

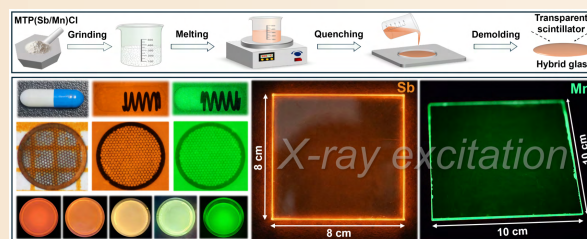
Transparent hybrid metal halide glassy scintillators for tunable multicolor and high-resolution X-ray imaging

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ABSTRACT: Current powder–polymer composite scintillator films typically require thicknesses of hundreds of micrometers to millimeters to ensure sufficient X-ray attenuation and brightness. However, intrinsic particle aggregation and interfacial heterogeneity inevitably induce severe light scattering, thereby compromising spatial resolution. Herein, we develop two optically homogeneous hybrid metal halide glasses, $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ (MTP = methyltriphenylphosphonium), via a scalable low-temperature melt quenching strategy. These glasses exhibit high visible–near-infrared transmittance up to ~90% and excellent glass-forming ability. At a thickness of 1 mm, they deliver near-complete attenuation of X-rays (> 99.8%) and intense radioluminescence with light yields of 5819 and 19,232 photons·MeV⁻¹, respectively. As a result, the glassy scintillators exhibit robust irradiation durability and achieve spatial resolutions of 18.8 and 22.5 lp·mm⁻¹, respectively, representing a substantial twofold improvement over previously reported crystalline counterparts. Impressively, they possess a stimulus-responsive reversible glass–crystal transition, low-temperature self-healing, and tunable radioluminescence from green to orange–red via compositional engineering. These features not only overcome the scattering and monochromatic-emission limitations of traditional scintillators but also establish a novel paradigm for next-generation recyclable materials and multicolor radiation visualization platforms.

KEYWORDS: metal halide glass; scintillator; X-ray imaging; radioluminescence; multicolor visualization



1 Introduction

X-ray imaging is indispensable in medical diagnostics, security screening, and industrial nondestructive testing, enabling noninvasive examination of internal structures [1–3]. Scintillators, which convert high-energy X-rays into visible light, play a crucial role in determining image quality, detection efficiency, and spatial resolution [4–7]. Currently, commercial inorganic scintillators such as $\text{Bi}_4\text{Ge}_3\text{O}_{12}$ (BGO), Ce-doped $\text{Lu}_3\text{Al}_5\text{O}_{12}$, LaBr_3 , and NaI have high X-ray stopping power and good radiation hardness [1,8,9]. However, these materials face significant challenges in terms of energy-intensive manufacturing processes at high temperature and high pressure, reliance on rare earth or toxic elements, and limited scalability with inherent rigidity. To overcome these limitations, organic–inorganic hybrid metal halides (OIMHs) have emerged as promising alternatives due to their tunable properties and ease of fabrication [9–12].

OIMHs combine the high X-ray attenuation capabilities of

heavy-metal halide units (Eu, Sb, Zr, Mn, Cu with I, Br, Cl) with flexible organic cations that offer tunable physical and chemical properties [4,13–16]. Among them, zero-dimensional (0D) OIMHs feature isolated inorganic units spatially separated by organic cations, which effectively suppress concentration quenching and enable efficient luminescence. In particular, Sb(III)- and Mn(II)-based OIMHs have gained attention due to their near-unity photoluminescence quantum yield (PLQY), superior solution processability, and environmental friendliness [14,17–19]. Despite these advantages, a critical bottleneck lies in translating these materials into large-area, high-performance scintillator screens. The current mainstream strategy involves embedding crystalline powders or nanocrystals into a polymer to produce composite scintillator films [20–22]. To ensure sufficient X-ray attenuation and scintillation brightness, the thickness of such composite films typically needs to reach several hundred micrometers or even a few millimeters. Unfortunately, this approach faces challenges such as particle agglomeration and

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nonuniform crystal distribution, often leading to significant light scattering and reduced optical transparency, generally limiting the spatial resolution to less than 12 lp-mm⁻¹ [23–25]. Importantly, these issues become more exacerbated with increasing film thickness, which severely restricts the practical application of polymer-based composite scintillators in high-resolution imaging [26,27]. Recent advances have demonstrated two representative morphology engineering strategies beyond conventional powder-polymer composite screens. One is the development of 0D hybrid metal halide glasses as bulk transparent scintillating media for large-area X-ray imaging, where the amorphous and optically homogeneous matrices suppress particle- and grain-boundary-induced light scattering while offering melt processability and potential recyclability [28,29]. The other is seed-crystal-induced cold sintering, which enables metal halide transparent ceramic scintillators through low-temperature densification [30]. Nevertheless, the limited number of metal centers leads to monochromatic luminescence (e.g., green from Mn²⁺ or yellow from Sb³⁺), which hinders the visualization of multicolor radiation detection [31,32]. Therefore, developing a new scintillator morphology that simultaneously offers high transparency, high homogeneity, tunable luminescence, and excellent stability is crucial for advancing the field toward practical applications.

Herein, we employ a low-temperature melt quenching strategy to develop a class of highly transparent hybrid metal halide glasses, MTP₂SbCl₅ and MTP₂MnCl₄ (MTP = methyltriphenylphosphonium). By eliminating the particulate aggregation and light scattering inherent to crystalline-polymer composite films, these glass-based scintillators achieve exceptional spatial resolutions of 18.8 and 22.5 lp-mm⁻¹, respectively—nearly double that of their recently reported crystalline composite counterparts, such as MTP₂SbCl₅ (10.2 lp-mm⁻¹) and MTP₂MnCl₄ (11.3 lp-mm⁻¹) [23,24]. We systematically characterize their structure, optical properties, scintillation performance, and imaging capabilities. These glasses exhibit high X-ray attenuation (100% for MTP₂SbCl₅ and 99.8% for MTP₂MnCl₄ at a thickness of 1 mm) and outstanding radiation stability. Furthermore, continuous multicolor radioluminescence from green to yellow and further to orange-red by simply adjusting the two-component glasses with different mass ratios is achieved, which enables the construction of high-quality visual radiation detection for medical imaging and security screening. With their special ability to change shape in response to different triggers and heal themselves at low temperatures, these hybrid glasses create a new standard for affordable, wide-area, and detailed X-ray imaging technologies.

2 Experimental

2.1 Preparation

Preparation of MTP₂SbCl₅ and MTP₂MnCl₄ single crystals: First, 1 mmol of MTPCl, 0.5 mmol of SbCl₃ (for MTP₂SbCl₅) and MnCl₂·4H₂O (for MTP₂MnCl₄), and 8 mL of N,N-dimethylformamide (DMF) were added to a 20 mL Teflon vessel and then placed in a muffle furnace. The furnace was heated at 100 °C for 2 h and then cooled to room temperature at a rate of 10 °C·h⁻¹. The resulting crystals were filtered, washed with ethanol, and dried overnight at 60 °C. MTPCl (97%), SbCl₃ (99.9%), MnCl₂·4H₂O (99.99%), and DMF (99.5%) were purchased from Macklin and used without further purification.

Preparation of MTP₂SbCl₅ and MTP₂MnCl₄ glasses: 10 mmol of MTPCl and 5 mmol of SbCl₃ (for MTP₂SbCl₅) and

MnCl₂·4H₂O (for MTP₂MnCl₄) precursors were added to a glass beaker and thermally processed under continuous agitation until they became a transparent molten material. The resulting melt is then cast into a preheated silicone mold and rapidly quenched to room temperature, yielding transparent glass. Multicolor-glass scintillators were prepared by adjusting the mass ratio of MTP₂SbCl₅ and MTP₂MnCl₄.

2.2 Characterizations

Single-crystal X-ray diffraction (XRD) data were measured using an Agilent Technologies Supernova diffractometer equipped with a graphite monochromator Mo Kα X-ray tube (λ = 0.71073 Å). Powder XRD measurements were conducted using a Rigaku SmartLab with a Cu Kα X-ray tube (λ = 1.5406 Å). Thermogravimetric analysis (TGA) was recorded using a HITACHI STA200 thermal analyzer. Differential scanning calorimetry (DSC) was measured on a Q2000 V24.11 Buid 124 instrument. X-ray photoelectron spectra (XPS) were recorded using an ESCALAB 250 spectrometer. Attenuated total reflectance Fourier transform infrared (ATR-FTIR) spectra were recorded on a Thermo Fisher Scientific Nicolet iS20 spectrometer. Energy-dispersive X-ray spectroscopy (EDS) and elemental mapping were conducted using a Hitachi SU 8010 under a voltage of 15 kV. Ultraviolet–visible absorption and transmittance spectra were measured using a UV2600 spectrophotometer. Steady-state photoluminescence (PL), photoluminescence excitation (PLE), radioluminescence (RL), time-resolved PL spectra, and decay curves were measured using an Edinburgh Instruments FLS1000 spectrofluorometer equipped with a continuous xenon lamp, a pulsed flash lamp, an X-ray tungsten target source, and a custom variable temperature system. PLQY was measured using a standard BaSO₄-coated integrating sphere as the sample chamber. All the recorded spectroscopic data were corrected for the spectral responses. Photographs of luminescence and X-ray imaging were taken using a Huawei Mate 80 Pro phone and a Nikon D850 camera without any filter.

2.3 Light yield and detection limit determination

Commercial BGO was used as a reference standard, which was measured as 10,000 photons·MeV⁻¹. All recorded RL spectra were corrected for the spectral responses of the detection system. To maintain data accuracy, transparent hybrid glasses of the same size were tested with BGO (10 mm in length, 10 mm in width, and 1 mm in thickness). All scintillators were measured at the same position under identical X-ray excitation and detection conditions. The emission photons of the scintillators were normalized to the same X-ray attenuation according to Eq. (1):

$$PC_{\text{normalized}} = \frac{PC_{\text{measured}}}{AE(\epsilon, d)} \quad (1)$$

where AE(ε, d) is the attenuation coefficient at the actual thickness. The corresponding photon counting (PC_{test}) is the integrated area of the RL spectra. The relative light yields (LY) of the prepared scintillator under X-ray excitation is defined as Eq. (2):

$$\begin{aligned} LY_{\text{sample}} &= LY_{\text{ref}} \times \frac{PC_{\text{normalized(sample)}}}{PC_{\text{normalized(ref)}}} \\ &= LY_{\text{ref}} \times \frac{PC_{\text{test(sample)}}}{AE_{\text{(sample)}}} \times \frac{AE_{\text{(ref)}}}{PC_{\text{test(ref)}}} \end{aligned} \quad (2)$$

where LY_{sample} and LY_{ref} are the light yields of the sample and reference, respectively. AE_{sample} and AE_{ref} are the attenuation coefficients of the sample and reference, respectively.

The detection limit (LOD) was calculated using the commonly adopted signal-to-noise criterion, as given by Eq. (3):

$$\text{LOD}(\sigma, n_{\text{slope}}) = \frac{3\sigma}{n_{\text{slope}}} \quad (3)$$

where σ is the standard deviation of the background signal measured without X-ray irradiation, and n_{slope} is the slope obtained from the linear fitting of the X-ray-excited luminescence (XEL) intensity as a function of the X-ray dose rate.

3 Results and discussion

Millimeter-sized single crystals of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ were synthesized via a hydrothermal method by reacting the organic salt MTPCl with inorganic precursors (SbCl_3 or MnCl_2) at a 2 : 1 molar ratio. Under daylight, the $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals appear white and pale yellow-green, respectively, whereas they exhibit intense yellow and green emissions under 365-nm ultraviolet (UV) light excitation (insets of Fig. 1). Single-crystal XRD analysis reveals that $\text{MTP}_2\text{SbCl}_5$ crystallizes in the monoclinic space group $P2_1/n$ with unit-cell parameters $a = 9.205 \text{ \AA}$, $b = 16.025 \text{ \AA}$, $c = 13.247 \text{ \AA}$, $V = 1906.9 \text{ \AA}^3$, and $Z = 2$, and $\text{MTP}_2\text{MnCl}_4$ belongs to the cubic $P2_13$ space group with $a = b = 15.603 \text{ \AA}$, $V = 3798.8 \text{ \AA}^3$, and $Z = 4$ (Fig. S1 and Table S1 in the Electronic Supplementary Material (ESM)). As illustrated in Figs. 1(a) and 1(b), both compounds show a characteristic 0D host-guest architecture, in which isolated inorganic polyhedra are fully separated by sterically bulky MTP^+ cations and periodically arranged. Figure 1 also displays the configurations of the MTP^+ , $[\text{SbCl}_5]^{2-}$, and $[\text{MnCl}_4]^{2-}$ units. Specifically, each Sb^{3+} ion is coordinated by five Cl^- ions to form a slightly distorted $[\text{SbCl}_5]^{2-}$ quadrangular pyramid [23,33,34], where the axial Cl^- site exhibits twofold disorder with an average occupancy of 1/2 (highlighted by red dashed circles). On the other

hand, each Mn^{2+} ion coordinates with four Cl^- ions to form a mildly distorted $[\text{MnCl}_4]^{2-}$ tetrahedron. The deviations in bond lengths ($\text{Sb}-\text{Cl}$ and $\text{Mn}-\text{Cl}$) and angles ($\text{Cl}-\text{Sb}-\text{Cl}$ and $\text{Cl}-\text{Mn}-\text{Cl}$) from the ideal geometries further corroborate the local polyhedral distortion (Table S2 in the ESM). Furthermore, the minimum inter-polyhedral distances exceed $9.205 \text{ \AA}$ for adjacent $[\text{SbCl}_5]^{2-}$ units and $10.009 \text{ \AA}$ for adjacent $[\text{MnCl}_4]^{2-}$ units (Fig. S2 in the ESM), which are significantly larger than the corresponding metal-halogen bond lengths. These results indicate negligible electronic interactions among inorganic moieties [18,35].

Transparent hybrid glasses were successfully fabricated via melt quenching using either as-synthesized $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals or their respective stoichiometric precursor mixtures. Specifically, the precursors were heated to a molten state and subsequently quenched into preheated silicone molds, yielding transparent glass bulks with controllable shape and thickness (Fig. 2(a)). These hybrid glasses exhibit excellent optical transparency across the visible to near-infrared range, with the Sb-based glass achieving a transmittance as high as 90%, as shown in Fig. 2(b). Powder XRD patterns of the glasses display the characteristic broad amorphous halos, confirming their noncrystalline nature (Fig. 2(c)). In contrast, the XRD patterns of the crystalline counterparts align well with the simulated results and remain distinct from the raw materials (Fig. 2(c) and Fig. S3 in the ESM), validating high phase purity and structural order. Generally, successful fabrication of glasses via melt quenching requires the melting temperature (T_m) to remain below the thermal decomposition temperature (T_d) [11,26,31]. Beyond this prerequisite, the ratio of the glass transition temperature (T_g) to T_m is widely used as a practical descriptor of glass-forming ability, with its magnitude reflecting to some extent the capacity to suppress recrystallization during cooling. According to the Kauzmann rule [10,36,37], a T_g/T_m ratio greater than 2/3 is

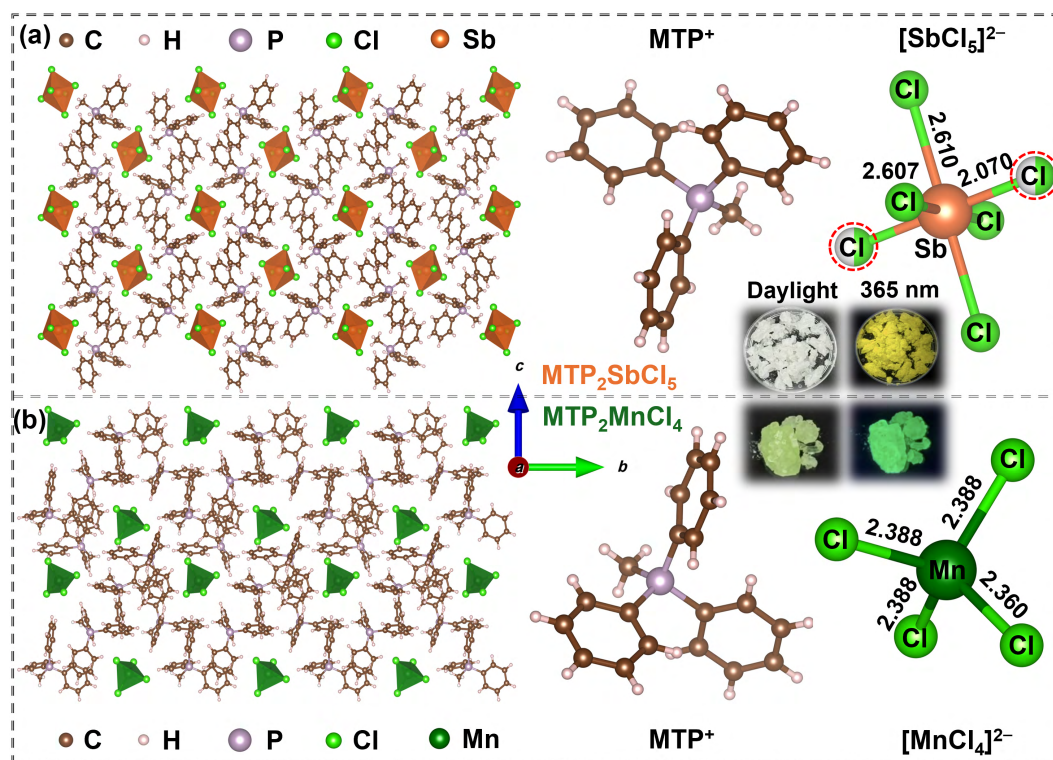


Fig. 1 Crystal structures of (a) $\text{MTP}_2\text{SbCl}_5$ and (b) $\text{MTP}_2\text{MnCl}_4$. Organic MTP^+ cations, inorganic $[\text{SbCl}_5]^{2-}$ quadrangular pyramids, and inorganic $[\text{MnCl}_4]^{2-}$ tetrahedra with key bond lengths (unit: $\text{\AA}$) are presented. Insets are optical images of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals under 365 nm light (top) and daylight (bottom).

generally considered to inhibit crystal nucleation, whereas a value below this threshold favors crystallization. TGA and DSC results (Figs. 2(d)–2(f)) reveal that the $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals possess T_d and T_m values of 268.4 and 220.5 and 309.1 and 214.7 °C, respectively. The second DSC heating scans further identify their T_g as 62.3 °C for $\text{MTP}_2\text{SbCl}_5$ and 66.7 °C for $\text{MTP}_2\text{MnCl}_4$ (Figs. 2(e) and 2(f)). The calculated T_g/T_m ratios of 0.68 and 0.70 further corroborate the strong glass-forming tendency of these systems and their capability to stabilize robust amorphous states.

The chemical composition and homogeneity of the glasses were comprehensively characterized by ATR-FTIR, EDS, elemental mapping, and XPS. As shown in Fig. 2(g), the ATR-FTIR spectra of both $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals and their corresponding glasses exhibit characteristic vibrational peaks, consistent with those of MTPCl , specifically the sp^3 -hybridized C–H bonds (450–1000 cm^{-1}) and C–H stretching vibrations of benzene rings ($\sim 3000 \text{ cm}^{-1}$) [11,38]. This consistency confirms the structural integrity of the organic cations and the negligibility of thermal degradation during the glass-forming process. Notably, the infrared absorption peaks of the glassy states exhibit discernible broadening compared with those of their crystalline counterparts (gray shaded areas in Fig. 2(g)), in line with an increased distribution of local coordination environments and enhanced structural disorder characteristic of amorphous states. High-resolution XPS and EDS spectra detected distinct signals of C, P, Cl, and Sb in $\text{MTP}_2\text{SbCl}_5$ glass and C, P, Cl, and Mn in $\text{MTP}_2\text{MnCl}_4$ glass (Fig. S4–S6 in the ESM). Elemental mapping further verified the homogeneous distribution of P, Cl, and Sb/Mn elements throughout the glass samples (Fig. S7 in the ESM). Collectively, these results confirm that the chemical composition

of the $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses comprises the expected organic cations and inorganic components.

Under 365-nm UV light excitation, the $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses exhibit red and green emission, respectively (Fig. 3(a)). Notably, a glass-to-crystal transformation can be deliberately triggered by external stimuli such as diethyl ether or mild thermal treatment (80–120 °C) (Fig. 3(a) and Figs. S8(a) and S8(c) in the ESM). Reapplying the melt quenching protocol described in Fig. 2(a) restores the amorphous state, enabling reversible glass–crystal–glass cycling. Beyond phase reversibility, these hybrid glasses also demonstrate excellent self-healing and reconfigurability after mechanical damage (Figs. S8(b) and S8(d) in the ESM), with healing temperatures ($< 210 \text{ °C}$) far below those required for conventional all-inorganic phosphate or silicate glasses (typically $> 1000 \text{ °C}$) [39,40]. These reversible phase transitions and low-temperature self-healing properties not only provide a sustainable strategy for the recycling and reuse of metal halide glasses but also greatly enhance the residual value of the materials after damage or end-of-life. It is noteworthy that the glassy and crystalline states of $\text{MTP}_2\text{SbCl}_5$ or $\text{MTP}_2\text{MnCl}_4$ exhibit distinct luminescence colors under UV light excitation, with the difference being particularly pronounced for the Sb-based system (yellow and red).

To elucidate the underlying mechanism governing the disparate luminescent properties between the crystalline and glassy states, we performed a series of optical characterizations. As shown in Fig. 3(b), the $\text{MTP}_2\text{SbCl}_5$ crystal exhibits broadband emission spanning from 470 to 850 nm under 365 nm excitation, with a dominant peak at 610 nm and a full width at half maximum (FWHM) of 129 nm. In stark contrast, the $\text{MTP}_2\text{SbCl}_5$ glass displays a significant redshift of the emission maximum to

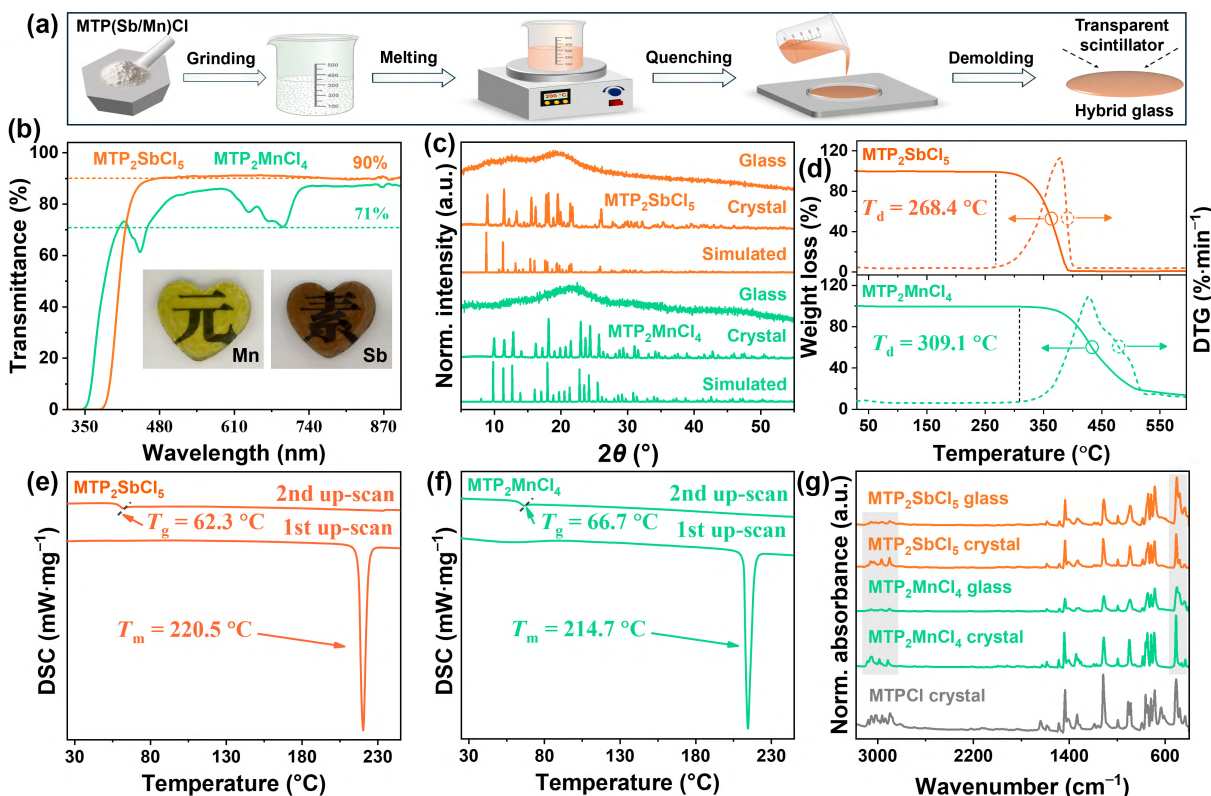


Fig. 2 (a) Preparation scheme of hybrid metal halide glass scintillator. (b) Transmittance curves of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses. Inset shows photographs of glasses under daylight. (c) XRD patterns of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ simulants, crystals, and glasses. (d) TG and DTG curves of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ crystals. First upscan and second upscan DSC curves in (e) $\text{MTP}_2\text{SbCl}_5$ and (f) $\text{MTP}_2\text{MnCl}_4$ crystals. (g) FTIR spectra of MTPCl crystal, $\text{MTP}_2\text{SbCl}_5$, and $\text{MTP}_2\text{MnCl}_4$ crystals and glasses.

650 nm, while the chromaticity coordinates change from (0.529, 0.459) to (0.581, 0.410) (Fig. S9 in the ESM) and the FWHM increases to 160 nm. Consistently, the excitation band, assigned to the $\text{Sb}^{3+} {}^1\text{S}_0 \rightarrow {}^3\text{P}_n$ transitions, shifts from 365 nm in the crystal to 397 nm in the glass [32,41], giving rise to a large Stokes shift of 2.09 eV for the glass (Fig. S10 in the ESM). In terms of efficiency and dynamics, the PLQY decreases from 99.4% in the crystal to 45.1% in the glass, and the corresponding decay lifetimes are 3.61 and 3.46 μs , respectively (Figs. 3(c) and 3(d)). Similarly, the $\text{MTP}_2\text{MnCl}_4$ crystal emits bright green luminescence under 450 nm excitation, peaking at 510 nm with a relatively narrow bandwidth (FWHM = 43 nm; Fig. 3(e)). Its glassy counterpart shows a red-shifted emission peak at 530 nm with a broadened FWHM of 69 nm and a change in chromaticity coordinates from (0.142, 0.662) to (0.296, 0.634) (Fig. S11 in the ESM). The PLE spectra exhibit a characteristic series of Mn^{2+} d–d transitions in the 320–500 nm range, which can be assigned to 318 nm (${}^6\text{A}_1 \rightarrow {}^4\text{T}_2(\text{F})$), 361 nm (${}^6\text{A}_1 \rightarrow {}^4\text{E}(\text{D})$), 382 nm (${}^6\text{A}_1 \rightarrow {}^4\text{T}_2(\text{D})$), 431 nm (${}^6\text{A}_1 \rightarrow {}^4\text{E}/{}^4\text{A}_1(\text{G})$), 446 nm (${}^6\text{A}_1 \rightarrow {}^4\text{T}_2(\text{G})$), and 467 nm (${}^6\text{A}_1 \rightarrow {}^4\text{T}_1(\text{G})$) [42,43]. These excitation peaks align well with the absorption peaks observed in the ultraviolet–visible absorption spectra (Fig. S12 in the ESM), and the Stokes shift of the $\text{MTP}_2\text{MnCl}_4$ glass is 1.57 eV. Quantitatively, the PLQY and lifetime changes from 93.8% and 2.10 ms in the crystal to 26.7% and 1.63 ms in the glass (Figs. 3(f) and 3(g)). Overall, the reduced PLQY and shortened lifetime in the glassy state point to enhanced nonradiative relaxation pathways enabled by increased structural disorder, which accelerates energy dissipation and suppresses radiative recombination.

To further pinpoint the origin of the broadband emission in

$\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses, we conducted excitation wavelength- and power-dependent steady-state spectroscopy and time-resolved PL measurements. As shown in Figs. 4(a) and 4(b), the excitation wavelength-dependent pseudocolor emission maps reveal that both glasses display a highly consistent emission response under different excitation wavelengths, suggesting that a common excitation pathway governs luminescence. This interpretation is reinforced by PLE spectra collected at different emission wavelengths, which exhibit nearly identical profiles and spectral features (Fig. S13 in the ESM), confirming that the broadband emission originates from the same excited state. Moreover, the linear dependence of the PL intensity on the excitation power (Fig. S14 in the ESM) rules out luminescence dominated by permanent defect or trap states and instead supports an intrinsic radiative process [44,45]. The time-resolved PL spectra acquired within time windows from 1.0 to 18.7 μs for $\text{MTP}_2\text{SbCl}_5$ glass and 0.1 to 9.3 ms for $\text{MTP}_2\text{MnCl}_4$ glass all exhibit consistent decay characteristics with minimal spectral reshaping (Figs. 4(c) and 4(d)). This phenomenon further supports that the radiative recombination process in each case stems from a single type of emission center. Considering that the broadband emission of $\text{MTP}_2\text{SbCl}_5$ glass is accompanied by a large Stokes shift and a millisecond-scale lifetime, it can be rationally assigned to self-trapped exciton (STE) emission in Sb-based halide systems [11,32]. In contrast, the microsecond-scale lifetime observed for $\text{MTP}_2\text{MnCl}_4$ glass is attributed to the spin-forbidden ${}^4\text{T}_1(\text{G}) \rightarrow {}^6\text{A}_1$ transition of the Mn^{2+} ion within $[\text{MnCl}_4]^{2-}$ tetrahedra [10,26].

To evaluate the optical thermal stability of the broadband emission, temperature-dependent PL spectra were measured from

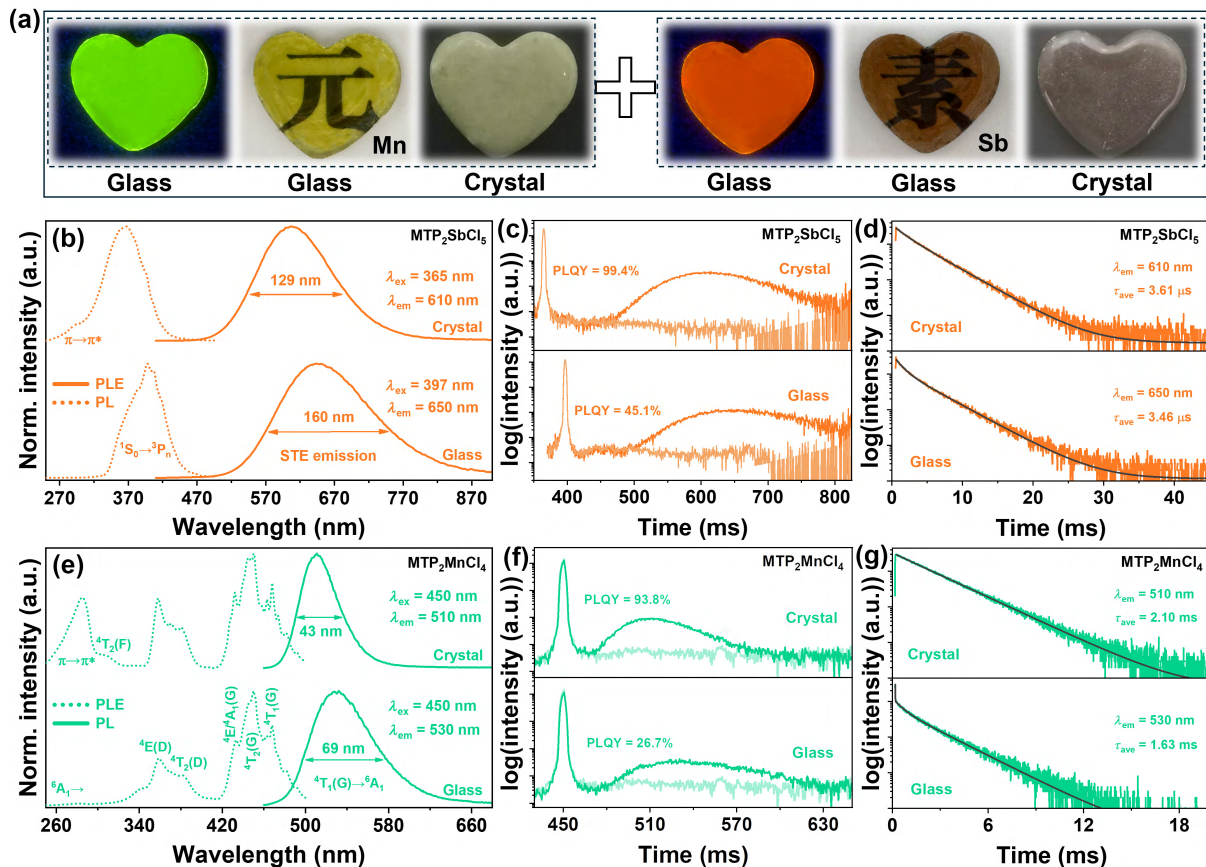


Fig. 3 (a) Photographs of $\text{MTP}_2\text{MnCl}_4$ and $\text{MTP}_2\text{SbCl}_5$ crystals and glasses under ultraviolet and daylight, respectively. (b) PLE/PL spectra, (c) PLQYs, and (d) PL decay curves of $\text{MTP}_2\text{SbCl}_5$ crystal and glass. (e) PLE/PL spectra, (f) PLQYs, and (g) PL decay curves of $\text{MTP}_2\text{MnCl}_4$ crystal and glass.

80 to 320 K. As shown in Figs. 4(e) and 4(f), the emission intensities of both glasses increase monotonically upon cooling, consistent with progressive suppression of nonradiative relaxation channels and enhanced contribution from radiative recombination at lower temperatures. Accordingly, the activation energy (E_b) can be calculated based on the Arrhenius equation (Eq. (4)) [46]:

$$I(T) = \frac{I_0}{1 + A \exp\left(-\frac{E_b}{k_B T}\right)} \quad (4)$$

where A , T , k_B , E_b , and $I(T)$ and I_0 represent the fitting constant, temperature, Boltzmann constant, activation energy, integrated PL intensities at different temperatures and 0 K, respectively. As shown in Fig. S15 in the ESM, the calculated E_b values for $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses are 66 and 159 meV, respectively. These results are higher than the thermal ionization energy at room temperature (~26 meV) [29,34], which facilitates efficient radiative recombination and overcomes thermal quenching, thereby supporting the excellent luminescent properties. Figure S16 in the ESM shows that the two individual emissions gradually broaden as the temperature increases. The electron-phonon couplings were evaluated by fitting the temperature-dependent FWHM of STE and Mn^{2+} emissions according to Eq. (5):

$$\text{FWHM} = 2.36\sqrt{S\hbar\omega_{\text{phonon}}}\sqrt{\coth\left(\frac{\hbar\omega_{\text{phonon}}}{2k_B T}\right)} \quad (5)$$

where S is the Huang–Rhys factor, and $\hbar\omega_{\text{phonon}}$ is the phonon energy. In this case, the fitting values of S and $\hbar\omega_{\text{phonon}}$ are 11.6 and 54.7 meV for STE recombination in $\text{MTP}_2\text{SbCl}_5$ glass and 6.0 and 5.1 meV for Mn^{2+} emission in $\text{MTP}_2\text{MnCl}_4$ glass, respectively (Fig. S17 in the ESM). Notably, the S values of glasses are higher than those of their crystalline counterparts (8.9 for $\text{MTP}_2\text{SbCl}_5$ and 4.4 for $\text{MTP}_2\text{MnCl}_4$, in Fig. S18 in the ESM) with a further enhanced electron-phonon coupling strength [29,32], which is

consistent with the structural transformation from crystalline order to amorphous disorder. This structural evolution rationalizes the concomitant decrease in luminescence efficiency, the expanded Stokes shift, and the redshifted emission.

Owing to their substantial deep-ultraviolet absorption, large Stokes shifts, and high optical transparency across the visible–near-infrared window, $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses exhibit considerable potential in scintillator applications, particularly for high-resolution X-ray imaging. Compared with conventional scintillator films composed of crystalline powders embedded in a polymer binder, transparent glass scintillators offer an amorphous and compositionally homogeneous matrix that minimizes grain-boundary- and particle-induced light scattering, thereby reducing optical loss and enhancing spatial resolution [10,26,27]. Using the National Institute of Standards and Technology XCOM database, the photon-energy-dependent X-ray absorption coefficients are presented in Fig. 5(a). Both $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ exhibit substantially stronger absorption than organic tetraphenylethylene (TPE)-based scintillators and approach that of commercial Si. Notably, $\text{MTP}_2\text{SbCl}_5$ even surpasses Si within the 10–120 kV range, which is most relevant to medical diagnostics and security screening. The incorporation of high- Z elements (Sb and Mn) markedly enhances X-ray attenuation relative to pure MTPCl . The X-ray attenuation efficiency (AE) was determined using Eq. (6) [47]:

$$\text{AE}(\varepsilon, d) = (1 - e^{-c(\varepsilon)\rho d}) \times 100\% \quad (6)$$

where $c(\varepsilon)$ is the photon cross-section function obtained from the XCOM database and ε , ρ , and d are the photon energy, density, and thickness, respectively. In this case, the densities of $\text{MTP}_2\text{SbCl}_5$ (1.487 g·cm⁻³) and $\text{MTP}_2\text{MnCl}_4$ (1.314 g·cm⁻³) are derived based on crystallographic information. Notably, increasing thickness improves absorption but simultaneously reduces transmittance and can degrade imaging sharpness [33], ultimately compromising spatial resolution. Here, we optimized the screen thickness to 1 mm. At this thickness, the $\text{MTP}_2\text{SbCl}_5$ and

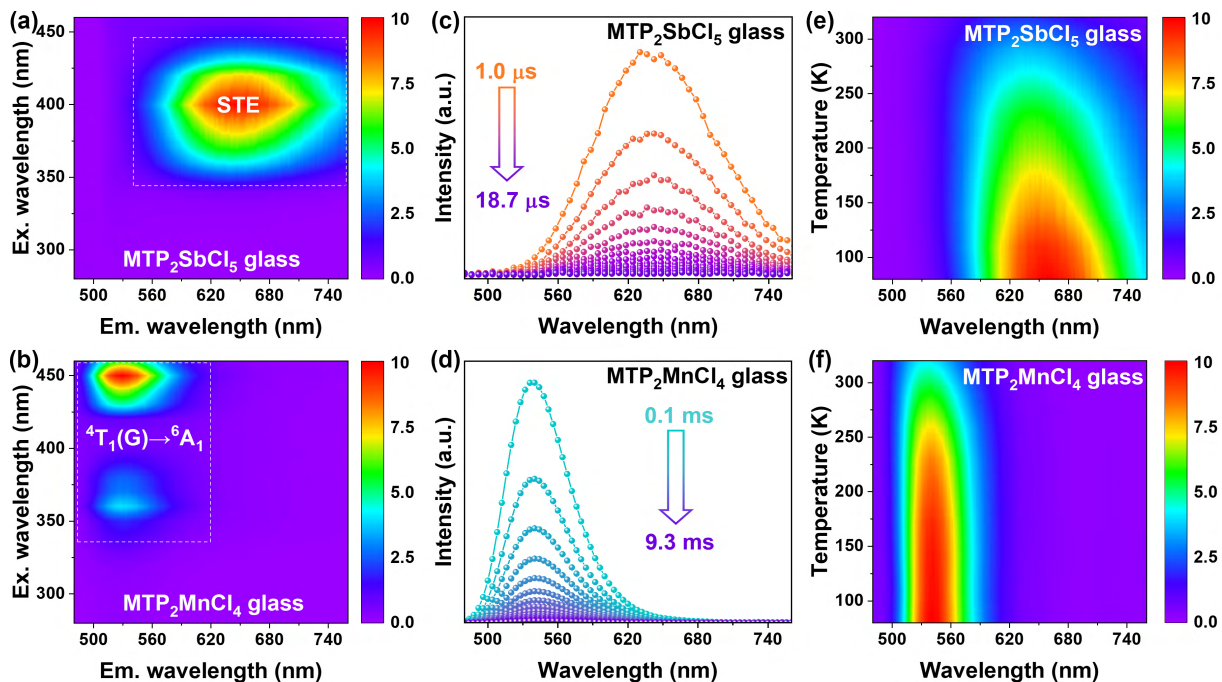


Fig. 4 Excitation wavelength-dependent PL spectra of (a) $\text{MTP}_2\text{SbCl}_5$ and (b) $\text{MTP}_2\text{MnCl}_4$ glasses. (c) Time-resolved PL spectra of $\text{MTP}_2\text{SbCl}_5$ glass from 1.0 to 18.7 μs . (d) Time-resolved PL spectra of $\text{MTP}_2\text{MnCl}_4$ glass from 0.1 to 9.3 ms. Temperature-dependent PL spectra of (e) $\text{MTP}_2\text{SbCl}_5$ and (f) $\text{MTP}_2\text{MnCl}_4$ glasses.

MTP₂MnCl₄ glasses attenuate 8-keV incident X-rays (tungsten-target energy) by 100% and 99.8%, respectively (Fig. 5(b)). The RL spectra of MTP₂SbCl₅ and MTP₂MnCl₄ glasses under X-ray excitation closely match their PL spectra obtained under ultraviolet excitation (Fig. S19 in the ESM), confirming the same radiative recombination pathways. At identical X-ray dose rates, the MTP₂MnCl₄ glass exhibits the highest XEL intensity, whereas the MTP₂SbCl₅ glass is only slightly weaker than the commercial BGO (Fig. 5(c)). Using BGO as a reference, the estimated light yields for MTP₂SbCl₅ and MTP₂MnCl₄ glasses are 5819 and 19,232 photons·MeV⁻¹, respectively. Remarkably, the light yield of MTP₂MnCl₄ glass exceeds that of commercial scintillators such as YAG:Ce (16,400 photons·MeV⁻¹) and CdWO₄ (13,000 photons·MeV⁻¹) [18]. While the transition from crystal to glass increases structural disorder and introduces nonradiative channels, leading to a reduction in PLQY, the resulting hybrid glasses maintain competitive radioluminescence performance. This is primarily attributed to their exceptional optical transparency (~90%), which substantially enhances light extraction efficiency compared with scattering-prone crystalline composites. Furthermore, the dense bulk nature of the glasses ensures efficient X-ray attenuation and energy deposition, while the isolated 0D inorganic units prevent severe luminescence quenching, collectively preserving high light yield and superior imaging resolution. Furthermore, the XEL intensity of both glasses increases progressively with increasing X-ray dose rate (Fig. S20 in the ESM). It maintains an excellent response to the dose rate (Fig. S21 in the ESM), which is advantageous for achieving high-contrast X-ray imaging. The detection limits of MTP₂SbCl₅ and MTP₂MnCl₄ glasses were 6.2 and 6.8 μGy_{air}·s⁻¹, respectively, which are comparable to typical values encountered in routine medical examinations (5.0 μGy_{air}·s⁻¹) [48,49].

Furthermore, the radiation stability of MTP₂SbCl₅ and MTP₂MnCl₄ glasses under high X-ray dose exposure, continuous irradiation, and on-off cycling tests was evaluated. As shown in Fig. 5(d), the XEL intensities of both glasses show no obvious degradation over 72 consecutive switching cycles. After continuous X-ray irradiation with a cumulative dose of 408.6 Gy, MTP₂SbCl₅ and MTP₂MnCl₄ glasses retain 98% and 95% of their initial intensity, respectively (Fig. 5(e)), underscoring excellent operational durability and resistance to radiation-induced photodegradation. As shown in Fig. S22 in the ESM, temperature-dependent RL measurements were further performed to compare the thermal quenching behavior under X-ray excitation with that under UV excitation. The RL activation energies were fitted to be 76.5 meV for MTP₂SbCl₅ glass and 188 meV for MTP₂MnCl₄ glass, which are close to the PL-derived values of 66 and 159 meV, respectively. This consistency indicates that PL and RL share similar thermal quenching pathways and originate from the same emissive centers. The functional stability after recycling and self-healing was also evaluated. After three crystal-glass cycles, both glasses retained nearly unchanged XEL intensity (Figs. S23(a) and S23(b) in the ESM). The self-healed samples obtained by heating fractured glass pieces at 200 °C for 5 min also preserved more than 98% of their original XEL intensity without an obvious spectral shift, confirming their good scintillation durability after recycling and repairing (Figs. S23(c) and S23(d) in the ESM). We next benchmarked their imaging performance using a custom-built X-ray imaging platform (Fig. S24 in the ESM). Spatial resolution was evaluated with a standard lead line-pair phantom using 1-mm-thick glass scintillation screens. The two glasses were measured using the same imaging platform, sample position, camera settings, exposure time, and MTF analysis method, while the X-ray excitation parameters were adjusted according to their

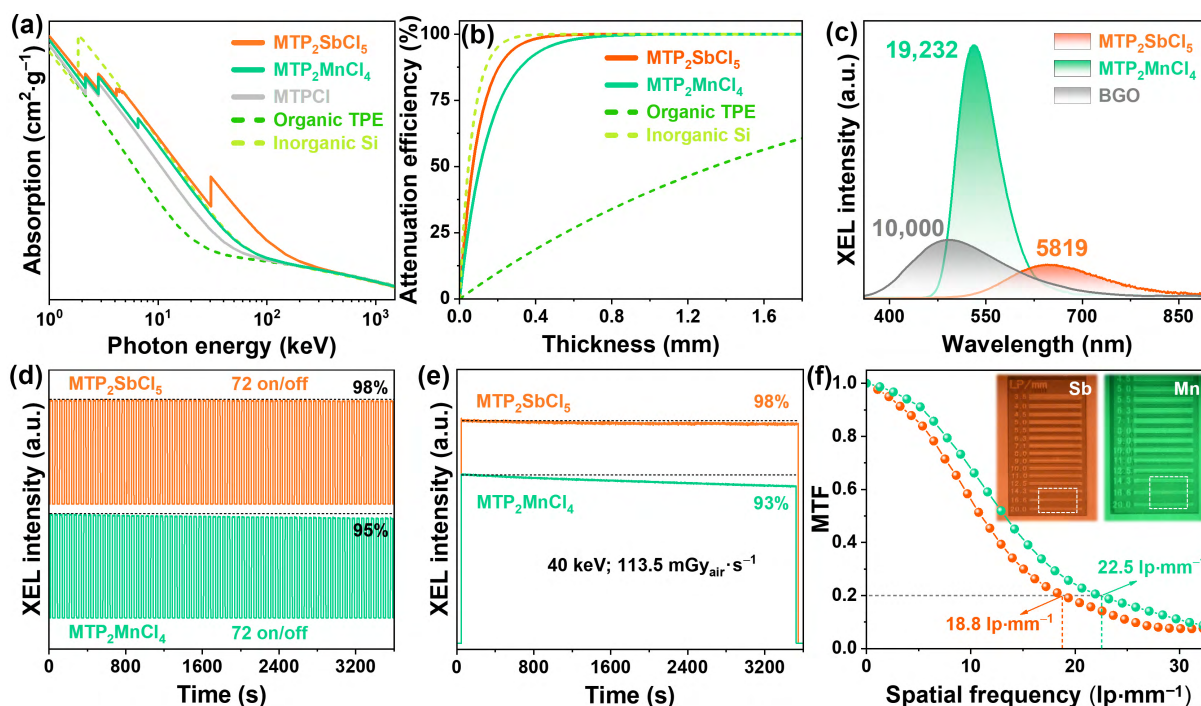


Fig. 5 (a) X-ray absorption coefficients of inorganic Si, organic TPE, MTPCl, MTP₂SbCl₅, and MTP₂MnCl₄ as functions of photon energy. (b) X-ray attenuation efficiencies of inorganic Si, organic TPE, MTP₂SbCl₅, and MTP₂MnCl₄ as a function of film thickness. (c) Radioluminescence spectra of BGO crystal, MTP₂SbCl₅ glass, and MTP₂MnCl₄ glass (dose rate: 71.1 mGy·s⁻¹, voltage: 30 kV). (d) XEL intensity of MTP₂SbCl₅ and MTP₂MnCl₄ glasses recorded over 72 continuous on-off cycles (dose rate: 113.5 mGy·s⁻¹, voltage: 40 kV). (e) XEL intensity of MTP₂SbCl₅ and MTP₂MnCl₄ glasses under X-ray irradiation (dose rate: 113.5 mGy·s⁻¹, voltage: 40 kV). (f) modulation transfer function (MTF) curves of MTP₂SbCl₅ and MTP₂MnCl₄ glasses. Insets show spatial resolution of corresponding glass scintillators, as determined by lead wire pair measurements.

emission intensity and imaging contrast: 50 kV and 34.8 $\text{mGy}_{\text{air}}\cdot\text{s}^{-1}$ for $\text{MTP}_2\text{SbCl}_5$ glass and 30 kV and 11.8 $\text{mGy}_{\text{air}}\cdot\text{s}^{-1}$ for $\text{MTP}_2\text{MnCl}_4$ glass. Clear separation of the interline gaps remains visible at 20 $\text{lp}\cdot\text{mm}^{-1}$, and the corresponding gray-value profiles further substantiate this resolving capability (Fig. S25 in the ESM). Quantitative MTF analysis, adopting an MTF value of 0.2 as the criterion, yields spatial resolutions of 18.8 $\text{lp}\cdot\text{mm}^{-1}$ for $\text{MTP}_2\text{SbCl}_5$ glass and 22.5 $\text{lp}\cdot\text{mm}^{-1}$ for $\text{MTP}_2\text{MnCl}_4$ glass (Fig. 5(f) and Fig. S26 in the ESM). These values markedly outperform those of previously reported crystal–polymer composite scintillator films, including $\text{MTP}_2\text{SbCl}_5$ (10.2 $\text{lp}\cdot\text{mm}^{-1}$) [23], $\text{C}_{38}\text{H}_{36}\text{P}_2\text{Sb}_2\text{Cl}_8$ (8.15 $\text{lp}\cdot\text{mm}^{-1}$) [14], $(\text{EOTP})_2\text{MnBr}_4$ (10.28 $\text{lp}\cdot\text{mm}^{-1}$) [50], and $\text{MTP}_2\text{MnCl}_4$ (11.3 $\text{lp}\cdot\text{mm}^{-1}$) [24], highlighting the intrinsic advantage of optically transparent glass scintillators for high-resolution X-ray imaging. To provide a clear overview of the advantages of the present glass scintillators, we further compared the key performance metrics with representative reported scintillators, as summarized in Table S3 in the ESM. The $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses combine low-temperature melt quenching preparation, high optical transmittance, competitive light yields, and high spatial resolutions within one material system. This balanced performance, together with their large-area processability, reversible glass–crystal transition, and self-healing capability, further highlights the advantages of transparent hybrid metal halide glasses for practical X-ray imaging.

To demonstrate the practical utility of $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses for X-ray imaging, we imaged representative objects, including a metallic spring sealed inside a capsule and a standard 200-mesh copper grid. As shown in Fig. 6(a), the spring

geometry is revealed, attesting to the excellent contrast and structural delineation enabled by these glass scintillation screens. Remarkably, the copper grid, originally designed for transmission electron microscopy and featuring line widths of only $\sim 37\ \mu\text{m}$, remains clearly distinguishable (Fig. 6(b)), highlighting a near-microscale resolving capability. Beyond imaging performance, these glass scintillators offer compelling engineering and commercial advantages. They can be rapidly fabricated via low-temperature melt quenching and readily scaled to large-area formats, reaching 8 cm $\times$ 8 cm and even 10 cm $\times$ 10 cm (Figs. 6(c) and 6(d)). Impressively, by tuning the two-component glasses with different mass ratios, the RL color of the hybrid glass can be continuously modulated from green to yellow and then to orange–red (Fig. S27 in the ESM and Fig. 6(e)), thereby overcoming a key limitation of conventional scintillators for multicolor visualization in radiation detection. The multicolor radioluminescence in the mixed-component glasses is achieved through homogeneous blending in the molten state rather than physical mixing of crystalline powders. Benefiting from the high homogeneity of the liquid phase and the subsequent rapid quenching process, the resulting glassy plates show spatially uniform RL emission without obvious phase separation or local color variation. Moreover, the emission color can be well reproduced across different synthesis batches by maintaining consistent precursor weight ratios and identical melt quenching conditions. These features support the scalable fabrication of large-area, color-tunable glass scintillator screens for visual radiation detection. Moreover, spatially splicing regions with distinct compositions enable a high-quality visual-radiation-detection interface with immediate relevance to medical imaging and

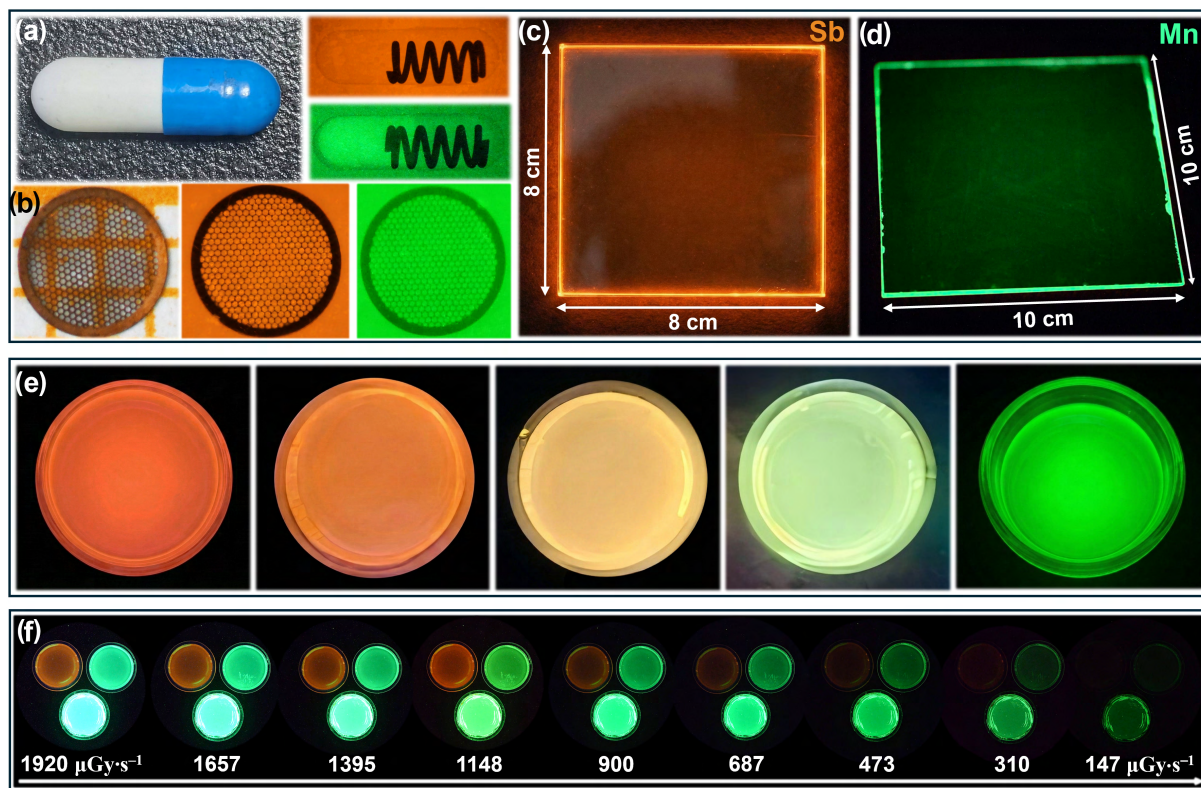


Fig. 6 Physical photographs and corresponding X-ray images of (a) a metallic spring sealed in a capsule and (b) a 200-mesh copper grid obtained using a 1-mm-thick glass scintillator screen. Transparent glasses with a large size of (c) 8 cm $\times$ 8 cm ($\text{MTP}_2\text{SbCl}_5$) and (d) 10 cm $\times$ 10 cm ($\text{MTP}_2\text{MnCl}_4$) under X-ray irradiation. (e) RL images of $\text{MTP}_2\text{SbCl}_5$ – $\text{MTP}_2\text{MnCl}_4$ hybrid glasses by adjusting the two-component glasses with different mass ratios of 1 : 0, 150 : 1, 90 : 1, 40 : 1, and 0 : 1. (f) RL images of color glass scintillators under different X-ray doses.

security screening. As illustrated in Fig. 6(f), when the dose rate decreases from 1920 to 147 $\mu\text{Gy}\cdot\text{s}^{-1}$, the splicing pattern of two-component glasses with different mass ratios can easily produce distinguishable changes in luminescence color, thereby enabling real-time and intuitive radiation warning and identification. This visual radiation detection modality operates without an external power source, underscoring its practical deployability.

4 Conclusions

In summary, we have successfully developed a novel class of highly transparent, 0D organic–inorganic hybrid $\text{MTP}_2\text{SbCl}_5$ and $\text{MTP}_2\text{MnCl}_4$ glasses that unify melt processability, recyclability, and high-performance scintillation. Centimeter-scale screens can be rapidly fabricated by cost-effective and scalable low-temperature melt quenching from either crystals or precursor mixtures, delivering high visible–near-infrared transparency (up to $\sim 90\%$ for Sb-based glass) and stable glass-forming characteristics ($T_g/T_m = 0.68$ and 0.70). At a thickness of 1 mm, the glasses provide near-complete X-ray attenuation (100% for $\text{MTP}_2\text{SbCl}_5$ and 99.8% for $\text{MTP}_2\text{MnCl}_4$) due to the high X-ray absorption of heavy atoms while exhibiting strong radioluminescence with light yields of 5819 and 19,232 photons·MeV $^{-1}$, respectively. Benefiting from effective light-scattering suppression in the amorphous state, these glasses achieved spatial resolutions of 18.8 and 22.5 lp·mm $^{-1}$, respectively. Moreover, we demonstrated multicolor radioluminescence tuning from green to orange–red via compositional engineering and constructed a regionally assembled, color-coded dose visualization, offering a scalable route toward high-resolution imaging and intuitive radiation detection beyond conventional powder–polymer composite scintillator films. This work not only expands the material library of metal halide glasses but also establishes a new paradigm for the development of low-cost, large-area, high-resolution, and self-healable X-ray imaging.

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Author contributions

Qixin Huang and Shilin Jin synthesized material; Qixin Huang and Shilin Jin measured the photophysical properties of the samples, X-ray scintillators, and X-ray imaging; Luyao Wei, Yuehua Chen, Huaixi Chen, and Lingwei Zeng assisted in the material characterization; Shilin Jin designed experiments and wrote the manuscript; Daqin Chen helped to analyze the experimental results, supervised this project, and revised the manuscript.

Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Conflict of interests

The authors have no competing interests to declare that are relevant to the content of this article. The author Daqin Chen is the Editorial Committee member of this journal.

Electronic Supplementary Material

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