acceleration voltage of 200 kV. Absorption and PL spectra were measured with a Shimadzu UV-3600 spectrophotometer and a Horiba spectrofluorometer, respectively. The PLQY was measured by using a 405 nm LED as the excitation source and a QE Pro (Ocean optics) as the spectrometer with a fiber-coupled calibration integrating sphere. Time-resolved PL decay curves were measured with a fluorescence lifetime 3 measurement system. XPS and UPS results were obtained from the Shimadzu AXIS SUPRA. Micro-area luminescence intensity and lifetime mapping images were acquired using a micro-fluorescence spectrometer equipped with a Nikon T Plane 50 $\times$ objective and a 405 nm pulsed semiconductor laser. Temperature-dependent PL spectra were recorded at various temperatures controlled by a charge-coupled device (CCD) detector (Linkam THMS600 INSTRON and Ocean Optics, USB 2000+), with a heating/cooling rate of 20 $^{\circ}\text{C}$ per minute.

The devices fabrication was conducted at room temperature (26 $^{\circ}\text{C}$) inside a nitrogen-filled glovebox with controlled atmospheres ($\text{O}_2 \leq 1$ ppm, $\text{H}_2\text{O} \leq 1$ ppm). Electroluminescence (EL) spectra, current density–voltage–luminance (J – V – L) curves, and EQE were collected using an LED measurement system equipped with a digital source meter (Keithley 2400), an integrating sphere (GPS-4P-SL, Lab ball), and a coupled spectrometer (QE Pro, Ocean Optics Co., Ltd.). For the measurement of electrical signals related to heart pulse, a silicon photodetector (S1223-01, Hamamatsu Photonics) was used in combination with both a common commercial LED and the PEA_2SnI_4 -based LED.

Electronic Supplementary Material: Supplementary material (Figs. S1–S20: precursor ratio optimization, PL/TRPL characterization of the heterophase, film stability, SCLC and single-carrier device measurements, UPS spectra, device performance and cardiac pulse detection data; Tables S1–S3: PL lifetime data, optical properties, and performance summary of reported PEA_2SnI_4 LEDs; Movies S1

and S2: luminescence evolution during annealing and cardiac pulse detection demonstration) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908887>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

Y. L. Y.: Data curation (lead), formal analysis (supporting), investigation (equal), writing – original draft (equal), writing – review & editing (equal). X. Z. J.: Data curation (supporting), formal analysis (lead), investigation (equal), experimental design (equal), writing – original draft (equal), writing – review & editing (equal). S. L. L.: Data curation (supporting), investigation (equal), writing – review & editing (equal). M. W.: Investigation (equal). W. H. C.: Conceptualization (equal). Q. L. L.: Writing – review & editing (equal). N. N. W.: Data curation (supporting). Z. H. X.: Data curation (supporting). H. F. J.: Project administration, funding acquisition (equal). J. L. M.: Investigation (supporting). Z. Z. M.: Investigation (supporting). J. B. Z.: Investigation (supporting). X. J. L.: Writing – review & editing (supporting). Z. F. S.: Project administration (lead), funding acquisition (equal), writing – review & editing (equal).

Use of AI statement

None.

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