

Rare-earth halide perovskite-like scintillators for X-ray detection and imaging

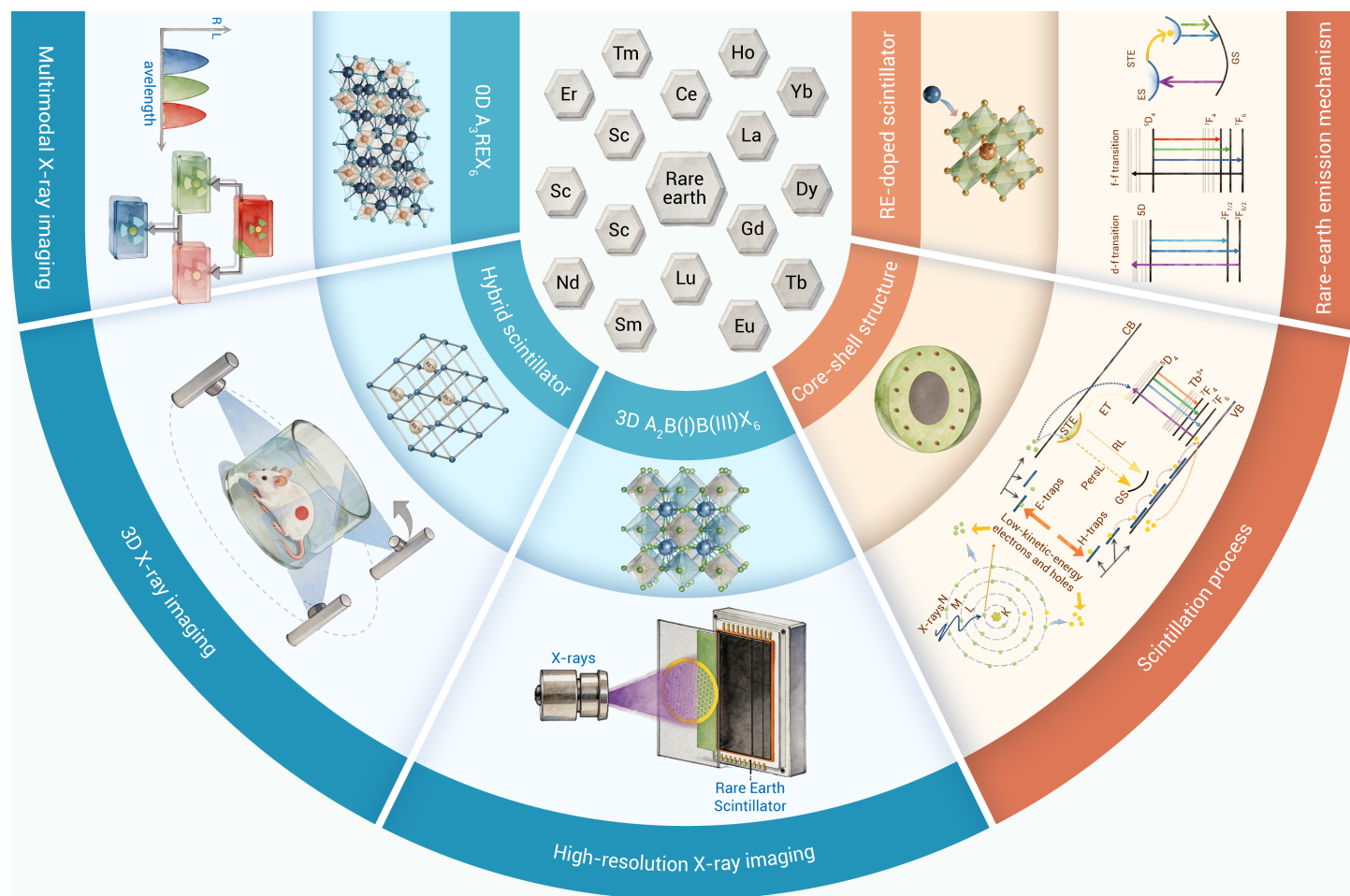
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Received: December 28, 2025; Accepted: July 13, 2026; Published Online: July 13, 2026; <https://doi.org/10.59717/j.xinn-mater.2026.100227>

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GRAPHICAL ABSTRACT



PUBLIC SUMMARY

- Rare-earth halide perovskite-like scintillators enable efficient and stable X-ray detection and imaging.
- This review outlines scintillation mechanisms and recent advances in inorganic, hybrid, and doped systems.
- These materials enable low-dose detection, high-resolution and dynamic imaging, and multimodal applications.

Rare-earth halide perovskite-like scintillators for X-ray detection and imaging

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Received: December 28, 2025; Accepted: July 13, 2026; Published Online: July 13, 2026; <https://doi.org/10.59717/j.xinn-mater.2026.100227>

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Citation: Wang N., Li B., Zhao Y., et al. (2026). Rare-earth halide perovskite-like scintillators for X-ray detection and imaging. *The Innovation Materials* 4:100227.

Scintillator-based X-ray detection technologies play an irreplaceable role in national security, medical diagnostics, and industrial non-destructive testing due to their broad dynamic response range, large effective imaging area, low maintenance requirements, and high operational reliability. In recent years, metal halide emitters with perovskite or perovskite-like structures have attracted extensive attention, among which rare-earth halides have shown remarkable scintillation characteristics, including high radiation conversion efficiency, tunable emission spectra, and excellent stability. These advantages arise from their structural diversity and rich photophysical properties, positioning them as strong candidates for next-generation scintillators. In this review, we provide a comprehensive overview of recent advances in rare-earth halide scintillators for X-ray detection and imaging. We first clarify the fundamental physical mechanisms governing the interaction between X-rays and rare-earth halide scintillators, along with the complete scintillation process. We then systematically summarize emerging classes of rare-earth halide scintillation materials, including fully inorganic compounds, organic-inorganic hybrids, and rare-earth-ion-doped systems, with emphasis on their design principles and strategies for modulating scintillation performance. In addition, we highlight the potential of these materials in high-resolution imaging, multimodal imaging, and X-ray photodynamic therapy. Finally, we identify the key challenges that currently constrain progress in this field and propose future research directions aimed at advancing rare-earth halide scintillator technologies.

INTRODUCTION

X-ray detection technologies play a pivotal role in homeland security, medical diagnostics, industrial inspection, and scientific research owing to their strong penetration capability, rapid response, and non-contact operation.^{1–3} According to the interaction mechanisms between X-rays and detecting media, X-ray detection technologies can be broadly classified into direct and indirect detection modes.⁴ In direct detection, a one-step energy conversion process is employed, in which high-quality single crystal or thick film semiconductor materials (such as Si, CdTe, and CdZnTe) serve as the detection medium.^{5–7} Upon X-ray irradiation, incident photons are directly absorbed by the semiconductor, generating electron-hole pairs. Under an externally applied electric field, these charge carriers are efficiently separated and transported toward the corresponding electrodes, producing a measurable electrical signal. To ensure high charge generation efficiency and long carrier transport lifetimes, semiconductor materials used for direct detection generally require high crystalline purity, excellent structural integrity, and a low density of defect states.^{8,9} These stringent material requirements are essential for suppressing carrier recombination and trap-assisted capture, which would otherwise lead to signal attenuation and degradation of spatial or energy resolution.¹⁰ By contrast, indirect X-ray detection relies on a two-step signal conversion mechanism. In this approach, scintillators act as energy conversion media that efficiently transform high-energy X-ray photons into ultraviolet or visible photons. These lower-energy photons are subsequently collected by photodetectors, such as silicon photodiodes or complementary metal-oxide-semiconductor sensors, and converted into readable electrical signals. Compared with the demanding requirements for high-quality semiconductor single crystals or thick films in direct detection, scintillators used in indirect detection exhibit greater tolerance with respect to material quality and processing conditions, offering significant advantages in terms of cost efficiency, scalability, and suitability for large-area and practical

applications.^{11–14}

In the 1940s, the scintillation effects of NaI:Tl and CsI:Tl were successfully developed, establishing these materials as cornerstones for early radiation detection and nuclear medical imaging.¹⁵ Subsequently, Bi₄Ge₃O₁₂ (BGO) crystals were widely adopted in high-energy radiation detection, owing to their high effective atomic number and excellent chemical stability.¹⁶ Since the 1990s, rare-earth-based oxide scintillator materials, represented by Lu_{1.8}Y_{0.2}SiO₅:Ce (LYSO), Gd₂SiO₅:Ce (GSO), and Gd₂O₂S:Tb/Pr (GOS), have been extensively developed.^{17–19} Compared with BGO, LYSO, and GOS simultaneously exhibit higher light yield and superior X-ray absorption efficiency, enabling rapid commercialization and driving rare-earth scintillators to the forefront of scintillator research.²⁰ However, these materials still suffer from intrinsic limitations. LYSO crystals are typically grown via the Czochralski method at temperatures exceeding 1500°C, resulting in high production costs and considerable processing complexity.²¹ GSO crystals are prone to cracking during growth and mechanical processing due to their layered crystal structure.¹⁸ GOS exhibits pronounced afterglow and light-scattering effects, which directly degrade the spatial resolution of imaging systems.^{19,22} These shortcomings substantially restrict their large-scale deployment in high-precision and cost-sensitive X-ray detection applications. Over the past decade, lead-based halide perovskites, typified by CsPbX₃ (X = Cl, Br, I, or mixed halide), have emerged as promising scintillator candidates due to their high X-ray absorption coefficients, high photoluminescence quantum efficiencies, tunable bandgaps, and compatibility with low-cost solution processing.^{23,24} However, unavoidable self-absorption losses, potential biological toxicity associated with lead, and poor stability against light, heat, and moisture have severely hindered their transition toward industrial implementation.^{25,26} In response to these challenges, extensive research efforts have been devoted to the development of rare-earth halide scintillators with perovskite-like structures.^{3,27,28} For instance, LiLuF₄:Tb exhibits a significantly higher X-ray absorption coefficient than conventional detector materials such as CdTe and BaF₂, achieving a minimum detectable dose rate as low as 36.31 nGy s^{−1}, well below the standard clinical diagnostic threshold of 5.50 μGy s^{−1}.²⁹ The NaLuF₄ system has been successfully extended to applications including flexible 3D memory imaging and precision biomedical diagnostics, benefiting from the processability of core-shell architectures and the structural tunability enabled by ionic doping.^{30–32} OD Cs₃CeBr₆ exhibits an ultrafast decay lifetime of only 29.6 ns, making it particularly suitable for dynamic imaging scenarios.³³ Moreover, Sb-doped 3D Cs₂NaLuCl₆ and OD Cs₃LuCl₆ can form 3D/OD heterostructures via a sodium-alloying strategy, realizing broadband color-integrated X-ray imaging.³⁴ Within hybrid structural systems, rare-earth ions can form optically transparent glasses through coordination with organic ligands or be incorporated into metal-organic frameworks to construct high-entropy scintillators via ligand coordination.³⁵ Furthermore, rare-earth ions also serve as highly efficient luminescence activators. For example, Yb³⁺ doping enables CsPbCl₃ scintillators to achieve light yields as high as 112,281 photons MeV^{−1}.³⁶ Combined with the near-infrared emission of Yb³⁺, this material has been employed for dual-modal visible-infrared imaging.³⁷ Fundamentally, the luminescence mechanisms of rare-earth halide scintillators primarily originate from electronic transitions between the f-f or f-d configurations of rare-earth ions. Their rich energy-level structures confer tunable emission spanning from ultraviolet to infrared wavelengths, high luminescence efficiency, and broadly adjustable decay times ranging from nanoseconds to milliseconds.³⁸ These intrinsic attributes collectively position rare-earth halides as one of the most promising material

platforms for high-performance X-ray scintillators.

In recent years, rare-earth halide perovskite-like materials have demonstrated broad application potential in X-ray detection owing to their tunable crystal structures, excellent radiation response characteristics, and flexible fabrication routes, leading to rapid progress in this field. This review provides a systematic overview of rare-earth halide perovskite-like structure materials for X-ray detection applications. First, the fundamental luminescence mechanisms of rare-earth scintillators under X-ray excitation are comprehensively elucidated. Subsequently, the key performance advantages of these materials for indirect X-ray detection are summarized based on structural classification, and their emerging applications in low-dose X-ray detection, dynamic imaging, and multimodal imaging are discussed. Finally, the critical challenges currently facing this material class are identified, including the regulation of scintillation decay lifetimes for dynamic imaging compatibility, low-cost and scalable fabrication coupled with precise valence-state control, and optimization of energy transfer pathways in rare-earth-doped systems. In light of ongoing trends in radiation detection, such as reduced radiation dose, enhanced spatial resolution, and multi-scenario adaptability, future research directions are proposed to facilitate the practical translation of rare-earth halide scintillator materials into advanced X-ray detection technologies.

SCINTILLATION MECHANISM OF RARE-EARTH HALIDE PEROVSKITE-LIKE SCINTILLATORS

The interaction of X-rays with rare-earth-based scintillators can be broadly divided into three stages: energy deposition, carrier transport, and radiative recombination. Upon entering the scintillator, incident X-ray photons transfer their energy predominantly through interactions with extranuclear electrons, including the photoelectric effect, Compton scattering, and electron-positron pair production. The relative contribution of each interaction pathway is jointly governed by the atomic number (Z) of the constituent elements and the energy of the incident photons. In the sub-MeV energy regime, the photoelectric effect dominates the energy deposition process, with its interaction cross-section in high- Z materials scaling approximately with Z^4 .³⁹ Consequently, rare-earth elements with high atomic numbers, such as Lu, Gd, and Tb are highly effective in enhancing X-ray absorption and energy deposition efficiency by increasing the probability of photoelectric interactions, enabling efficient conversion of high-energy photons into cascades of secondary electron-hole pairs. As the photon energy increases, Compton scattering becomes increasingly significant, whereas electron-positron pair production only becomes relevant once the photon energy exceeds 1.02 MeV. Following energy deposition, a high density of electron-hole pairs or excitons is generated and subsequently undergoes thermalization and transport within a characteristic timescale of $\sim 10^{-12}$ – 10^{-8} s.^{1,40,41} During this process, lattice imperfections, such as vacancies, impurity atoms, and grain boundaries, introduce trap states that perturb carrier motion. The associated carrier dynamics can be quantitatively described by trap depth: shallow traps (typically 0.1–0.6 eV) may temporarily capture carriers and release them through thermal activation under the room-temperature activation energy, giving rise to microsecond-to-millisecond slow decay components and afterglow in the scintillation signal. In contrast, deep traps (> 1 eV) tend to immobilize carriers over extended timescales and act as nonradiative recombination centers via the Shockley-Read-Hall process, increasing the likelihood of energy dissipation and thereby reducing prompt luminescence efficiency.^{42–44} When excitation energy reaches the luminescence centers, photon emission occurs through radiative recombination pathways. These include direct electron-hole recombination, exciton recombination mediated by luminescent centers, and characteristic intra-ionic electronic transitions of rare-earth ions activated through energy transfer processes. Band-edge exciton recombination typically occurs on the nanosecond timescale (< 10 ns), whereas self-trapped exciton (STE), involving significant lattice relaxation, generally exhibits broadband emission with large Stokes shifts and microsecond-scale lifetimes (~ 1 μ s).^{45,46} In addition, rare-earth luminescence follows quantum selection rules: allowed $5d \rightarrow 4f$ transitions of Ce^{3+} or Eu^{2+} possess large oscillator strengths and display fast nanosecond decay (typically ~ 30 ns), making them particularly suitable for time-resolved imaging. By contrast, $4f \rightarrow 4f$ transitions of ions such as Tb^{3+} or Eu^{3+} exhibit much longer lifetimes ranging from hundreds of microseconds to milliseconds (typically 2–8 ms).⁴⁵ After cessation of X-ray

irradiation, thermally stimulated detrapping enables carriers to re-enter the conduction or valence bands, followed by delayed radiative recombination.^{47,48} This process gives rise to persistent luminescence, commonly referred to as long afterglow emission, which can span from several minutes to several days depending on the depth, distribution, and population of the involved trap states.⁴⁷ Overall, the macroscopic scintillation characteristics of rare-earth halide scintillators, including light yield, decay time, and afterglow, arise from the interplay among Z -dependent energy deposition efficiency, defect-regulated carrier transport kinetics, and the radiative recombination pathways determined by the electronic energy-level structure.

Clear distinctions arise between rare-earth halide scintillators and their oxide counterparts in terms of emission mechanisms and spectral tunability. Rare-earth oxide scintillators typically rely on a limited set of intrinsic luminescence pathways, predominantly governed by $f-f$ or $f-d$ electronic transitions of rare-earth ions.⁴⁹ In contrast, rare-earth halide scintillators exhibit markedly richer emission behaviors, enabled by the structural flexibility and the compositional diversity.³⁸ These materials can simultaneously support multiple radiative pathways, including band-edge emission, STE recombination, and characteristic rare-earth ionic emission (Figure 1A). Among these pathways, band-edge emission arises from electronic transitions between the conduction band minimum and the valence band maximum. The emission channel is characterized by high radiative efficiency and nanosecond-scale decay lifetimes. In addition, characteristic emission from rare-earth ions substantially enriches the spectral versatility of halide scintillators. Such emission primarily originates from intra-configurational $4f-4f$ transitions and inter-configurational $4f-5d$ transitions. Through judicious selection of rare-earth activators, radioluminescence spanning the ultraviolet, visible, and near-infrared spectral regions can be achieved.³⁸ As illustrated in Figure 1B, ions such as Ce^{3+} , Pr^{3+} , Nd^{3+} , Gd^{3+} , Sm^{3+} , Eu^{3+} , Tb^{3+} , Dy^{3+} , Ho^{3+} , Er^{3+} , and Tm^{3+} exhibit visible emission, while Nd^{3+} , Ho^{3+} , Er^{3+} , Tm^{3+} , and Yb^{3+} additionally display near-infrared emission.^{38,50,51} Except for divalent rare-earth ions, such as Eu^{2+} and Yb^{2+} , whose luminescence is dominated by parity-allowed $d-f$ transitions, most trivalent rare-earth ions emit via parity-forbidden $4f-4f$ transitions.⁵² The radiative decay characteristics of rare-earth scintillators are therefore closely related to the change in orbital angular momentum (Δl) between the excited and ground states.⁵³ When Δl equals one, the transition is parity-allowed and typically involves $5d$ orbitals.⁵⁴ These orbitals are spatially extended and weakly shielded by inner electrons, making them highly sensitive to the surrounding crystal field and prone to band broadening. Consequently, such transitions produce broadband emission with nanosecond-scale decay times. In contrast, transitions with Δl equal to zero are parity-forbidden, resulting in sharp-line emission and longer decay times, typically spanning the microsecond to millisecond regime.^{55,56} Low-dimensional metal halides introduce an additional emission pathway owing to their intrinsically soft lattice frameworks. Strong coupling between charge carriers and lattice vibrations readily induces local lattice distortion, thereby stabilizing STE.^{44,57,58} Radiative recombination of these excitons typically produces broadband emission accompanied by large Stokes shifts and pronounced temperature dependence.^{59,60} In rare-earth halide scintillators, STEs can further function as intermediate energy states that mediate the transfer of absorbed X-ray energy to the $4f$ levels of rare-earth ions, thereby compensating for the intrinsically low absorption cross-section associated with parity-forbidden $f-f$ transitions.^{44,46,61} The generation of STEs results from lattice relaxation following high-energy excitation, during which photo-generated electron-hole pairs become localized through lattice distortion and form bound excitonic states with relatively long radiative lifetimes. From a kinetic perspective, the scintillation process is governed by the competition between STE formation and energy transfer to luminescent activator centers.^{62,63} STE formation generally involves a relatively low activation barrier for carrier self-trapping, leading to a large formation rate constant.⁶³ Importantly, lattice dimensionality plays a key role in regulating these processes: reduced structural dimensionality enhances quantum confinement and strengthens electron-phonon coupling, which lowers the self-trapping barrier and increases the STE rate constant while simultaneously shortening the carrier diffusion length and suppressing the activator energy-transfer rate constant.^{64–66} In contrast, higher-dimensional frameworks promote carrier delocalization and long-range migration, increasing diffusion length and facilitating more efficient energy transfer to

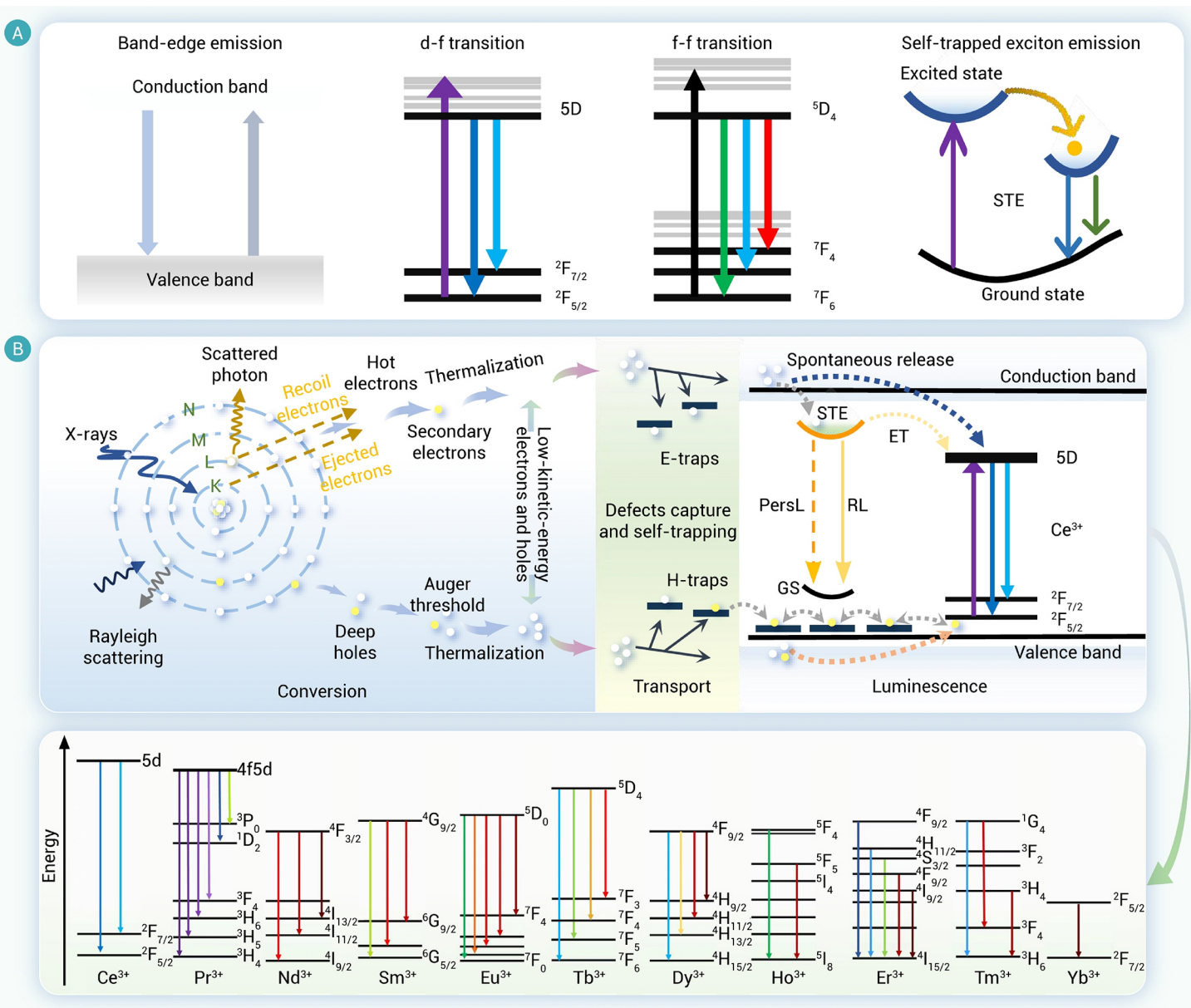


Figure 1. Scintillation mechanisms of rare-earth halide perovskite-like scintillators (A) Photoluminescence mechanisms of rare-earth halide scintillators: band-edge emission, self-trapped exciton emission, d-f transition, and f-f transition. (B) Schematic illustration of the scintillation process in rare-earth halide scintillators.

activator centers.⁶⁷ Consequently, the balance between the STE rate constant and energy-transfer rate constant, modulated by lattice dimensionality, ultimately determines the dominant scintillation pathway and emission characteristics in rare-earth halide scintillators. In rare-earth organic-inorganic hybrid systems or doped halide materials, the scintillation mechanisms become more complex, necessitating interpretation in the context of the specific structural, electronic, and energy-transfer characteristics of the host lattice.

RARE-EARTH HALIDE PEROVSKITE-LIKE SCINTILLATORS

All-inorganic rare-earth halide scintillators

AREF₄ rare-earth halides. Rare-earth fluorides are characterized by low lattice phonon energies and dense crystal structures, which effectively suppress nonradiative relaxation processes.⁶⁸ This intrinsic advantage underpins their high luminescence efficiency and favorable energy conversion performance. According to their chemical composition, rare-earth fluorides are commonly divided into binary fluorides (REF₃) and composite fluorides (AREF₄, where A denotes an alkali metal ion). Binary fluorides such as CeF₃ and LaF₃ often exhibit pronounced hygroscopicity and typically require high-purity anhydrous precursors, thereby increasing synthesis cost and complexity.

In contrast, composite fluorides, including NaYF₄, NaGdF₄, and NaLuF₄, have attracted growing attention due to their lower effective phonon energies, enhanced structural stability, and flexible solid solubility for rare-earth dopants.^{38,68} AREF₄ compounds are particularly well known for their efficient upconversion luminescence, wherein the sequential absorption of two or more low-energy photons produces higher-energy ultraviolet or visible emission.⁶⁹ The efficiency of this process is strongly sensitive to surface-related nonradiative relaxation and defect-associated energy levels. Materials with high upconversion generally exhibit low defect densities, efficient energy migration, and well-passivated luminescent centers.³² These attributes are likewise beneficial under X-ray excitation, as they facilitate radiative recombination and contribute to enhanced radioluminescence intensity as well as trap-regulated afterglow behavior.⁷⁰ By contrast, surface ligands with high phonon energies, lattice distortions, or unpassivated defects promote nonradiative energy dissipation.⁷¹ Moreover, overly strong sensitization pathways may compete with X-ray-induced carrier recombination, limiting trap filling and reducing scintillation efficiency.⁷² To address these challenges, structural design strategies have increasingly focused on core-shell architectures to optimize the scintillation behavior of AREF₄ materials. Introducing a shell material with low lattice mismatch and low phonon energy facilitates coher-

ent epitaxial growth while effectively passivating surface defects and suppressing surface-related energy losses.⁷³ Such architectures provide a more favorable environment for carrier confinement and energy transfer. Based on lattice compatibility and the spatial distribution of rare-earth ions, core-shell structures are generally classified as homogeneous core-shell, heterogeneous core-shell, or active-core/active-shell systems.

The lanthanide contraction results in highly similar ionic radii and physico-chemical properties between Y^{3+} and rare-earth ions ranging from Gd^{3+} to Lu^{3+} , thereby providing a robust structural basis for introducing low-lattice-mismatch rare-earth ions into $AREF_4$ hosts and for constructing heterogeneous core-shell architectures.^{76,77} Leveraging this principle, Ou et al. prepared a $NaLuF_4:Tb^{3+}@NaYF_4$ heterogeneous core-shell structure by combining co-precipitation with epitaxial growth (Figure 2A).³² The resulting material delivers markedly stronger X-ray radioluminescence than $SrAl_2O_4:Eu^{2+}/Dy^{3+}$ and sustains persistent luminescence for more than 30 days. This exceptional performance arises from the high-Z Lu ions in the $NaLuF_4$ core, which enhance X-ray absorption efficiency and induce F⁺ displacement, thereby generating vacancy-type electron traps and interstitial-type hole traps (Figure 2B). Short-range lattice displacements give rise to shallow traps, whereas long-range displacements produce deep traps, enabling effective energy storage. Holes recombine at Tb^{3+} centers to generate characteristic emission upon release, while the $NaYF_4$ shell suppresses surface-related nonradiative energy losses. Consequently, the core-shell architecture achieves a 1.5-fold enhancement in radioluminescence and a 6.5-fold increase in persistent luminescence compared with the bare core (Figure 2C). Furthermore, rational modulation of rare-earth dopants within the $NaLuF_4$ core enables multicolor afterglow emission and supports an X-ray luminescence extension imaging scheme that integrates X-ray writing, long-term afterglow storage, and thermal or optical readout (Figures 2D-E). This approach delivers spatial resolutions exceeding 20 lp mm⁻¹ and optical memory retention times longer than 15 days, enabling three-dimensional imaging of complex circuit architectures. In addition, Wu et al. demonstrated that carrier dynamics can be effectively tuned through the combined use of phase regulation and defect engineering.⁷⁴ By controlling the α/β phase structure of $NaLuF_4$ and introducing Mn^{2+} co-doping, they synthesized a $NaLuF_4:Mn^{2+}/Gd^{3+}$ storage phosphor in which energy trapping and release processes were deliberately modulated (Figures 2F-G). This structural and compositional design yielded an exceptionally prolonged afterglow persisting for up to 108 h, thereby enabling long-term imaging of biological skeletal structures with a storage duration approaching 30 days (Figure 2H). Additionally, Hou et al. developed a swallowable X-ray dosimeter, which integrates long-afterglow nanoscintillators and a pH-responsive module to monitor radiation dose, pH, and temperature in real time during gastrointestinal radiotherapy.⁷⁸ Beyond optical storage applications, these advances underscore rare-earth fluorides as versatile material platforms with increasing potential in information security technologies.⁷⁹ A representative example is the encryption-type scintillator reported by Yang et al. based on $NaLuF_4:Gd^{3+}/Ce^{3+}$ (Figure 2I), which enables X-ray-encrypted information writing within silicone rubber films and programmable optical readout through coupling with the thermally responsive emission of a perovskite nanocrystal layer (Figure 2J).⁷⁵ More recently, such structural and compositional design strategies have been extended to hybrid heterojunctions. Li et al. developed a $NaYF_4:Yb,Tm@Cs_4PbBr_6$ architecture in which upconversion emitters are epitaxially embedded within a perovskite matrix rather than being confined to a conventional core-shell configuration.⁸⁰ By tuning the thickness of the Cs_4PbBr_6 layer, full-color upconversion emission spanning the blue, green, and red spectral regions was achieved. Furthermore, solid-solution fluorides formed via substitution of Na^+ with Li^+ , such as $LiLuF_4:Tb^{3+}$ and $LiYbF_4:Tb^{3+}$, have further broadened the application of rare-earth fluorides in high-performance X-ray and γ -ray detection.^{29,81} Notably, Lu et al. reported a $LiLuF_4:Tb^{3+},Cu^{2+}$ scintillator with an ultralow X-ray detection limit of 2.7928 nGy s⁻¹, an imaging resolution of 22 lp mm⁻¹, and an information storage time exceeding 48 h.⁸² Cu^{2+} co-doping not only enhances radioluminescence intensity but also introduces additional carrier trapping states, thereby reinforcing persistent luminescence.

Active molecular doping has emerged as a powerful strategy for improving the performance of core-shell structured materials.^{68,87} Representative exam-

ples include the work by Huang and Liu groups, who developed a $NaLuF_4:Tb^{3+}@NaYF_4@SiO_2$ core-shell nanocomposite in which organic 4-PP molecules were incorporated into the outer SiO_2 layer (Figure 3A).⁸³ Under X-ray irradiation, the inorganic core absorbs high-energy photons to generate persistent luminescence, while the SiO_2 shell stabilizes the triplet states of the organic molecules through hydrogen bonding and spatial confinement, leading to long-lived cyan afterglow emission (Figure 3B). With an afterglow duration exceeding two months and good aqueous dispersibility, the material was applied as an anti-counterfeiting ink for screen-printed information encryption, where the distinct afterglow colors arising from the inorganic and organic components under combined UV and X-ray excitation enable reliable identification of encoded information (Figure 3C). Building upon this design principle, the authors further extended the concept to a more complex RE- SiO_2 architecture by selectively doping different organic molecules into the core, intermediate shell, and outer layer of monodisperse SiO_2 microspheres. By rationally matching molecular energy levels with excitation wavelengths, both emission color and afterglow lifetime were finely tuned over a broad temporal window ranging from 8.6 μ s to 3.42 s.⁸⁸ In addition, Wang et al. reported a multilayer core-shell nanomaterial featuring spatially separated activators (Figure 3D). Upon X-ray excitation, radioluminescence is simultaneously generated from the $NaYF_4:Tb^{3+}$ core and the $NaYF_4:Eu^{3+}$ shell. Subsequent electron trapping and interfacial energy transfer to Tb^{3+} centers sustain persistent emission. Leveraging this delayed luminescence, X-ray-written Morse code patterns were encoded in nanoscintillator films, where information storage relies on afterglow persistence, and decoding can be achieved through simple mechanical friction or thermal stimulation.⁸⁴ Moreover, Lei et al. successfully achieved delayed 3D imaging based on a multi-layer core-shell structure, with a spatial resolution of approximately 25.64 lp mm⁻¹.⁸⁹ Beyond luminescence modulation, practical X-ray imaging applications require high scintillator loading while maintaining optical transparency. To meet this requirement, Zhang group prepared a flexible and transparent scintillating film by drop casting, achieving an exceptionally high nanoparticle loading of 83% at a thickness of ~55 μ m while retaining 89% optical transparency. The resulting film delivers radioluminescence intensity surpassing that of commercial LYSO scintillator, a low detection limit of 661 nGy s⁻¹, and excellent imaging performance, including a spatial resolution of 8 μ m and a spatial resolution of 122 lp mm⁻¹ when the modulation transfer function is 0.2 (Figures 3E-F).⁸⁵ In addition to structural and compositional design, emission color can also be regulated through dopant concentration control. Lu et al. demonstrated that increasing the Ho^{3+} concentration in $NaLuF_4:Ho^{3+}$ shortens interionic distances and enhances the cross-relaxation process of [$(^6F_4, ^5S_2, ^5I_7) \rightarrow (^6F_5, ^5I_6)$], which promotes red emission while suppressing green emission. Under identical X-ray irradiation doses, systematic variation of Ho^{3+} content induces a continuous green-to-red emission shift governed by the competition between cross-relaxation and concentration quenching (Figure 3G). The concentration-dependent color tunability provides a practical strategy for radiometric and color-encoded X-ray dosimetry (Figures 3H-I).⁸⁶

Overall, the scintillation performance of $AREF_4$ rare-earth halides is intrinsically governed by the interplay between their crystal structure and carrier relaxation pathways. The rigid fluoride lattice with low phonon energy effectively suppresses multiphonon nonradiative relaxation, thereby promoting efficient radiative recombination at rare-earth activator centers. At the same time, the structural compatibility among lanthanide ions enables flexible dopant engineering and the construction of core-shell architectures that regulate carrier confinement and defect distribution. As a result, these structural characteristics directly translate into enhanced light yield, prolonged persistent luminescence, and improved radioluminescence stability. Consequently, $AREF_4$ systems illustrate how lattice rigidity, phonon energy, and defect engineering collectively determine the scintillation mechanism and ultimately govern key performance metrics such as emission efficiency, afterglow duration, and imaging sensitivity.

A_3REX_6 rare-earth halides. The luminescence performance of rare-earth-activated materials is closely governed by the efficiency of energy transfer between the host lattice and the activator centers. When a significant mismatch exists between the host-related excitation states and the energy levels of the rare-earth ions, the energy transfer process becomes inefficient, leading to severe luminescence losses.^{90,91} Under such conditions, minimiz-

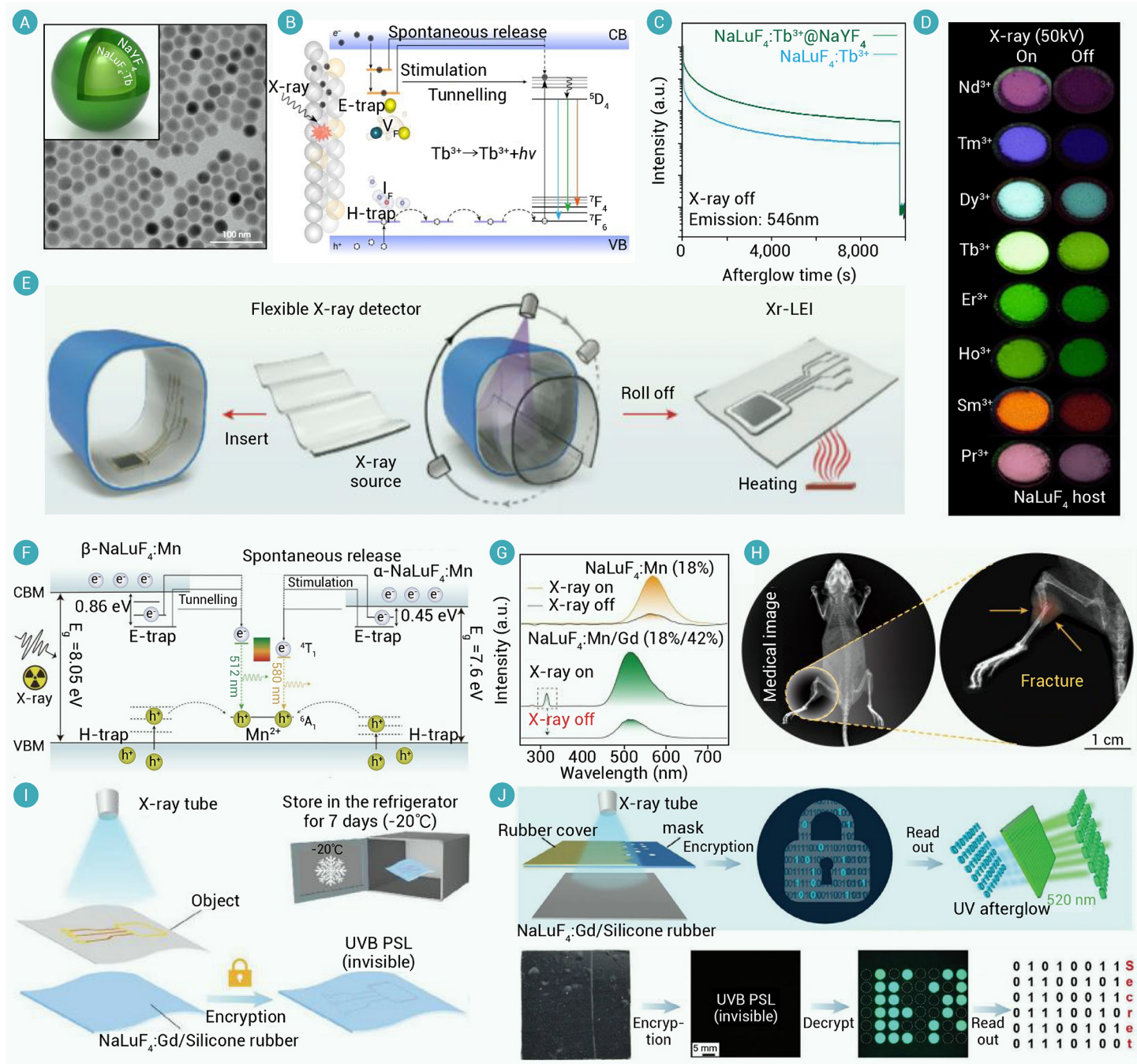


Figure 2. Scintillation mechanisms of AREX₄ rare-earth halides (A–C) Core-shell NaLuF₄:Tb³⁺@NaYF₄ nanocrystals showing microstructure, persistent radioluminescence mechanism, and enhanced 546 nm emission. **(D)** Radioluminescence and afterglow of NaLuF₄ nanocrystals doped with various activators (Nd³⁺, Tm³⁺, Dy³⁺, Tb³⁺, Er³⁺, Ho³⁺, Sm³⁺, and Pr³⁺). **(E)** Schematic of a flexible detector integrated with nano-scintillators for X-ray luminescence extension imaging. Copyright 2021, Springer Nature.³² **(F–G)** Charge trapping, Mn²⁺-mediated persistent emission, and co-doping modulation of afterglow intensity. **(H)** X-ray imaging using persistent luminescence scintillators and digitally processed medical imaging of a mouse fracture. Copyright 2024, Wiley.⁷⁴ **(I–J)** Schematic of the X-ray encryption and decryption process using NaLuF₄:Gd³⁺ persistent luminescence scintillators. Copyright 2023, Wiley.⁷⁵

ing chemical complexity and suppressing impurity-induced perturbations are particularly critical, as additional components often introduce unwanted energy level disturbances.²⁷ OD rare-earth halides with the general formula A₃REX₆ have emerged as an attractive class of materials. Their crystal and electronic structures can be regulated through rational selection of constituent ions without incorporating extraneous dopants, thereby enabling precise control over the energy landscape (Figure 4A). As a result, the luminescence behavior of these materials is predominantly dictated by the intrinsic electronic transitions of the rare-earth ions (Figure 4B). Within this framework, Im group synthesized Rb₃TbCl₆ and Cs₃TbCl₆ single crystals using a hydrothermal method, achieving light yields of 88,800 and 56,800 photons MeV⁻¹, respectively, both of which markedly exceed that of commercial GOS

scintillators (Figure 4C).⁹² Such exceptional radioluminescence performance was attributed to the low exciton binding energy of these hosts, which promotes efficient generation of free charge carriers during thermalization and subsequently enhances radiative recombination at the Tb³⁺ emission centers. In contrast, Chai et al. reported complementary behavior in Cs₃EuCl₆ microcrystals prepared via high-temperature calcination.⁹³ The material exhibits extraordinary resistance to thermal quenching, maintaining stable luminescence up to 730°C. The feature is attributed to the combined effect of a relatively high exciton binding energy and the parity-forbidden nature of 4f–4f transitions, maintaining a light yield of 12,877 photons MeV⁻¹ and a low X-ray detection limit of 643.1 nGy s⁻¹. Beyond intrinsic luminescence properties, synthetic accessibility and scalability are critical considerations for practical

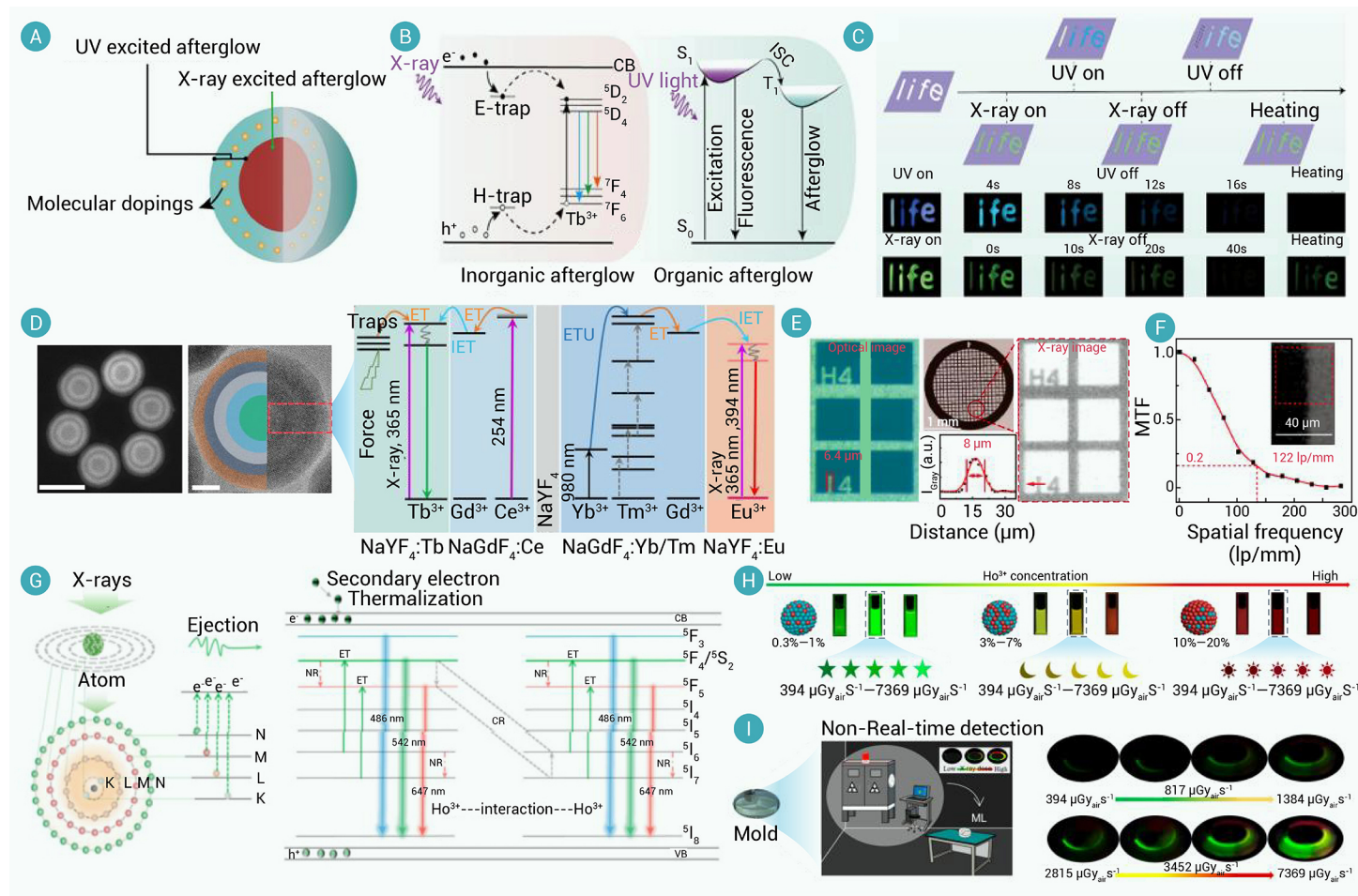


Figure 3. AREX₄ rare-earth halides for multi-scenario X-ray applications (A–C) Core-shell-molecule composite (NaLuF₄:Tb³⁺@NaYF₄@SiO₂) and its X-ray/UV-induced persistent luminescence mechanism for patterned information storage. Copyright 2025, Wiley.⁸³ (D) Morphology diagram of a multilayer core-shell nanoparticle and the luminescence mechanism between different core-shell layers. Copyright 2024, American Chemical Society.⁸⁴ (E–F) Ultra-high-loading ceramic film enabling high-resolution X-ray imaging (up to 122 lp mm^{−1}). Copyright 2022, American Chemical Society.⁸⁵ (G–I) Radioluminescence and dose-dependent mechanoluminescence in NaLuF₄:Ho³⁺ nanocrystals, enabling non-real-time visual X-ray dosimetry. Copyright 2023, American Chemical Society.⁸⁶

applications. Conventional solvothermal and solid-state methods often involve complex procedures, elevated temperatures, and limited production efficiency. To overcome these challenges, Im group introduced a room-temperature grinding strategy, enabling the high-yield preparation of Cs₃CeCl₆, Cs₃TbCl₆, and Cs₃EuCl₆ microcrystals.⁹⁴ Building upon this progress, Li et al. developed a room-temperature recrystallization approach that exploits solubility differences to induce selective co-precipitation, thereby allowing efficient synthesis of Cs₃LnCl₆ microcrystals.⁹⁵ Among the resulting compositions, Cs₃TbCl₆ exhibited a light yield of 51,800 photons MeV^{−1}. Notably, this strategy was further extended to the fabrication of high-entropy rare-earth halide crystals, exemplified by Cs₃(TbDyHoErTm)Cl₆, opening a viable pathway for designing high-entropy compositions within all-inorganic rare-earth halide scintillators.

Cs₃CeBr₆ nanocrystals reported by Xu group exhibit high material stability, an ultrafast radioluminescence decay time of approximately 29 ns, and an exceptionally low detection limit of 11.2 nGy s^{−1}.³³ Under X-ray irradiation, charge carriers rapidly thermalize to form excitons, which are efficiently captured by Ce³⁺ luminescent centers and subsequently emit light via allowed 5d → 4f transitions (Figure 4D). This ultrafast decay renders Cs₃CeBr₆ well suited for dynamic X-ray imaging applications that require rapid temporal response (Figure 4E). Beyond intrinsic emission, controlled impurity modulation offers an additional pathway for performance optimization. In Cs₃TbCl₆, trace Sb³⁺ doping has been shown to enhance luminescence by inducing the formation of STE associated with [SbCl₆]^{3−} units, which establish an efficient energy transfer channel toward Tb³⁺ centers.⁹⁶ However, the light yield gradually decreases due to concentration quenching as the Sb³⁺ concentration increases, underscoring the importance of precise dopant concentration

control (Figure 4F). Structural flexibility further expands the tunability of A₃REX₆-type halides.⁹⁷ Cs₃YCl₆ can be synthesized via an anti-solvent route, and systematic variation of the CsCl content drives a phase transformation to Cs₃YCl₇ (Figure 4G).⁶¹ Subsequent co-doping with activator ions such as Sb³⁺, Ce³⁺, and Ho³⁺ enables continuous modulation of emission wavelengths over a wide spectral range, highlighting the compositional and structural versatility of A₃REX₆ rare-earth halides. In summary, the scintillation performance of A₃REX₆ rare-earth halides is closely associated with their zero-dimensional crystal framework composed of isolated [REX₆]^{3−} octahedra. This structural configuration favors localized excitation and efficient capture of carriers by rare-earth luminescent centers. Through rational dopant incorporation and compositional modulation, the radiative recombination pathways and emission characteristics can be effectively tuned. As a result, A₃REX₆ materials can achieve high light yield, fast radioluminescence response, and low detection limits.

A₂BREX₆ rare-earth halides. Cs₂AgBiBr₆ was among the early double-perovskite materials investigated for scintillation applications.^{98,99} However, its relatively modest scintillation efficiency underscored the importance of highly localized luminescent centers, thereby motivating the subsequent development of rare-earth-based double-perovskite scintillators.⁹ In contrast to low-dimensional rare-earth halides, three-dimensional A₂B(II)RE(III)X₆ double-perovskite framework features a continuous crystal lattice and enhanced structural robustness, which facilitates long-range carrier transport (Figure 5A). The structural stability of double perovskites is commonly assessed using the tolerance factor (τ), which reflects A-site ionic size matching, and the octahedral factor (μ), which describes the geometric stability of BX₆ units.¹⁰⁰ When both parameters fall within appropriate ranges, a stable three-

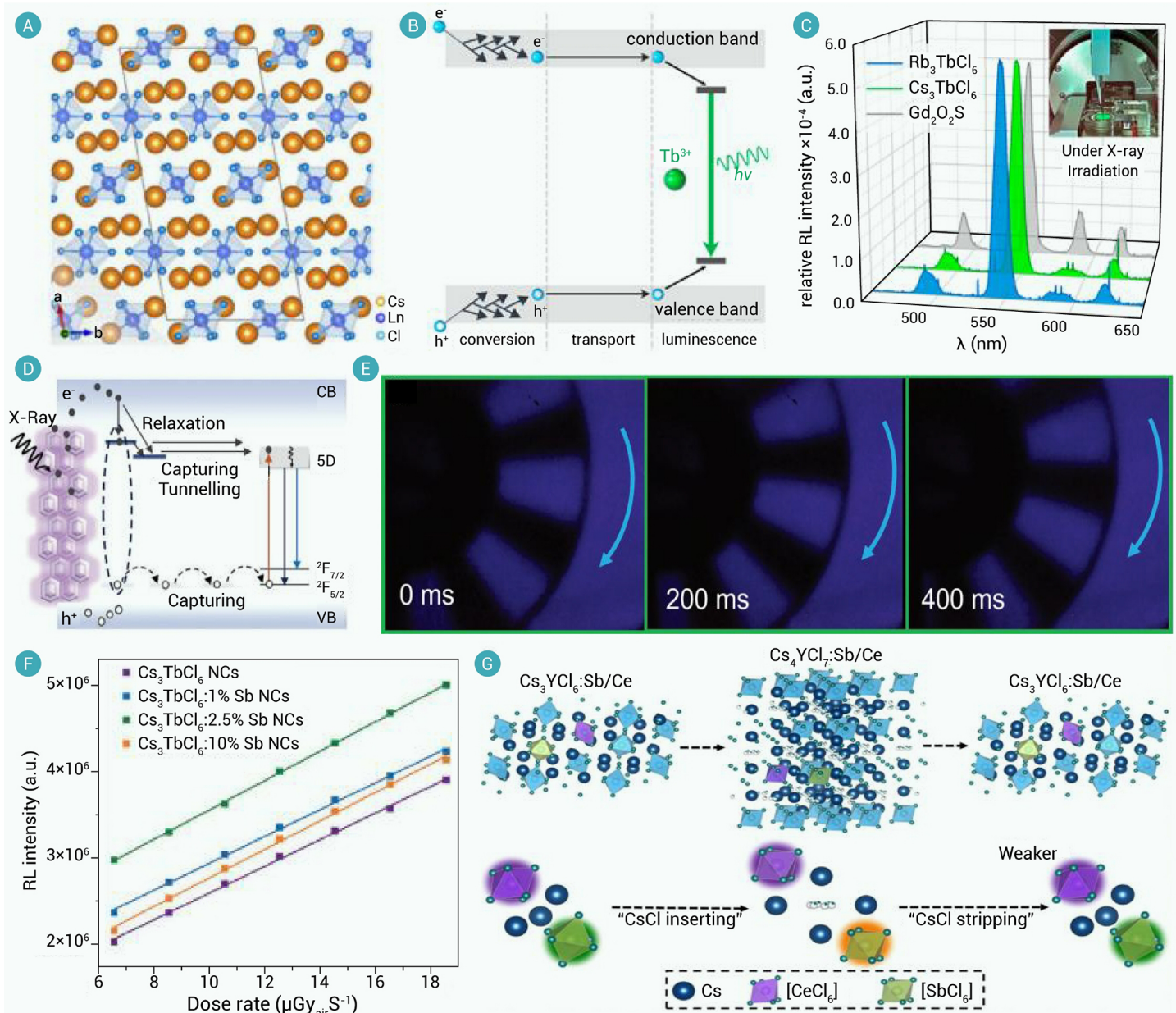


Figure 4. Scintillation mechanisms and properties of A₃REX₆ rare-earth halides (A) Crystal structure of Cs₃LnCl₆. Copyright 2024, American Chemical Society.⁹⁴ (B) Scintillation conversion mechanism in Cs₃TbCl₆ and Rb₃TbCl₆. (C) Light yield of Rb₃TbCl₆ (88800 photons MeV⁻¹) and Cs₃TbCl₆ (56800 photons MeV⁻¹). Copyright 2022, American Chemical Society.⁹² (D) X-ray scintillation mechanism of Cs₂CeBr₆ nanocrystals. (E) Dynamic X-ray imaging using Cs₃CeBr₆ nanocrystals. Copyright 2025, Wiley.³³ (F) Relationship between emission intensity and dose rate for Sb³⁺-doped Cs₃TbCl₆ nanocrystals. Copyright 2024, Wiley.³⁶ (G) Schematic illustration of the reversible structural transition between Cs₃YCl₆:Sb³⁺/Ce³⁺ and Cs₄YCl₇:Sb³⁺/Ce³⁺. Copyright 2025, Wiley.⁶¹

dimensional lattice can be realized. Within the Cs₂NaREX₆ system, well-matched τ and μ values ensure lattice integrity and high compositional compatibility, thereby providing a favorable platform for alloying and dopant engineering.¹⁰¹ Moreover, the incorporation of high-Z Cs element enhances X-ray absorption efficiency, further supporting the suitability of this system for radiation detection. Early rare-earth double perovskites were predominantly synthesized as bulk single crystals using concentrated hydrochloric acid evaporation crystallization or the Bridgman method, resulting in limited control over material morphology and composition.¹⁰² With the evolution of synthetic strategies, including grinding-sintering, hydrothermal processing, evaporation-based growth, vacuum-assisted deposition, and hot-injection techniques, Cs₂NaREX₆ materials have become accessible across multiple dimensionalities. Using a solvothermal approach, Hu et al. reported Cs₂NaTbCl₆ and Cs₂NaEuCl₆ in 2018, with Cs₂NaTbCl₆ achieving a light yield of 46,600 photons MeV⁻¹.¹⁰³ Luminescence performance can be further enhanced through sensitizer incorporation. In particular, Ce³⁺ effectively

promotes energy transfer. Ce-doped Cs₂NaTbCl₆ reported by Wang et al. achieved a light yield of 52,153 photons MeV⁻¹.¹⁰⁴ Using an anti-solvent method, Zhou et al. further improved Cs₂NaTbCl₆:Ce³⁺, realizing a higher light yield of 53,200 photons MeV⁻¹ together with an imaging resolution of 31.0 lp mm⁻¹ and a detection limit of 28.8 nGy s⁻¹.¹⁰⁵ Introducing Sb³⁺ generates STE in Cs₂NaTbCl₆, thereby reducing the detection limit to 140 nGy s⁻¹ and enhancing detection sensitivity by nearly fivefold.¹⁰⁶ Additional Mn²⁺ co-doping introduces persistent luminescence, extending the functionality to delayed imaging.¹⁰⁷ Exploiting the chemical stability of Gd³⁺, Nohyun et al. developed multi-ion-doped Cs₂NaGdCl₆:Tb³⁺, achieving a substantially reduced detection limit to 41.32 nGy s⁻¹. Environmental adaptability has also been demonstrated.^{108–110} More recent studies have clarified the critical role of B(l)-site cations in governing structure-performance relationships in rare-earth double perovskites. In Cs₂NaRECl₆, the ionic radius of Na⁺ closely matches the lattice cavities of the double perovskite framework, contributing to structural stabilization and promoting efficient carrier localization within

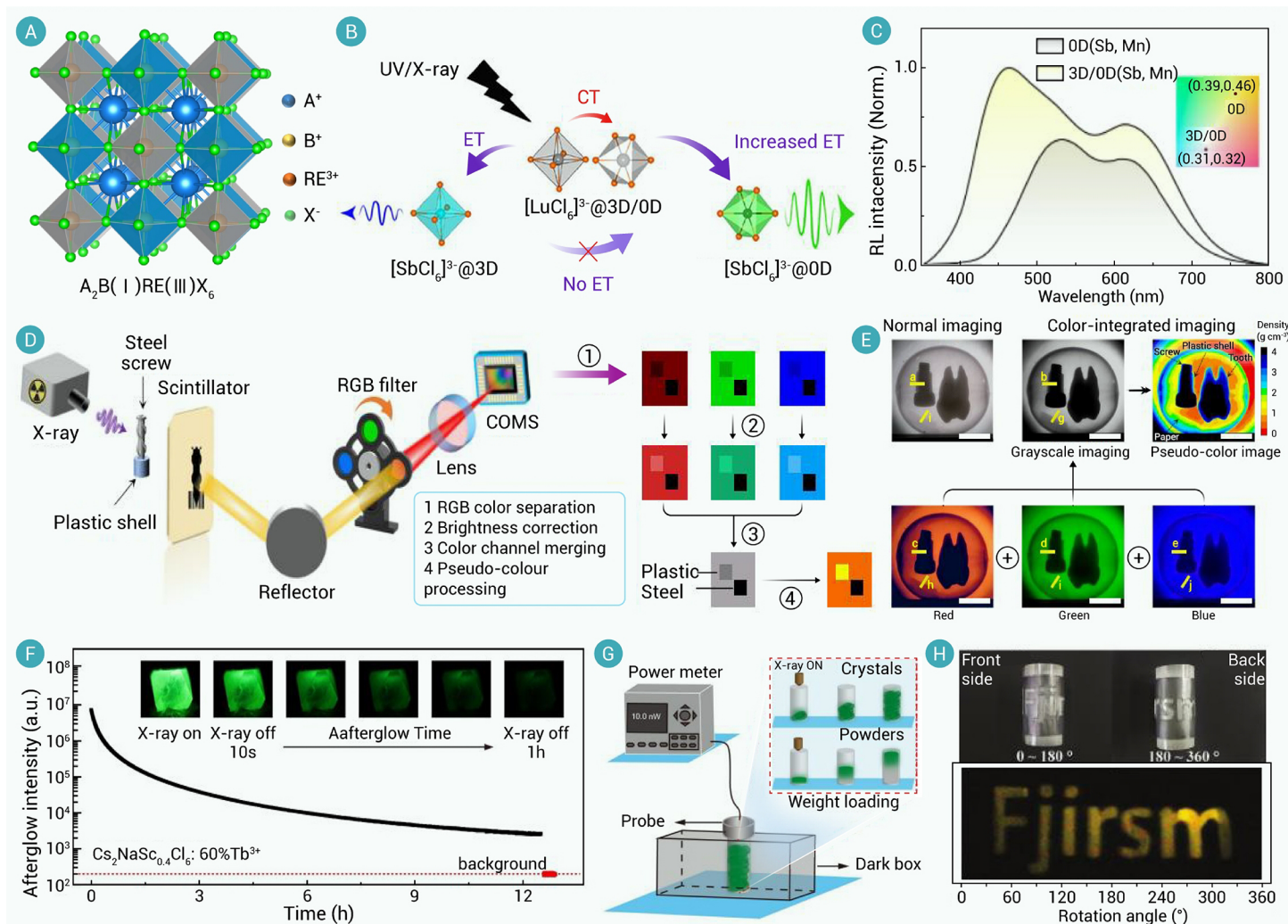


Figure 5. Scintillation mechanisms and properties of $A_2B(III)RE(III)X_6$ rare-earth halides (A) Crystal structure of $A_2B(III)RE(III)X_6$. (B–E) Dimensionality-dependent luminescence mechanisms and radioluminescence spectra of 3D $Cs_2NaLuCl_6(Sb)$ and 0D $Cs_3LuCl_6(Sb)$ heterostructures, enabling broadband emission for color-integrated X-ray imaging through spectral modulation and image fusion processing. Copyright 2025, Springer Nature.³⁴ (F) Ultra-long persistent luminescence decay curve of $Cs_2NaScCl_6:Tb^{3+}$ crystals over 12 hours. (G) Working principle of a radiation storage battery using persistent luminescence materials. Copyright 2022, Wiley.¹¹⁴ (H) 3D X-ray imaging using a flexible scintillator film of $Cs_2Na_{0.9}Ag_{0.1}LuCl_6:Dy^{3+}$. Copyright 2023, Wiley.¹¹⁵

the $[RECl_6]^{3-}$ octahedra. Partial substitution of Na^+ with Li^+ or K^+ introduces an additional degree of freedom by balancing X-ray absorption efficiency with defect-state regulation.¹¹¹ Wang et al. synthesized Sb^{3+} -doped $Cs_2B(III)GdCl_6$ ($B = Li, Na, K$), revealing that the K-based compound exhibited the highest light yield of 39,900 photons MeV^{-1} , enabled by an ultrafast self-trapped exciton formation time of 279 fs and efficient Sb^{3+} -related radiative recombination.¹¹¹ The Li-based variant also outperformed the Na-based counterpart, delivering a light yield of 20,800 photons MeV^{-1} . Liu et al. introduced trace Li^+ into $Cs_2NaScCl_6$ to induce lattice distortion, thereby enhancing carrier localization and significantly improving both blue and near-infrared emission efficiencies.¹¹² These findings highlight the critical role of B(III)-site engineering in optimizing the scintillation performance of rare-earth double perovskites and provide practical guidance for rational material design. Beyond localized emission processes, Saghy et al. identified a ligand-to-metal charge transfer (LMCT) mechanism in $Cs_2NaLuCl_6$.¹¹³ The absorption spectra display broad bands with FWHMs ranging from 570 to 940 meV, with peak positions corresponding to the energy separation between Cl 3p and Ln 4f orbitals. Notably, the LMCT absorption energy scales linearly with the Ln^{2+}/Ln^{3+} redox potential, while the electronic structure exhibits an indirect bandgap. Photophysical analysis, together with the observed low photoluminescence quantum yield, indicates that excitation energy is predominantly dissipated through the LMCT pathway. Density functional theory calculations further corroborate the Cl 3p $\rightarrow$ Ln 4f transition mechanism.

Dimensional engineering has become an effective route to optimize scintil-

lation performance and elucidate mechanisms in rare-earth scintillators.^{3,116} Yang et al. constructed a Sb^{3+} -doped 3D/0D $Cs_2NaLuCl_6/Cs_3LuCl_6$ heterostructure via sodium alloying, integrating the advantages of three- and zero-dimensional halides while preserving a low effective electronic dimensionality.³⁴ The heterostructure design enhances electron-phonon coupling of localized excitons, promotes self-trapped exciton formation, and gives rise to broadband emission with a large Stokes shift and high radiative efficiency. Under either optical or X-ray excitation, interfacial charge transfer enables energy migration from the $[LuCl_6]^{3-}$ lattice to $[SbCl_6]^{3-}$ centers, thereby amplifying STE emission and establishing a charge-transfer-dominated luminescence mechanism (Figure 5B). Building upon this heterostructure design, further incorporation of Mn^{2+} yielded a broadband-emitting 3D/0D $Cs_2NaLuCl_6/Cs_3LuCl_6:Sb^{3+}, Mn^{2+}$ scintillator (Figure 5C). The broadened emission enabled a contrast-enhanced, color-integrated X-ray imaging strategy based on spectral separation and signal fusion. By exploiting the wavelength-dependent responses of optical filters and complementary metal-oxide-semiconductor sensors, RGB (i.e., Red, Green, Blue) channel separation, reconstruction, and pseudo-color mapping were successfully implemented (Figure 5D). This imaging approach enhances contrast sensitivity for low-density materials while preserving reliable identification of high-density objects, thereby demonstrating strong versatility in complex imaging scenarios (Figure 5E).

Rare-earth double perovskites exhibit considerable potential for X-ray-induced long-afterglow applications, owing to the intrinsically long decay life-

times of rare-earth ions.¹⁰¹ Wang et al. synthesized $\text{Cs}_2\text{NaScCl}_6\cdot\text{Tb}^{3+}$ via a solvothermal method, in which efficient Tb^{3+} cross-relaxation alleviates electron saturation at the $^5\text{D}_3$ level, enhances radiative transitions, and suppresses nonradiative quenching.¹¹⁴ As a result, intense persistent luminescence with a duration of up to 12 h was achieved under X-ray excitation (Figure 5F). On this basis, a radiation battery concept was proposed, wherein afterglow powders or crystals are encapsulated within plastic tubes to store X-ray energy in the form of persistent luminescence (Figure 5G). Multicolor long-afterglow emission was subsequently achieved through $\text{Mn}^{2+}/\text{Sb}^{3+}$ co-doping, which combines energy transfer from STE to Mn^{2+} centers with precise dopant concentration control.¹¹⁷ In addition, Suo et al. reported that Sb^{3+} -doped $\text{Cs}_2\text{NaScCl}_6$ exhibits Sc^{3+} emission in the 350–400 nm region together with a long-afterglow band centered at approximately 450 nm under X-ray excitation, delivering a radioluminescence yield of 11,000 photons MeV^{-1} , a detection limit of 26.2 nGy s^{-1} , and an afterglow duration exceeding 9 h.¹¹⁸ Zhang et al. further developed flexible afterglow films based on $\text{Dy}^{3+}/\text{Ag}^+$ co-doped $\text{Cs}_2\text{NaLuCl}_6$.¹¹⁵ Deposited onto highly curved three-dimensional surfaces via blade coating, these films enable X-ray imaging of nonplanar objects. Moreover, thermal stimulation after X-ray pre-irradiation allows faithful reproduction of stored structural information (Figure 5H). Notably, Tong et al. designed a core-shell $\text{Cs}_2\text{NaYCl}_6\cdot\text{Er}/\text{KBr}$ architecture that exploits the near-infrared emission of Er^{3+} , further illustrating the versatility of core-shell strategies in rare-earth double perovskite materials.¹¹⁹

Compared with low-dimensional rare-earth halides, A_2BREX_6 double-perovskite scintillators possess a three-dimensional framework that enables more efficient carrier transport and reduced exciton localization. The continuous lattice network facilitates long-range migration of photo-generated carriers, thereby increasing the probability of energy transfer toward luminescent activator centers. Meanwhile, the structural stability governed by tolerance and octahedral factors provides a robust platform for compositional alloying and dopant engineering. These structural characteristics modulate the competition between STE formation and activator-centered emission, which ultimately determines scintillation efficiency, decay dynamics, and detection sensitivity. Therefore, the A_2BREX_6 system exemplifies how lattice dimensionality and structural rigidity regulate carrier transport pathways and energy-transfer kinetics, establishing a direct structure-mechanism-performance relationship for high-efficiency X-ray scintillation.

Organic-inorganic Hybrid Rare-earth Halides

In recent years, metal-organic frameworks (MOFs), constructed through the coordination of inorganic metal nodes and organic bridging ligands, have emerged as structurally well-defined and rigid scaffolds for functional optoelectronic materials.^{120,121} Their ordered architectures allow the precise spatial integration of X-ray-absorbing centers with emissive units, making them attractive platforms for scintillation design.^{122–124} Along this direction, Wang et al. developed MOF-based scintillators by incorporating high-Z elements such as Hf and Zr into the framework, thereby enhancing X-ray absorption coupled with radioluminescence.¹²⁵ Beyond elemental incorporation, recent studies have increasingly exploited the structural versatility of MOFs to realize more refined energy management strategies. Liu et al. proposed a cluster sensitization concept by atomically integrating strongly X-ray-absorbing $[\text{Cu}_4\text{L}_4]$ clusters with lanthanide ions (e.g., $\text{Tb}^{3+}/\text{Eu}^{3+}$) within a MOF lattice (Figure 6A).¹²⁶ Upon X-ray excitation, the generated high-energy electron-hole pairs undergo cascade relaxation and populate hybrid excited states characterized by mixed halogen-ligand and metal-ligand charge-transfer components (Figure 6B). The rigid and spatially confined MOF environment suppresses vibrational dissipation, immobilizes lanthanide emitters, and reduces nonradiative losses, thereby facilitating efficient energy transfer from cluster-derived excitons to lanthanide centers and enhancing scintillation output. Building upon such framework-level energy control, Li et al. introduced the concept of radiative luminescence building blocks, in which deliberate electronic structure matching among organic ligands, Ln^{3+} ions, and high-Z constituents enables efficient ligand-to-lanthanide energy transfer while suppressing backward channels.¹²⁷ This design yields spectrally pure, characteristic Ln^{3+} emission with minimal interference. $\text{Tb}^{3+} \rightarrow \text{Eu}^{3+}$ energy transfer in this system exhibits pronounced thermal modulation, as elevated temperatures activate phonon-assisted transitions and gradually shift the

emission color from green to orange-red. The corresponding scintillation film maintains stable output up to 368 K and under high radiation doses. Extending MOF scintillator design into the compositional entropy regime, Yang et al. leveraged the comparable ionic radii and favorable mixing thermodynamics of lanthanide ions to construct a family of high-entropy lanthanide-organic frameworks (He-Ln-MOFs).³⁵ By incorporating five rare-earth ions (La, Ce, Eu, Dy, and Er) into a porous lattice, they formed the He-Ln-FA system (Figure 6C), which delivers a light yield of approximately 17,000 photons MeV^{-1} , a low detection limit of 302 nGy s^{-1} , and an exceptionally high uranium adsorption capacity of 1,532 mg g^{-1} . This material operates through a dual-mode mechanism. It enables ratiometric fluorescence detection of UO_2^{2+} under UV-visible excitation, whereas it produces a broadband scintillation signal mediated by the $\text{Ce}^{3+}/\text{Ce}^{4+}$ redox couple under radiation exposure, allowing robust and interference-resistant radiometric sensing (Figure 6D). In MOFs, X-ray energy conversion pathways can be constructed via directional energy transfer from ligand triplet excitons to lanthanide luminescent centers. Zhang et al. demonstrated that in the Tb -MOF-76 scintillator, the heavy-atom effect at the metal-molecule interface and strong electronic coupling generate a high density of electron-hole pairs upon X-ray absorption, which are subsequently converted into abundant triplet excitons.¹²⁸ These excitons, in synergy with carrier trapping processes, effectively activate the Tb^{3+} luminescent centers, leading to efficient radioluminescence under X-ray excitation. By precisely tuning the $\text{Tb}^{3+}/\text{Eu}^{3+}$ doping ratio, tunable multicolor emission was further achieved. Xu et al. proposed a strategy based on precise energy level alignment between ligands and lanthanide ions. By designing the ligand triplet energy level to be slightly higher than the emission level of the lanthanide ions, a unidirectional energy transfer pathway from ligand to lanthanide centers is established, effectively suppressing back energy transfer (Figure 6E).¹²⁹ This strategy enables over 99% of the X-ray excitation energy to be funneled through the ligand triplet exciton channel to the lanthanide centers, resulting in an enhancement of radioluminescence intensity by more than three orders of magnitude and a reduced detection limit down to 31 nGy s^{-1} . This provides a promising material platform for deep-tissue X-ray-mediated radiotherapy. Furthermore, based on a lanthanide-assisted triplet exciton cycling mechanism, an X-ray-mediated photodynamic therapy was developed (Figure 6F). Upon X-ray penetration into tissues, tumor-targeting nanomicelles are activated, and the near-unity energy transfer efficiency between ligands and lanthanides significantly enhances the emission intensity. Rose Bengal, serving as a photosensitizer, absorbs the emitted light to generate reactive singlet oxygen. The strong fluorescence signal induced thereby directly confirms the effective intracellular reactive oxygen species generation and tumor cell killing capability of this X-ray-mediated photodynamic therapy (Figure 6G).

Optical crosstalk arising from photon scattering remains a principal limitation to spatial resolution in X-ray imaging. To address this challenge, Liu et al. drew inspiration from the intrinsic optical transparency of water and developed a highly uniform and optically transparent liquid scintillator via in situ coordination between lanthanide ions and ionic liquid ligands.¹³⁰ By suppressing photon scattering, the liquid scintillator achieves a transmittance exceeding 93% and delivers a spatial resolution of 26 lp mm^{-1} at a thickness of 1 mm. Its inherent fluidity and shape adaptability further enable the fabrication of convex and zoomable scintillating lenses, broadening the functional scope of liquid scintillators. Han et al. introduced a structural-optical co-regulation strategy by incorporating ammonium salts and alcohols to induce local clustering and hydrogen bonding, which drives a dimensional reduction from three-dimensional lattices to lower-dimensional (0D/1D) architectures.¹³¹ Within these confined structures, spin-allowed 5d–4f transitions promote carrier transport, while edge- and face-sharing connectivity among $[\text{EuBr}_6]^{4-}$ octahedra effectively narrows the bandgap and enhances quantum confinement. Exploiting the solution processability of MeEu , a transparent AAO@MeEu composite film was fabricated by infiltrating the scintillator into an AAO template, where the aligned AAO nanochannels function as optical waveguides that confine emitted photons and markedly suppress lateral and edge scattering, enabling X-ray imaging with a spatial resolution exceeding 27 lp mm^{-1} (Figures 7A–C). Beyond liquid scintillators, highly transparent scintillating gels and glassy materials have attracted increasing attention. Zhao et al. prepared a europium-based transparent glass through a low-temperature dehydration process, in which Eu^{3+} and Cl^-

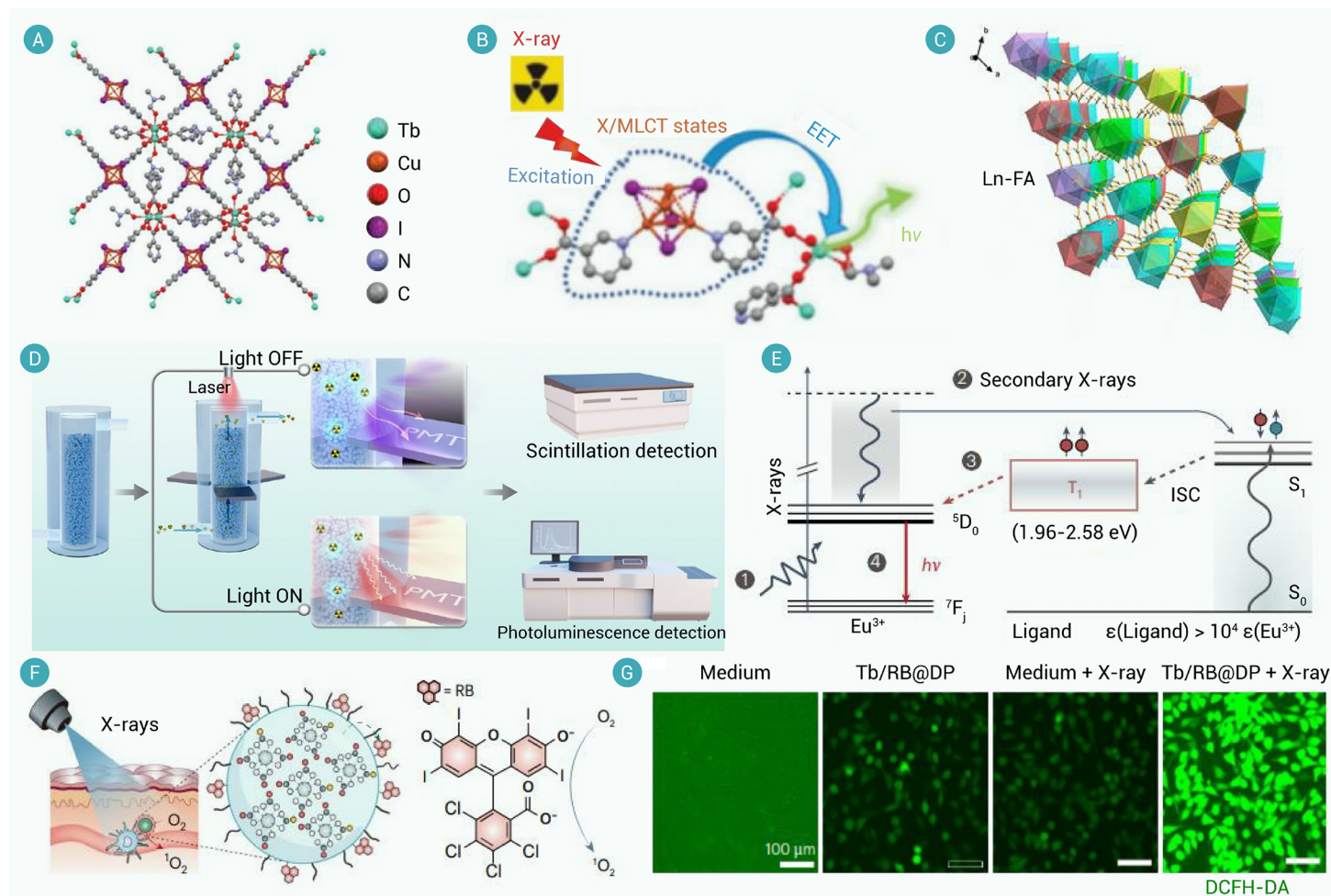


Figure 6. Scintillation mechanism and properties of organic-inorganic hybrid rare-earth halides (A–B) Crystal structure of TbCu_4I_4 MOFs and cluster-to- Tb^{3+} energy transfer under X-ray excitation. Copyright 2023, Wiley.¹²⁶ (C–D) High-entropy rare-earth MOFs and their dual-mode (photoluminescence/radioluminescence) detection mechanism. Copyright 2025, Wiley.³⁵ (E–G) Triplet exciton cycling in organic lanthanide molecules enabling X-ray-activated energy conversion for in vivo photodynamic therapy and cellular imaging. Copyright 2024, Springer Nature.¹²⁹

ions are immobilized within a high-viscosity amorphous network (Figure 7D).¹³² This structural confinement suppresses ion migration and crystallization while eliminating non-radiative losses associated with O–H vibrational modes, resulting in radioluminescence intensities that significantly exceed those of the corresponding crystalline phases (Figure 7E). Subsequent epoxy encapsulation further improves moisture resistance without compromising optical transparency. In parallel, Wang et al. fabricated a transparent $(\text{Bzmim})_3\text{TbCl}_6$ ($\text{Bzmim}^+ = \text{C}_{11}\text{H}_{13}\text{N}_2^+$) hybrid glass through a low-temperature melting process.¹³³ By tuning the rare-earth doping ratio, continuous radioluminescence modulation from green to red was achieved under X-ray excitation, enabling real-time visual radiation detection without external power input (Figures 7F–H). Lian et al. developed a water-assisted rapid evaporation strategy to synthesize hybrid glasses incorporating 16 lanthanide elements.¹³⁴ Their results revealed a triplet-exciton-mediated energy transfer mechanism, wherein heavy lanthanides promote intersystem crossing, and the triplet energy level of the organic cation $\text{C}_{22}\text{H}_{24}\text{P}^+$ ($\text{BuTPP}^+ = \text{C}_{22}\text{H}_{24}\text{P}^+$) is well matched to the excited states of Ln^{3+} emitters. Among these materials, $(\text{BuTPP})_3\text{TbCl}_6$ glass delivered a light yield of $8,100 \text{ photons MeV}^{-1}$ and retained 80% of its emission intensity after 30 days of air exposure, demonstrating excellent environmental stability. Complementarily, Wei et al. established a mild, solvent-free, rapid synthesis route for highly transparent rare-earth hybrid glasses with the general composition $\text{RE}(\text{NO}_3)_3(\text{C}_6\text{H}_2\text{N}_4)_2$.¹³⁵ The material exhibits an X-ray absorption coefficient 1.4 times higher than that of commercial LYSO scintillators and achieves a low detection limit of $1.87 \mu\text{Gy s}^{-1}$. Meanwhile, the Tb-based glass reaches a spatial resolution of 20 lp mm^{-1} , highlighting the great potential of rare-earth glasses for high-resolution X-ray imaging applications.

Overall, the scintillation properties of hybrid rare-earth halides arise from the cooperative interaction between inorganic metal nodes and organic ligands. The rigid framework structures suppress vibrational dissipation, while organic ligands provide additional energy-transfer pathways and structural flexibility. Such hybrid architectures enable precise regulation of exciton confinement, energy transfer, and photon propagation, which is beneficial for achieving high optical transparency and improved imaging resolution. Therefore, hybrid rare-earth halides offer a unique platform where structural modularity directly translates into tunable scintillation mechanisms and device-level imaging performance.

Rare-earth-doped Halides

With deeper insights into rare-earth ion doping, together with advances in tunable multicolor emission and quantum-cutting mechanisms, rational rare-earth incorporation effectively reduces lattice defects, suppresses unfavorable phase transitions, and enhances structural stability.^{79,90,136} Using a melt-quenching approach, Eu-doped CsPbBr_3 quantum dots can be grown in situ within a transparent oxide glass matrix, forming a glass-ceramic composite characterized by reduced particle size and improved spatial uniformity.¹³⁷ According to Rayleigh scattering theory, in which scattering intensity scales with the sixth power of particle size, such microstructural refinement markedly suppresses internal light scattering. Simultaneously, the glass matrix acts as a robust barrier against moisture ingress, elevated temperatures, and prolonged X-ray irradiation, thereby inhibiting lead leaching and improving radiation tolerance (Figure 8A), which collectively enhances imaging stability (Figure 8B). Quantum-cutting behavior of rare-earth ions further broadens the functional imaging applications. Yb^{3+} -doped CsPbCl_3 nanocrystals

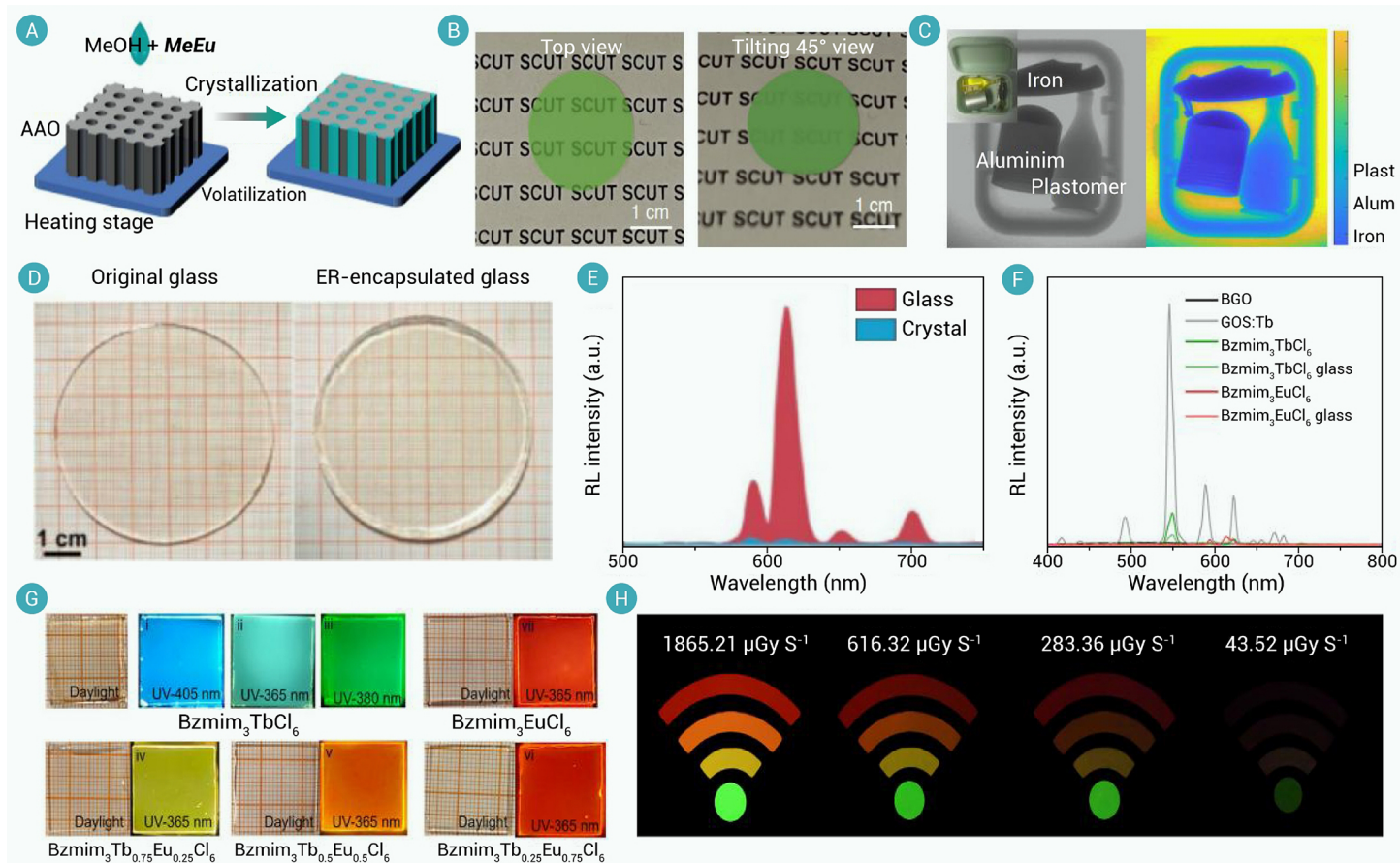


Figure 7. Organic-inorganic hybrid rare-earth halides for multi-scenario X-ray applications (A–C) Fabrication and imaging performance of AAO@MeEu scintillator films prepared via filtration-crystallization. Copyright 2024, Springer Nature.¹³¹ (D–E) Dehydration-induced transparency enhancement and corresponding radioluminescence improvement in Eu-based glass. Copyright 2025, Wiley.¹³² (F–H) Structure-dependent radioluminescence of Bzmm₃LnCl₆ (glass vs. crystalline states) and its dose-dependent scintillation imaging performance. Copyright 2025, Wiley.¹³³

tals exhibit photoluminescence quantum yields exceeding 100% and a pronounced Stokes shift from blue host emission to Yb³⁺-centered near-infrared emission, effectively suppressing interlayer crosstalk in multi-energy X-ray imaging.^{138,142} CsPbCl₃:Yb³⁺ is particularly well suited as the top scintillation layer in stacked detectors, where it preferentially absorbs low-energy X-rays and emits NIR photons. Optimization of layer thickness enables a balance between X-ray absorption efficiency and minimized interlayer interference (Figures 8C–E). Synergistic visible–NIR imaging strategies have also been realized. CsPbCl₃:Yb³⁺ single crystals reported by Zhu et al. exhibit strong resistance to ambient visible-light interference during NIR imaging, eliminating the need for light-tight encapsulation.¹³⁹ Ran et al. combined NIR-emitting CsPbCl₃:Yb³⁺ with visible-emitting CsCu₂I₃ to fabricate a dual-modal scintillation film spanning both spectral regions, enabling simultaneous multichannel signal acquisition (Figures 8F–H).³⁷ Lu³⁺-doped CsPbBr₃ nanocrystals embedded in a high-viscosity amorphous matrix undergo localized recrystallization upon thermal treatment, restoring scintillation performance via a self-healing mechanism (Figure 8I).¹⁴⁰ Furthermore, the lead-free halide Cs₂ZrCl₆:Lu³⁺ delivers a light yield as high as 94,190 photons MeV^{−1} under X-ray excitation and supports clear imaging under bright and low-light conditions, demonstrating its suitability for diverse detection environments (Figure 8J).¹⁴¹

In rare-earth-doped halide systems, the introduction of Ln³⁺ ions plays a dual role by modifying the host electronic structure and acting as efficient luminescence centers. Doping not only introduces additional energy levels that facilitate energy transfer and quantum-cutting processes but also improves structural stability and defect tolerance of the host lattice. Through rational dopant selection and concentration control, the balance between host emission, sensitization processes, and carrier recombination pathways can be precisely regulated, enabling enhanced light yield, spectral tunability, and multifunctional imaging capabilities.

CONCLUSION AND PERSPECTIVE

This review provides a systematic overview of the core applications and recent advances of rare-earth halide perovskite or perovskite-like structure scintillators in X-ray detection and imaging. Benefiting from their high light output, spectrally tunable emission, flexible fabrication routes, as well as pronounced structural diversity and favorable processability, these materials have substantially advanced X-ray detection toward high sensitivity and low-dose operation. As summarized in Table 1, many rare-earth halide scintillators exhibit scintillation performances comparable to those of conventional commercial scintillators. For example, all-inorganic 0D Rb₃TbCl₆ single crystals exhibit light yields up to 88,800 photons MeV^{−1}, while 3D Cs₂NaTbCl₆:Ce³⁺ microcrystals deliver light yields of 53,200 photons MeV^{−1} with detection limits as low as 28.8 nGy s^{−1}. From an imaging perspective, materials such as AAO@MeEu composite films and Bzmm₃TbCl₆ hybrid glass demonstrate spatial resolutions exceeding 20 lp mm^{−1}, meeting the requirements for high-resolution X-ray imaging. Table 1 also summarizes the representative preparation strategies for these materials, highlighting that the synthesis routes are closely related to the structural dimensionality and targeted device architectures. As summarized in Table 1, the fabrication strategies of rare-earth halide scintillators show a clear dependence on their structural dimensionality. Three-dimensional frameworks with extended corner- or edge-sharing octahedral networks generally require crystal-growth approaches such as Bridgman growth, melting, or evaporation crystallization to maintain long-range structural order and minimize defect formation. These methods enable the preparation of highly crystalline materials with efficient X-ray absorption and carrier transport, which are advantageous for achieving high scintillation efficiency. However, the stringent growth conditions, including elevated temperatures and long crystallization times, limit their scalability and large-area device fabrication. In contrast, lower-dimensional structures, such as zero-dimensional isolated octahedra, one-dimensional chains, or two-dimen-

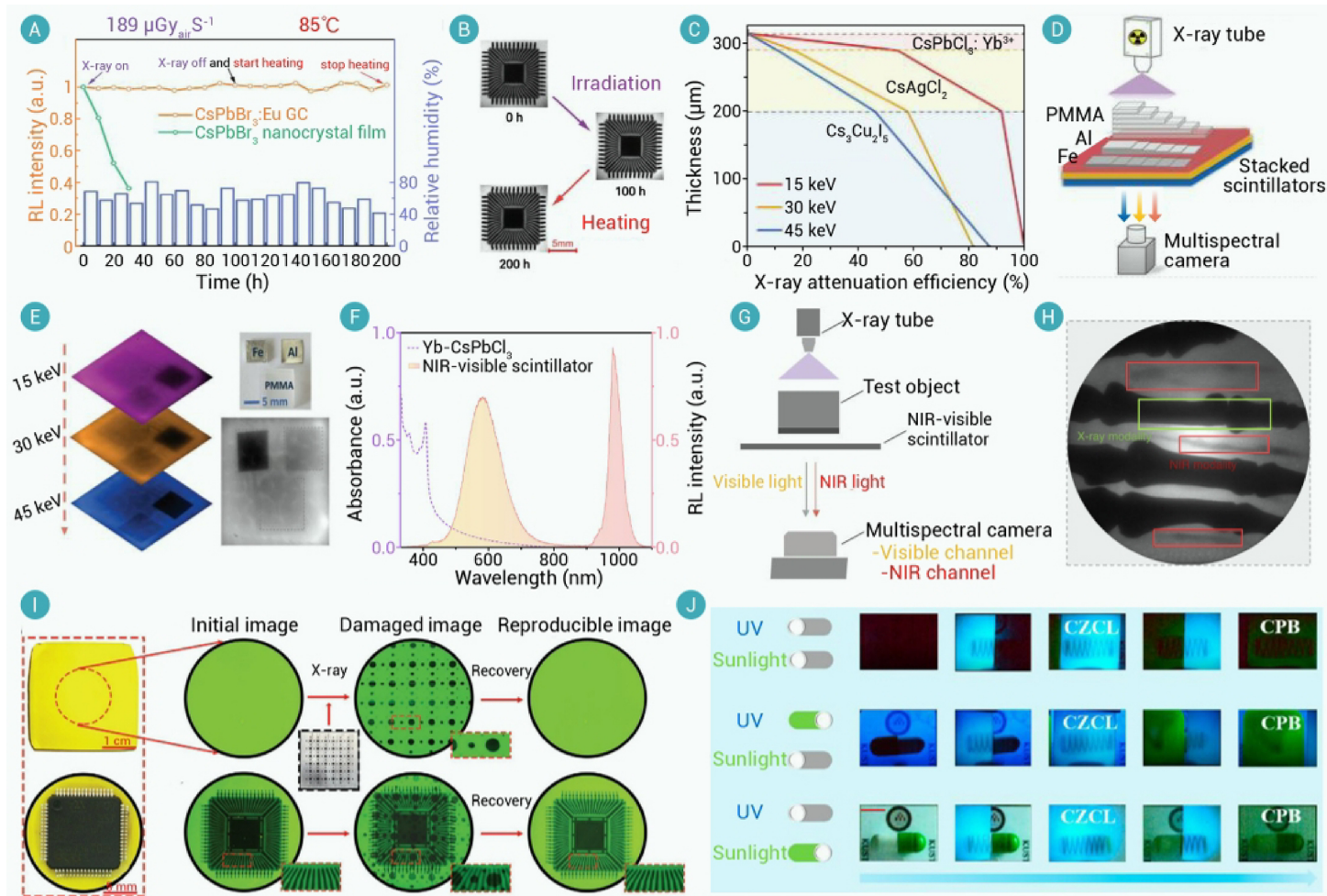


Figure 8. Rare-earth-doped halides for multi-scenario X-ray applications (A–B) X-ray and environmental stability, and imaging durability of CsPbBr₃:Eu glass ceramics. Copyright 2021, Wiley.¹³⁷ (C) The correlation between thickness and absorption efficiency at varying X-ray energies, with data derived from XCOM. (D–E) Multispectral imaging system based on stacked scintillators for material identification. Copyright 2025, Wiley.¹⁴² (F–H) Near-infrared/visible dual-emission scintillator (CsPbCl₃:Yb/CsCu₂I₃) demonstrating spectrally separated radioluminescence and dual-mode X-ray imaging for simultaneous visualization of bones (X-ray mode) and blood vessels (near-infrared mode). Copyright 2025, Springer Nature.³⁷ (I) X-ray irradiation damage and recovery process in CsPbBr₃:Lu³⁺. Copyright 2021, Wiley.¹⁴⁰ (J) Application of the solar-blind characteristic of Cs₂ZrCl₆:Lu³⁺ in X-ray imaging. Copyright 2021, American Chemical Society.¹⁴¹

sional layered motifs, can often be obtained through solution-based routes, including solvothermal synthesis, coprecipitation, or hot-injection methods. These approaches provide milder reaction conditions and flexible compositional tunability, while also facilitating the integration of the resulting materials into scintillation layers through solution processing. Nevertheless, the reduced structural connectivity in low-dimensional systems may lead to stronger exciton localization and increased optical scattering in polycrystalline films, which can influence carrier transport and spatial resolution in imaging devices. Collectively, these results demonstrate that rational control over structural dimensionality, dopant configuration, hybrid design, and fabrication strategy enables rare-earth halide scintillators to achieve high emission efficiency, low detection limits, and excellent imaging resolution. Despite the significant progress achieved to date, several challenges remain to be addressed in the future, and future research efforts should focus on the following directions.

(1) *Decay lifetime control and compatibility with dynamic imaging.* Defect states, deep traps, and self-trapped exciton processes are commonly present in rare-earth halides, leading to delayed carrier recombination, millisecond-scale decay components, or persistent afterglow. These effects cause signal accumulation and motion blur under high-frame-rate imaging conditions. Improving compatibility with dynamic X-ray imaging requires shifting the dominant emission pathway toward parity-allowed 5d–4f transitions (e.g., Ce³⁺ or Eu²⁺ activated systems) and suppressing self-trapped exciton formation through structural and defect engineering. For example, Samanta et al. reduced the Cs₃TbCl₆ lifetime from 8.5 ms to 7.27 ns by doping sensitizers

for material modification.⁹¹ In addition, core-shell or surface-passivated architectures, inspired by composite fluoride scintillators, offer an effective strategy to suppress surface-defect-induced nonradiative relaxation while simultaneously enhancing emission efficiency and response speed.¹¹⁹ It should be emphasized that translating intrinsically fast scintillation into practical dynamic imaging performance requires coordinated optimization of scintillator response, detector readout speed, and device architecture.^{167,168}

(2) *Low-cost scalable synthesis and valence-state control.* Current synthesis of inorganic rare-earth halide scintillators relies heavily on solvothermal or hot-injection methods, which typically require elevated temperatures and strictly oxygen-free environments, thereby limiting scalability and processing flexibility. In addition, commonly used precursors such as REX₃ exhibit strong hygroscopicity and limited thermal stability, further increasing the difficulty of material preparation and large-scale production. Therefore, the development of low-temperature, rapid, and scalable synthetic routes is crucial for translating these materials from laboratory studies to practical applications. Recent reports on coprecipitation-based synthesis of Cs₃LnCl₆ and antisolvent-assisted preparation of Cs₂NaLnCl₆ provide promising strategies for room-temperature, large-scale fabrication.⁹⁵ However, even if scalable synthesis can be achieved at the material level, converting these high-performance materials into large-area and uniform scintillation screens remains a significant challenge.^{12,13} The fabrication of thick-film or bulk scintillation layers typically requires precise control over crystal growth kinetics, particle size distribution, and microstructural morphology, as structural inhomogeneity or irregular crystallization can introduce additional optical scattering and

Table 1. Scintillation performance of rare-earth halide perovskite-like scintillators.

Material	Synthesis method	Emission peak (nm)	Decay time	Light yield (photons MeV ⁻¹)	Detection limit (nGy s ⁻¹)	Resolution (lp mm ⁻¹)	Ref.
NaLuF ₄ :Tb ³⁺ @NaYF ₄	epitaxial growth	546	> 30 days	/	/	> 20	32
NaYF ₄ :Tb ³⁺ @NaYF ₄	epitaxial growth	545	> 1,800 s	/	/	/	143
NaYF ₄ :Nd ³⁺ @NaYF ₄	epitaxial growth	862	> 1,800 s	/	/	/	
NaGdF ₄ :Eu ³⁺ @NaGdF ₄ :Ce ³⁺	epitaxial growth	616	/	/	164.56	11	70
NaLuF ₄ :Gd ³⁺	thermal decomposition	314	7 days	/	/	20	75
NaLuF ₄ :Mn ²⁺ ,Gd ³⁺	thermal decomposition	512	30 days	/	/	22	74
LiYbF ₄ :Tb ³⁺	thermal decomposition	544	4.794 ms	/	360	20	81
LiLuF ₄ :Tb ³⁺	thermal decomposition	544	5.466 ms	13,015	36.31	20	29
LiLuF ₄ :Tb ³⁺ ,Zn ²⁺	hydrothermal	544	/	37,241	3.83	35	144
KLaF ₄ :Gd ³⁺ ,Ce ³⁺	melt-quenching	380	53.6 ns	17,000	1.8×10 ⁴	/	145
LiLuF ₄ :Tb ³⁺ ,Cu ²⁺	solvothermal	544	> 48 h	28,902	2.7928	22	82
Cs ₃ CeCl ₆ ·3H ₂ O	hot injection	373	~ 32 ns	31,900	108	~ 12	146
Cs ₃ CeBr ₆	hot injection	390	29.6 ns	/	11.2	12.21	33
Cs ₃ EuCl ₆	solid-state sintering	614	4.11 ms	12,877	643.1	9.17	93
Rb ₃ TbCl ₆	hydrothermal	548	4.38 ms	88,800	115.38	3.9	92
Cs ₃ TbCl ₆	anti-solvent	547	6.8 ms	51,800	63	12	95
Cs ₃ TbCl ₆ :Sb ³⁺	thermal decomposition	548	7.6 ms	20,300	212	9.6	96
CsLu ₂ F ₇ :Tb ³⁺	co-precipitation	542	3.9 ms	11,525	480	30.5	147
CsLu ₂ F ₇ :Eu ³⁺	co-precipitation	611	4.2 ms	11,915	1930	25	
Cs ₂ NaScCl ₆ :Tb ³⁺	solvothermal	548	7.45 μs	/	/	/	148
Cs ₂ NaScCl ₆ :Sm ³⁺	solvothermal	448	/	18,887	76.51	11.9	
Cs ₂ NaScCl ₆ :Nd ³⁺	solvothermal	1072	/	/	/	24	149
Cs ₂ NaYCl ₆	hot injection	461	1.32 μs	/	/	/	150
Cs ₂ NaYCl ₆ :Sb ³⁺	hot injection	461	/	22,500	143	2.04	
Cs ₂ NaGdCl ₆	solvothermal	470	2.55 μs	/	/	2.93	108
Cs ₂ NaGdCl ₆ :Tb ³⁺	solvothermal	549	4.2 ms	39,100	41.32	10.75	
Cs ₂ NaTbCl ₆	hot injection	548	5 ms	14,800	681	4.4	106
Cs ₂ NaTbCl ₆ :Sb ³⁺	hot injection	548	7.5 ms	23,500	140	7.9	
Cs ₂ NaTbCl ₆	solvothermal	548	6.24 ms	93,105	255.19	17	151
Cs ₂ NaTbCl ₆ :Ce ³⁺	solvothermal	548	5.69 ms	52,153	30	10	104
Cs ₂ NaTbCl ₆	anti-solvent	548	6.4 ms	18,700	/	/	105
Cs ₂ NaTbCl ₆ :Ce ³⁺	anti-solvent	548	6.4 ms	53,200	28.8	31	
Cs ₂ NaEuCl ₆	solvothermal	593	2.61 ms	4,400	/	/	
Cs ₂ NaHoCl ₆	solvothermal	657	2.04 ms	9,600	/	/	
Cs ₂ NaErCl ₆	solvothermal	660	3.45 ms	3,300	/	/	51
Cs ₂ NaTmCl ₆	solvothermal	806	560 μs	1,400	/	/	
Cs ₂ NaYbCl ₆	solvothermal	996	2.73 ms	6,000	/	/	
Cs ₂ NaYbCl ₆ :Er ³⁺	solvothermal	663	6.88 ms	16,000	144.9	12	152
Cs ₂ LiGdCl ₆ :Sb ³⁺	hot injection	528	2.44 μs	20,800	94	/	
Cs ₂ NaGdCl ₆ :Sb ³⁺	hot injection	474	1.47 μs	6,200	236	/	111
Cs ₂ KGdCl ₆ :Sb ³⁺	hot injection	522	2.31 μs	39,900	86	~ 15	

(Continued)

Table 1. Scintillation performance of rare-earth halide perovskite-like scintillators.

Material	Synthesis method	Emission peak (nm)	Decay time	Light yield (photons MeV ⁻¹)	Detection limit (nGy s ⁻¹)	Resolution (lp mm ⁻¹)	Ref.
Tb-Cu ₄ I ₄	solvothermal	547	1.52 ms	29,379	45.2	12.6	126
Tb-Eu-Cu ₄ I ₄ MOFs (4:1)	solvothermal	547	387.4 μs	12,707	/	/	
Tb-HNIC	solution method	547	908 μs	11,513	/	/	
Eu-pba	solvothermal	615	430 μs	/	4.92×10 ³	8	153
Tb-pba	solvothermal	543	750 μs	/	3.17×10 ³	10	
Tb-MOF-76	solvothermal	546	1.4 ms	48,221	23	16.6	128
Eu-MOF-76	solvothermal	616	710 μs	3901	/	/	
Tb-TPC	solvothermal	543	134 μs	5,453	1.114×10 ⁴	4.46	154
Eu-TPC	solvothermal	619	463 μs	6,121	5.2×10 ³	5.15	
Tb-BPTC	solvothermal	544	/	38,800	156.1	18	127
Eu-BPTC	solvothermal	615	/	6,500	/	8	
[Me ₄ N] ₆ Eu ₅ Br ₁₆ ·MeOH	solvent evaporation	500	368.2 ns	73,100	18.6	16.2	131
[Et ₄ N]EuBr ₃ ·MeOH	solvent evaporation	450	387.1 ns	32,200	69.3	/	
Ce(Tp) ₃	co-precipitation	430	38 ns	5,177	/	/	129
Eu(TpCF ₃) ₂	co-precipitation	490	506 ns	8,650	/	/	
Eu(NTA) ₃ DPEPO	co-precipitation	~ 615	621 μs	38,589	20	/	
Tb(CPMIP) ₃ (TPPO) ₂	co-precipitation	~ 545	/	17,251	/	/	155
Eu(TTA) ₃ Phen	co-precipitation	612	180 μs	24,877	19.97	23	
(MTP) ₃ EuCl ₆ glass	melt slow-cooling	592	118.48 μs	7,820	95.8	14.3	
(BuTPP) ₃ TbCl ₆ glass	melt slow-cooling	549	2.41 ms	8,100	930	26.8	134
RECl ₃ (ETPCl) ₂	melt slow-cooling	613	0.967 ms	8,600	/	8	156
Bzmim ₃ TbCl ₆	ionothermal	547	2.81 ms	14,839	/	/	133
Bzmim ₃ TbCl ₆ glass	melt slow-cooling	547	2.47 ms	4,101	1.56×10 ³	25	
Bzmim ₃ EuCl ₆	ionothermal	611	5.99 ms	4,879	/	/	
Bzmim ₃ EuCl ₆ glass	melt slow-cooling	611	3.57 ms	1,419	308.3	/	157
(R-3BrMBA) ₃ TbCl ₆	anti-solvent	549	4.95 ms	/	50.3	/	
(S-3BrMBA) ₃ TbCl ₆	anti-solvent	549	4.89 ms	/	/	/	
PDMIMEuBr ₃	solvent evaporation	513	603.87 ns	89,380	42.61	21.1	158
Tb(IL) ₃	solvent evaporation	548	686.67 μs	2,771	21.48	26	130
(La, Ce, Eu, Dy, Er)-FA	solvothermal	617	59.05 ns	17,400	302.4	/	35
(La, Ce, Eu, Dy, Er)-SA	solvothermal	/	57.58 ns	8,200	836	/	
(La, Ce, Eu, Dy, Er)-NTA	solvothermal	616	9.95 ns	7,600	960	/	
[PP14]EuBr ₃ MeOH	solvent evaporation	495	370.8 ns	24,000	/	/	159
[PP13]EuBr ₃ MeOH	solvent evaporation	490	312.1 ns	19,000	/	/	
[Emim]EuBr ₃ MeOH	solvent evaporation	505	382.3 ns	43,000	405.4	10	
CsPbBr ₃ :Eu ³⁺	melt-quenching	616	6.78 ns	10,100	/	15	137
CsPbBr ₃ :Ce ³⁺	hot injection	~ 485	7.09 ns	33,000	/	580	160
CsPbCl ₃ :Yb ³⁺	solvothermal	980	/	112,281	176.5	/	36
CsPbBr ₃ :Gd ³⁺	melt-quenching	520	44 ns	/	/	12.7	161
CsPbBr ₃ :Lu ³⁺	melt-quenching	516	27 ns	7,820	50	16.8	140
Cs ₂ ZrCl ₆ :Lu ³⁺	solvothermal	460	/	94,190	15.8	16.6	141

(Continued)

Table 1. Scintillation performance of rare-earth halide perovskite-like scintillators.

Material	Synthesis method	Emission peak (nm)	Decay time	Light yield (photons MeV ⁻¹)	Detection limit (nGy s ⁻¹)	Resolution (lp mm ⁻¹)	Ref.
Cs ₂ Ag _{0.6} Na _{0.4} In _{0.95} Bi _{0.05} Cl ₆ : Tm ³⁺	solvothermal	800	2.56 ms	33,500	55.2	11.2	162
CsCaCl ₃ :Eu ²⁺	melt-quenching	434	991 ns	9,300	213.8	30.0	163
CsSrCl ₃ :Eu ²⁺	melt-quenching	428	820 ns	11,000	151.7	/	
Cs ₂ BaCl ₄ :Eu ²⁺	melt-quenching	422	643 ns	8,600	305.4	/	
Cs ₃ Cu ₂ I ₅ :Eu ²⁺	bridgman	460	1,119 ns	55,000	91.1	23	164
CsPbCl ₃ :Yb ³⁺	hot injection	983	/	36,740	/	13.6	142
CsPbCl ₃ :Yb ³⁺	solvent evaporation	980	/	31,223	100	14.1	37
CsPbI ₃ :Sc ³⁺	melt-quenching	~ 720	37.67 ns	5,572	/	17	165
Cs ₂ ZrCl ₆ :Sb ³⁺ ,Nd ³⁺	co-precipitation	450	13.74 μs	13,300	/	5.77	166

lateral photon diffusion, thereby degrading imaging resolution and device reproducibility. Consequently, the development of scalable synthesis strategies together with controllable deposition techniques for constructing uniform and high-density scintillation layers is an essential prerequisite for practical device implementation.¹⁶⁹ With respect to valence-state control, most existing studies focus on trivalent rare-earth ions, whereas divalent and tetravalent rare-earth systems remain relatively underexplored. Compounds such as CsEuBr₃, Cs₄EuBr₆, and Cs₄EuI₆ exhibit blue emission, and Cs₄EuBr₆ and Cs₄EuI₆ achieve light yields of 78,000 and 53,000 photons MeV⁻¹ under γ-ray excitation, respectively.^{97,170} In addition, Li et al. developed a series of organic Eu²⁺-based halide crystals exhibiting efficient 5d-4f emission.¹⁵⁸ In Ce-doped Cs₂NaTbCl₆, energy transfer between Ce³⁺ and Ce⁴⁺ states enhances emission intensity by nearly 40-fold.¹⁷¹ These studies suggest that expanding the accessible valence states of rare-earth ions and constructing mixed-valence systems could provide new opportunities to modulate energy-transfer processes and further improve scintillation performance, offering an important direction for the development of next-generation rare-earth halide scintillators.

(3) *Optimizing material stability and dopant-mediated energy transfer through intelligent design.* Excessive dopant concentrations can induce cross-relaxation and other nonradiative energy-transfer pathways, while also introducing lattice strain and structural defects that degrade emission efficiency and radiation stability. Addressing these challenges requires the integration of advanced characterization techniques with machine-learning-assisted approaches.¹⁷²⁻¹⁷⁴ Such strategies enable efficient analysis of large experimental datasets, facilitate the tracking of defect evolution and carrier dynamics, and improve the predictive accuracy of compositional and structural optimization.¹⁷⁵ Meanwhile, rare-earth halide scintillators are generally susceptible to environmental factors such as humidity, oxygen, and temperature fluctuations, as well as prolonged exposure to high-energy radiation, which can lead to structural defects or trap states, chemical degradation, and luminescence quenching during long-term operation.¹⁷⁶ Therefore, improving material stability is equally critical for the practical deployment of rare-earth halide scintillators. Current mainstream strategies typically rely on surface passivation or protective encapsulation.¹³² However, compared with external encapsulation, stabilization strategies based on intrinsic structural regulation are of greater long-term significance. Artificial-intelligence-assisted materials design provides new opportunities for exploring effective structural regulation pathways, including: (i) dimensional engineering (e.g., constructing 0D or 3D frameworks) to introduce intrinsic diffusion barriers that suppress moisture penetration,^{34,61} (ii) lattice densification and coordination saturation to minimize under-coordinated sites and defect densities, thereby reducing water adsorption and chemical reactivity,⁴⁴ (iii) strengthening RE-X bond stability through hard-halide selection or mixed-halide strategies, together with the rational utilization of internal hydrogen-bonding networks to mitigate water-induced degradation.^{134,156,177} Nevertheless, these structural engineering strategies often involve inherent trade-offs with carrier transport, scintillation efficiency, or thermal stability. Consequently, the development of

stable rare-earth halide scintillators should be guided by application-specific requirements, seeking an optimal balance among structural robustness, scintillation performance, and manufacturability. In addition, a deeper understanding of energy-transfer pathways and charge-transport processes under X-ray excitation, particularly when combined with data-driven modeling, will further promote the rational design and accelerated discovery of next-generation rare-earth halide scintillators.

(4) *Development of highly transparent rare-earth halide scintillators.* Further development of rare-earth glass scintillators is constrained by the insufficient co-optimization of processing and performance. Conventional rare-earth oxide glasses typically require high-temperature melting, resulting in high energy consumption and limited compositional flexibility. Recently, rare-earth halide glasses, such as MTP₃EuCl₆ and (BuTPP)₃LnCl₆, have been reported. Notably, C₂₀H₂₀PCI (ETPCI) can form glassy phases at low temperatures, either independently or in combination with PbCl₂, suggesting that triphenylphosphonium-based organic cations can actively participate in glass formation.¹⁷⁸ This provides a viable low-temperature route toward hybrid rare-earth halide glasses. Meanwhile, rare-earth glass scintillators generally face a trade-off between light yield and structural stability, and rational exploitation of glass-crystal transformation offers an effective strategy to optimize both stability and overall performance. For instance, Wang et al. enhanced stability and suppressed crystallization by tailoring triphenylphosphonium cation structures.¹⁷⁹ Wen et al. employed quartz-glass encapsulation and waterproof coatings to suppress glass-crystal phase transitions, improving stability by more than two orders of magnitude.¹⁸⁰ Xiong et al. further demonstrated in situ crystallization to form glass-ceramic scintillators, increasing the light yield from 2,190 to 56,486 photons MeV⁻¹ while maintaining high optical transparency.¹⁸¹ Systematic studies on low-temperature glass-formation mechanisms and local rare-earth coordination environments will be essential for advancing high-performance rare-earth halide glass scintillators.

(5) *Expansion toward multimodal and spectral imaging.* With growing demand for high-resolution, color, and energy-resolved imaging, researchers have introduced spectral information through detector architecture design and signal modulation strategies. For example, Shao et al. achieved indirect dual-energy imaging by integrating bilayer scintillators with optical filters.¹⁸² The authors further proposed an energy-loss-residual-energy telescope architecture composed of six stacked detection units, in which detector-level energy modulation enables the extension of conventional grayscale imaging to color imaging.¹³⁸ In addition, Fan et al. developed a rare-earth-doped oxide scintillator thin film stack based on narrowband emission of rare earths, successfully achieving dual energy imaging of ultra-narrowband scintillators.¹⁸³ Benefiting from their high effective atomic numbers, strong X-ray absorption, and rich, tunable emission manifolds, rare-earth halide materials offer distinct advantages for color and multi-energy X-ray imaging. Recently, Song et al. reported heterostructured scintillators with broadband emission, and Wang et al. developed multicolor rare-earth halide glass systems.^{34,133} Looking forward, the intrinsic emission tunability and structural

design flexibility of rare-earth halides position them as promising candidates for color and energy-resolved X-ray imaging, with significant potential in medical diagnostics and industrial computed tomography.

(6) *Toward unified radiation performance testing standards.* In scintillator-based X-ray detection, key performance metrics, such as light yield, detection limit, and spatial resolution, are often evaluated under variable experimental conditions and sample geometries, which complicates direct, quantitative comparisons across studies.¹⁸⁴ Light yield is typically inferred from relative radioluminescence intensity, but this estimate is highly sensitive to extrinsic factors, including sample size and morphology, transparency, and self-absorption.^{185,186} The detection limit, which is associated with light yield, further depends on X-ray tube voltage, beam filtration, and the accuracy of dose-rate calibration.^{184,187,188} In practical imaging configurations, scintillators are coupled with high-resolution thin-film transistor or complementary metal-oxide-semiconductor arrays, so the measured spatial resolution is influenced by both material properties and device architecture, including optical coupling and crosstalk.¹⁸⁹⁻¹⁹¹ Accordingly, rigorous, transparent, and standardized reporting of sample characteristics and measurement conditions is essential. Only within a controlled and reproducible evaluation framework can meaningful comparisons be achieved among rare-earth halide perovskite scintillators and other emerging scintillator materials, ultimately accelerating the translation of laboratory research into high-performance X-ray imaging technologies.

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FUNDING AND ACKNOWLEDGMENTS

This work was supported by the National Natural Science Foundation of China (No. 62541508, 52572168), National Key Research and Development Program of China (No. 2022YFC3700801), Key R&D Program of Shandong Province, China (No. 2024CXGC010302), Young Taishan Scholars Program of Shandong Province (No. tsqn202312213), and Shandong Provincial Natural Science Fund for Excellent Young Scientists Fund Program (Overseas, 2025HWYQ-049). The funders had no role in study design, data collection and analysis, decision to publish, or preparation of the manuscript.

AUTHOR CONTRIBUTIONS

N. Wang collected the literature necessary for this review and wrote the original manuscript. Y. Zhao and G. Zhou collected the necessary literature. B. Li and S. Huang revised the manuscript. Y. Zhou and B. Cao supervised all the tasks and revised the manuscript. All authors contributed to the article and approved the submitted version.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AND CODE AVAILABILITY

No new data and code were created or analyzed in this review.