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Research Article

Defect-engineered Ce³⁺-doped borosilicate glass scintillator for multifunctional and environment-adaptive X-Ray imaging

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Abstract: Advanced scintillation materials capable of simultaneously delivering ultrahigh efficiency, exceptional stability, and multifunctional X-ray imaging remain elusive because of intrinsic trade-offs in conventional scintillators. Herein, a defect-engineered Ce^{3+} -activated borosilicate glass was developed. It exhibits high internal/external quantum efficiencies of 92.4%/85%, an ultrafast lifetime of 142.3 ns, and XEL intensity further reaching 190% of BGO after “bright-burning effect”, setting a new performance benchmark for Ce^{3+} -activated glass scintillators. The glass combines remarkable thermostability, superior environment durability and outstanding long-term radiation tolerance. Furthermore, the formation of irradiation-induced Frenkel defects enables the unprecedented integration of efficient scintillation and persistent information retention into a single material. Consequently, the glass supports high-resolution multifunctional X-ray imaging across real-time (22.1 lp/mm), time-lapse (12 lp/mm), dynamic, high-temperature, and underwater modes. This work advances Frenkel defect engineering in amorphous scintillators and provides a viable route toward next-generation intelligent X-ray imaging under complex conditions.

Keywords: glass; Ce^{3+} ; luminescence; scintillator; X-ray imaging

1 Introduction

Scintillators are indispensable radiation-conversion materials underpinning X-ray detection technologies, with extensive applications in security screening, medical CT/PET imaging, industrial non-destructive testing, and high-energy physics detection.[1-8] Driven by the rapid advancement of imaging systems toward large-area integration, real-time acquisition, dynamic operation, and harsh-environment deployment, the requirements for scintillator performance have expanded far beyond conventional metrics.[9-11] High light yield and spatial resolution fundamentally remain the primary determinants of detection sensitivity and imaging fidelity. Meanwhile, emerging imaging modalities increasingly require the simultaneous optimization of environmental resilience, temporal response, and information-retention capability (Fig. 1).[12] Specifically, thermal and chemical robustness dictate operational reliability in high-temperature and high-humidity conditions.[13-15] Fast scintillation kinetics help suppress motion-induced artifacts during dynamic imaging.[16, 17] In addition, defect-mediated carrier trapping opens opportunities for time-lapse imaging.[18, 19] Accordingly, developing multifunctional scintillators combining high efficiency, ultrahigh imaging fidelity, rapid response, long-term durability, controllable energy-storage behavior, and scalable manufacturability has become a critical challenge for both radiation detection and materials science.

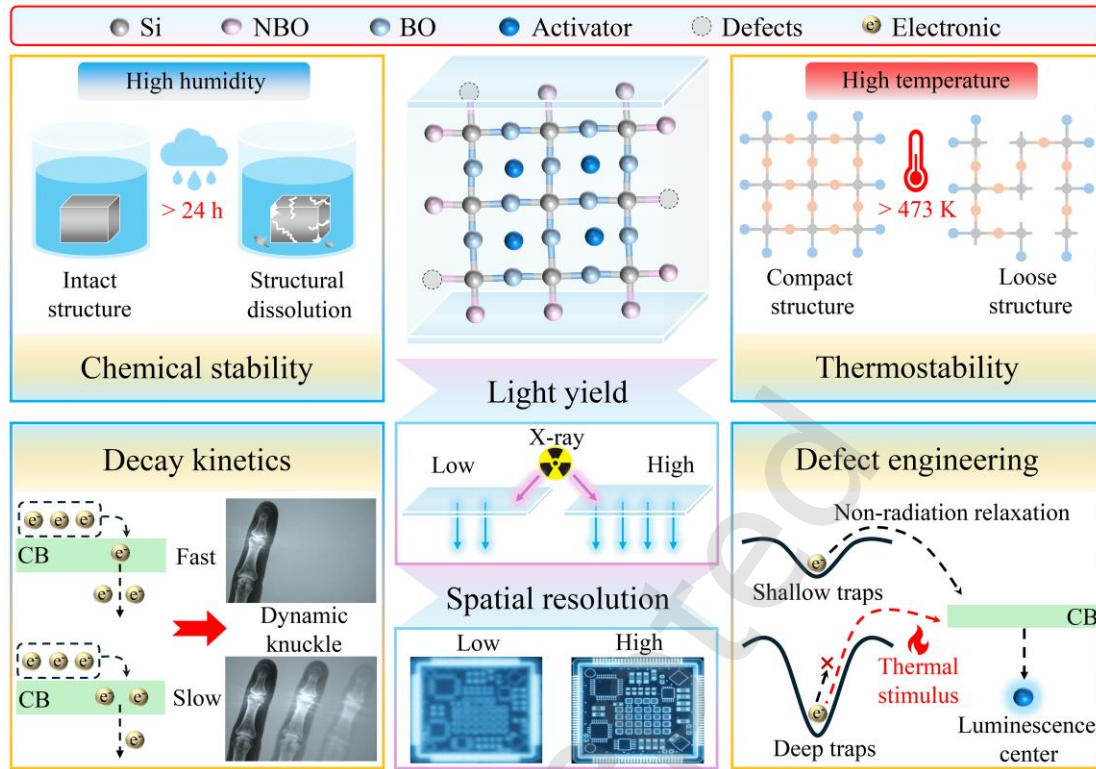


Fig. 1 Schematic overview of application-driven performance requirements.

To meet these demands, extensive efforts have been devoted to inorganic crystals, organic and hybrid scintillators, and halide perovskites.[20-22] Despite substantial progress, each class suffers from intrinsic limitations. Crystalline scintillators (e.g. BGO, CsI:Tl, and YAG:Ce) rely on energy-intensive crystal growth and complex processing, limiting cost-effectiveness and scalability for large-area fabrication.[22] Organic and MOF-based systems generally display inferior thermal and radiation stability.[23, 24] While halide perovskites continue to face challenges associated with environmental instability and toxicity concerns.[21, 25] Consequently, these materials struggle to fully satisfy the requirements of versatile imaging under increasingly demanding operational conditions. Among emerging alternatives, glass scintillators have attracted considerable attention as a promising platform due to their compositional

flexibility, crystal-growth-free fabrication, low-cost melt processing, excellent scalability, and superior physicochemical stability.[26-28] Although their performance is constrained by the disordered amorphous network that introduces abundant nonradiative defects and hampers carrier utilization efficiency,[29] such structural flexibility simultaneously provides opportunities for coordinated regulation of luminescent centers and functional defect states. This characteristic renders glass scintillator a viable candidate for integrating efficient scintillation, environmental durability, and multifunctional imaging within a single material system.

Herein, we develop a high-performance Ce^{3+} -activated oxyfluoride borosilicate glass scintillator by reduction-assisted defect engineering. The combination of parity-allowed Ce^{3+} emission, fluoride-mediated phonon suppression, and a robust borosilicate framework concurrently promotes efficient radiative recombination and exceptional environmental resilience. Moreover, X-ray-induced Frenkel defects provide effective carrier trapping while retaining strong X-ray excited luminescence (XEL), extending functionality beyond conventional real-time detection. As a result, the optimized composition delivers IQE of 92.4% and EQE of 85%, with XEL intensity reaching up to 190% of BGO after “bright-burning effect”, representing state-of-the-art performance among Ce^{3+} -activated glass scintillators. Its rapid scintillation response and negligible room-temperature afterglow further enable high-fidelity imaging of transient events with minimal artifacts. Coupled with defect-enabled carrier storage, the material affords multifunctional high-resolution X-ray imaging including real-time (22.1 lp/mm), time-lapse (12 lp/mm), dynamic, high-temperature and underwater

modes. This work establishes a general design strategy for integrating high scintillation efficiency, environmental robustness, and defect-enabled functionality in glass systems, paving the way toward next-generation intelligent X-ray imaging.

2 Experiment

G- x Ce- y Al and host samples were synthesized via melt quenching method. More details are presented in Electronic Supplementary Material.

3 Results and discussion

3.1 Structure and PL properties

X-ray diffraction (XRD) patterns of G-1Ce- x Al samples reveal a typical broad diffuse halo without any discernible crystalline reflections (Fig. 2(a)), indicative of a fully amorphous structure. Four absorption bands centered at 565, 690, 949, 1284 and 1382 cm^{-1} appear in Fourier transform infrared (FTIR) (Fig. 2(b)), attributable to Al-F vibrations, asymmetric Si-O-Al stretching in $[\text{AlO}_4]/[\text{SiO}_4]$ tetrahedra, symmetric Si-O-B stretching of $[\text{SiO}_4]/[\text{BO}_4]$ units, and B-O stretching modes of $[\text{BO}_3]$ groups.[30, 31] Consistently, Raman spectra (Fig. S1) display two broad envelopes situated at 501 and 1079 cm^{-1} , which are ascribed to the bending modes of four-membered $[\text{SiO}_4]$ rings and the symmetrical stretching vibrations of Si-O in $[\text{SiO}_4]$ tetrahedra, respectively.[26, 32] Notably, no detectable shifts are discerned in FTIR or Raman features following CeF_3 or Al incorporation. Combined with XRD results, these observations demonstrate that the amorphous glass network structure remain essentially unchanged.

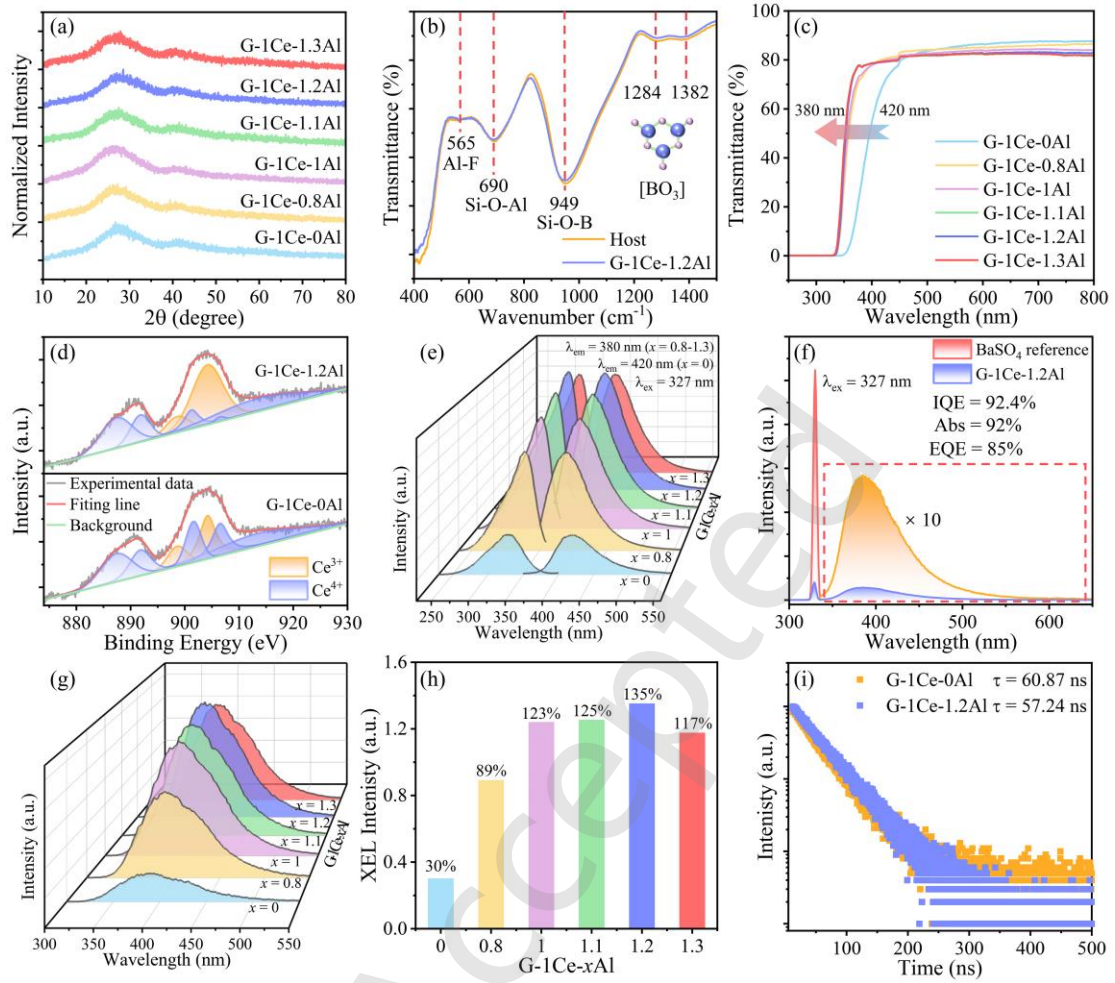


Fig. 2 (a) XRD patterns of G-1Ce- x Al. (b) FTIR patterns of the host and G-1Ce-1.2Al. (c) Transmission spectra of G-1Ce- x Al. (d) XPS spectra of G-1Ce-0Al and G-1Ce-1.2Al. (e) PLE and PL spectra of G-1Ce- x Al. (f) Quantum efficiency of G-1Ce-1.2Al. G-1Ce- x Al: (g) XEL spectra; (h) XEL relative intensity comparison with BGO. (i) Decays curves of G-1Ce-0Al ($\lambda_{\text{ex}} = 327$ nm, $\lambda_{\text{em}} = 420$ nm) and G-1Ce-1.2Al ($\lambda_{\text{ex}} = 327$ nm, $\lambda_{\text{em}} = 380$ nm).

In scintillator materials, incident X-ray photons or high-energy particles are first absorbed, and subsequently transferred to luminescent centers, where it is converted into visible photons for detection and signal acquisition.[33] The efficiency of this

conversion process is strongly governed by the optical transparency of the scintillator. Low transmittance intensifies photon absorption and scattering losses during light propagation, thereby diminishing the signal-to-noise ratio and spatial resolution, ultimately compromised imaging fidelity.[33] Consequently, high optical transmittance is crucial for achieving superior imaging performance in glass scintillators. As depicted in Fig. 2c, all G-1Ce-*x*Al samples maintain excellent optical transparency, with visible-light transmittance exceeding 80%, ensuring minimal optical attenuation and rendering them highly suitable for scintillation imaging. Their refractive indices (*n*) are summarized in Table S1, providing essential optical parameters for evaluating light propagation and extraction in these glasses. Obviously, the incorporation of Al powder brings about a markedly blue shift of the absorption edge from 420 to 380 nm. Given that the 420 nm absorption feature originates from the Ce⁴⁺ charge-transfer transition, this spectral evolution indicates that the Al powder effectively suppresses the oxidation of Ce³⁺ during the melt-quenching process.[34, 35]

To further elucidate the Al-mediated valence-state modulation of Ce ions. X-ray photoelectron spectroscopy (XPS) measurements were performed on G-1Ce-0Al and G-Ce-1.2Al samples (Fig. 2(d) and Table S2). The spectra can be satisfactorily deconvoluted into two characteristic Ce³⁺ components at 904.2 and 898.6 eV, accompanied by five Ce⁴⁺ contributions located at 913.5, 906.5, 901.6, 892.2 and 887.4 eV, respectively.[34] Upon Al introduction, the fraction of Ce³⁺ rises substantially from 26.42 to 44.84%, evidencing an Al-driven shift of the Ce³⁺/Ce⁴⁺ equilibrium toward the scintillation-active Ce³⁺ state. In addition, the XPS survey spectra (Fig. S2) clearly

identify all constituent elements, including Si, Al, B, C, O, F, Ba, Na, and Ce, consistent with the designed compositions.

Under excitation at 327 nm, the Al-free glass (G-1Ce-0Al) generates a broad emission band centered at 420 nm, characteristic of the parity-allowed $5d-4f$ transition of Ce^{3+} (Fig. 2(e) and S3).[36] Al incorporation triggers a blue shift of the emission maximum to 380 nm, reflecting a profound change in the electronic environment of the luminescent centers. In the absence of Al powder, a considerable fraction of Ce ions exists as Ce^{4+} . The broad $\text{O}^{2-}\text{-Ce}^{4+}$ charge-transfer absorption extends deep into the visible region and overlaps extensively with the Ce^{3+} emission band, causing severe self-absorption. As a consequence, the intrinsic Ce^{3+} emission is significantly attenuated and appears red-shifted to 420 nm. Following the addition of Al, Ce^{4+} species are efficiently reduced to optically active Ce^{3+} centers, substantially alleviating reabsorption losses and unveiling the intrinsic Ce^{3+} luminescence centered at 380 nm. The corresponding PLE spectra are dominated by an excitation band located at 327 nm, assigned to the $4f-5d$ transition of Ce^{3+} (Fig. 2(e) and S4). Both PL and PLE intensities follow a volcano-shaped trend with increasing Al content, attaining their optimum at $x = 1.2$ before progressively declining due to concentration-induced quenching. Quantitative PL quantum yields (PLQY) of G-1Ce- x Al glasses were further determined under 327 nm excitation (Fig. 2(f) and S5) employing Eq. (S1-S3), which reveal the same compositional dependence as the PL intensity. When the irradiation source is switched to X-ray, the XEL spectra (Fig. 2(g) and S6(a)) closely match their PL counterparts (Fig. 2(e)), indicative of that radiative recombination under both excitation

modes stems from the identical Ce^{3+} luminescent centers. Consistent with the PL and PLQY results, the XEL intensity is maximized at $x = 1.2$ and delivers an approximately 4.5-fold enhancement over the Al-undoped sample (Fig. 2(h)).

With the Al content fixed at $x = 1.2$, the activator Ce^{3+} concentration was subsequently optimized in G- $y\text{Ce}$ -1.2Al ($y = 0.8$ -1.6) series. Among these compositions, the G-1Ce-1.2Al achieves the highest XEL intensity, reaching 135% of commercial BGO (Fig. S6(b) and S7). More importantly, it displays an internal quantum efficiency (IQE) of 92.4%, an external quantum efficiency (EQE) of 85% and an absorption efficiency (Abs) of 92% (Fig. 2(f)). Such exceptionally high IQE reflects highly efficient radiative recombination with minimal non-radiative losses. This originates from the synergistic effect of low intrinsic defect density, homogeneous Ce^{3+} distribution, and the low-phonon-energy glass matrix, which collectively facilitate carrier capture by Ce^{3+} luminescent centers.[37, 38] Meanwhile, the high EQE demonstrates efficient photon extraction from the bulk glass with limited optical loss, further contributing to the superior scintillation performance.

Generally, the ultrafast decay kinetics of the material can effectively shorten both signal response and recovery times, which alleviates inter-frame signal crosstalk in high-speed imaging systems.[39] The PL decay profiles of G-1Ce-0Al and G-1Ce-1.2Al samples (Fig. 2(i)), monitoring the $\text{Ce}^{3+} 5d \rightarrow 4f$ emission under 327 nm excitation, were quantitatively analyzed via Eq. (S4).[40] Upon Al incorporation, the UV-excited decay lifetime reduces from 60.87 to 57.24 ns, hinting slightly accelerated luminescence relaxation. While excited at X-ray, G-1Ce-1.2Al provides a scintillation

decay lifetime of 142.3 ns (Fig. S8), which is shorter than those of most reported scintillator materials (Table S3). Such sub-microsecond scintillation response minimizes residual luminescence and temporal crosstalk, thereby favoring rapid-event detection and high-frame-rate dynamic X-ray imaging.

Rational defect engineering has emerged as a powerful means for endowing scintillators with advanced multifunctional imaging capabilities. In particular, the deliberate construction of deep trapping centers capable of efficient charge-carrier capture and controlled release provides the physical basis for time-lapse imaging. As portrayed in Fig. S9 and 3(a), neither the unirradiated sample nor the sample subjected to 340 nm UV light excitation displays any appreciable thermoluminescence (TL) response. In striking contrast, X-ray irradiation gives rise to a prominent TL band centered at 409 K, the intensity of which progressively elevates with irradiation power and exposure duration. These observations manifest that the associated trapping centers are generated exclusively through high-energy X-ray radiation and cannot be activated by UV excitation.

Detailed TL deconvolution identifies three energetically distinct trapping centers at 372, 411 and 491 K (Fig. S10), with estimated trap depths of 0.75, 0.82 and 0.98 eV, respectively, according to the empirical relationship $E_T = T_M/500$. [18, 33] Notably, the marked growth of the 491 K component under intensified X-ray irradiation signifies a gradual enrichment of deep traps (Fig. 3(a)). Such irradiation-driven evolution hints that X-ray not only promote defect generation but also enhance the energetic stability of trapped carriers, enabling more efficient charge accumulation and long-term carrier

retention.

3.2 XEL properties

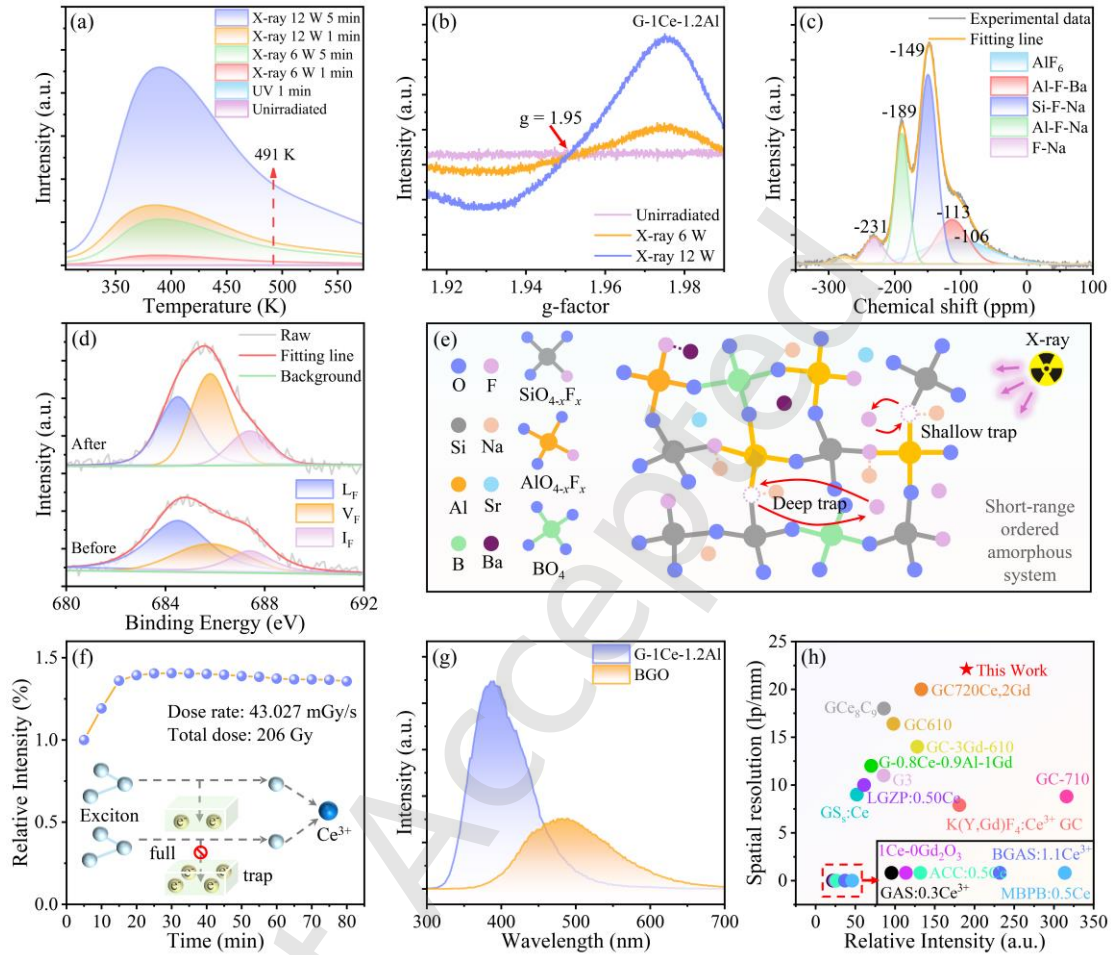


Fig. 3 G-1Ce-1.2Al: (a) TL curves recorded after X-ray irradiation at different powers (6 W: 30 kV, 200 μA ; 12 W: 60 kV, 200 μA) and exposure times, UV irradiation ($\lambda_{\text{ex}} = 340$ nm), and without irradiation; (b) EPR spectra before and after X-ray irradiation at different power levels (6 W: 30 kV, 200 μA ; 12 W: 60 kV, 200 μA); (c) Fitting results of the ^{19}F NMR spectra; (d) F1s XPS spectra before and after X-ray irradiation; (e) Schematic illustration of Frenkel defect formation induced by high-energy X-ray irradiation; (f) Evolution of XEL intensity under continuous X-ray irradiation for 80 min; (g) Comparison of XEL intensity between G-1Ce-1.2Al and BGO after 15 min

irradiation; (h) Comparison of relative XEL intensity and spatial resolution between the resultant sample and previously reported Ce^{3+} -doped glass and glass-ceramics.

Further evidence for defect formation is provided by electron paramagnetic resonance (EPR) measurements (Fig. 3(b)). The signal $g = 1.95$ progressively strengthens with increasing X-ray power. In fluoride-containing materials, such paramagnetic centers are commonly associated with radiation-induced Frenkel defects. Previous studies have predominantly reported this process in long-range ordered fluoride nanocrystals.[41-44] Remarkably, the present findings suggest that a similar defect-formation mechanism can also operate within a structurally disordered amorphous glass matrix.

To validate this hypothesis, the local coordination environment of F ions was systematically investigated using ^{19}F nuclear magnetic resonance (NMR) and XPS analyses. The ^{19}F spectra (Fig. 3(c)) disclose characteristic peaks at -106 , -113 , -149 , -189 and -231 ppm, which can be ascribed to the chemical shift signals of AlF_6 , Al-F-Ba , Si-F-Na , Al-F-Na and F-Na configurations, respectively.[45-47] These results indicate that F ions simultaneously participates as both a network modifier and a network-forming constituent, thereby regulating the structure compactness of the glass network. Meanwhile, distinct variations in the $\text{F}1s$ XPS profiles are observed after X-ray irradiation (Fig. S11), directly confirming radiation-induced reconstruction of the F coordination environment and migration of F ions from their original lattice-like sites.[48-50]

In this glass system, F primarily coordinates with Na^+ and resides within localized lattice-like environments, denoted as lattice F (L_F), with a characteristic binding energy of 684.5 eV based on the NIST XPS database. Upon X-ray irradiation, partial migration of F^- leads to the generation of F vacancies (V_F) within the lattice. The reduced local electron density around these vacancies weakens the electronic shielding of adjacent L_F sites, resulting in a positive shift of their binding energy. Accordingly, the relative fraction of the high-binding-energy component rises with irradiation time, enabling semi-quantitative evaluation of the concentration of radiation-induced V_F . [51-53] While inherent interstitial F (I_F), situated in a more electron-deficient coordination environment, intrinsically exhibits a higher binding energy. [51-53] To quantitatively elucidate the defect evolution process, the $\text{F}1\text{s}$ XPS spectra (Fig. 3(d)) were deconvoluted into three well-resolved components assigned to L_F (684.5 eV), V_F (685.8 eV) and I_F (687.4 eV). Following X-ray exposure, the relative fraction of V_F increases significantly from 32.7% to 45.0%, accompanied by a decrease in L_F content from 50.5% to 37.1% (Table S4), achieving a well-matched quantitative conversion between these two species. Taken together, the converging evidence from TL, EPR, ^{19}F NMR and XPS analyses establishes an unconventional defect-evolution pathway in the short-range ordered amorphous glass.

X-ray excitation triggers the migration of F ions toward interstitial positions, generating Frenkel defects. Depending on the displacement extent of F ions, the resulting defects can be categorized into shallow traps with spontaneous recovery capability or deep traps requiring external activation for de-trapping (Fig. 3(e)). [54]

Such dynamically generated trapping states offers efficient channels for charge accumulation, storage, and controlled release, laying the foundation for X-ray time-lapse imaging. As a direct consequence of this defect-mediated carrier-storage mechanism, G-1Ce-1.2Al glass displays robust X-ray-induced afterglow at 423 K (Fig. S12), with a persistent luminescence (PersL) duration beyond 600 s (Fig. S13). More importantly, the contribution of PersL becomes negligible at ambient temperature (Fig. S12), effectively circumventing afterglow-related image lag and motion-induced ghosting during dynamic imaging. This self-adaptive trapping behavior reconciles the seemingly contradictory requirements of long-term temporal information storage and artifact-free real-time imaging, enabling advanced multifunctional X-ray imaging.

Exceptional radiation tolerance is critical for ensuring the long-term operational stability of scintillators under prolonged X-ray exposure. Notably, G-1Ce-1.2Al emerges a pronounced irradiation-induced “bright burning effect” (Fig. 3(f) and S14), in which the XEL intensity increases continuously during the first 20 min of X-ray irradiation and subsequently reaches a stable plateau even after 80 min of continuous exposure.[55] This behavior arises from the progressive saturation of radiation-induced traps, which gradually redirects charge carriers from trap filling to Ce^{3+} radiative recombination.[27] Consequently, this irradiation-caused self-optimization boosts the integrated XEL intensity to 190% of commercial BGO (Fig. 3(g)), representing a state-of-the-art level among Ce^{3+} -doped glass scintillators and surpassing most reported Ce^{3+} -activated glass-ceramics (Fig. 3(h) and Table S5). Moreover, such irradiation-induced XEL enhancement is reversible and reproducible. As demonstrated by five

consecutive X-ray irradiation-heating cycles (Fig. S15), the “bright-burning effect” can be repeatedly activated during X-ray irradiation and subsequently erased by thermal treatment, exhibiting excellent cycling reproducibility. These results demonstrate the robust reversibility of the defect-mediated luminescence modulation and highlight the superior radiation tolerance of the sample.

Under intense X-ray irradiation, abundant free electrons and holes are generated in the glass matrix, while electron trapping at anion-vacancy-related defects leads to the formation of radiation-induced color centers.[56, 57] In conventional glass scintillators, the accumulation of these defects typically causes pronounced optical darkening, manifested as radiation-induced coloration with transmittance losses exceeding 30%, thereby deteriorating light transport and imaging performance.[27, 58] By contrast, even under high dose X-ray irradiation at 12 W (81 mGy/s), G-1Ce-1.2Al reveals only a 14% reduction in optical transmittance (Fig. S16). Moreover, such irradiation-caused coloration is fully reversible, with the original optical transparency completely restored upon subsequent thermal treatment. G-1Ce-1.2Al displays no detectable attenuation in emission intensity following 25 consecutive X-ray on-off cycles at a dose rate of 43 mGy/s (Fig. S17). These results unambiguously verify the outstanding resistance of G-1Ce-1.2Al to radiation-induced optical and structural degradation as well as its excellent fatigue durability under repeated exposure.

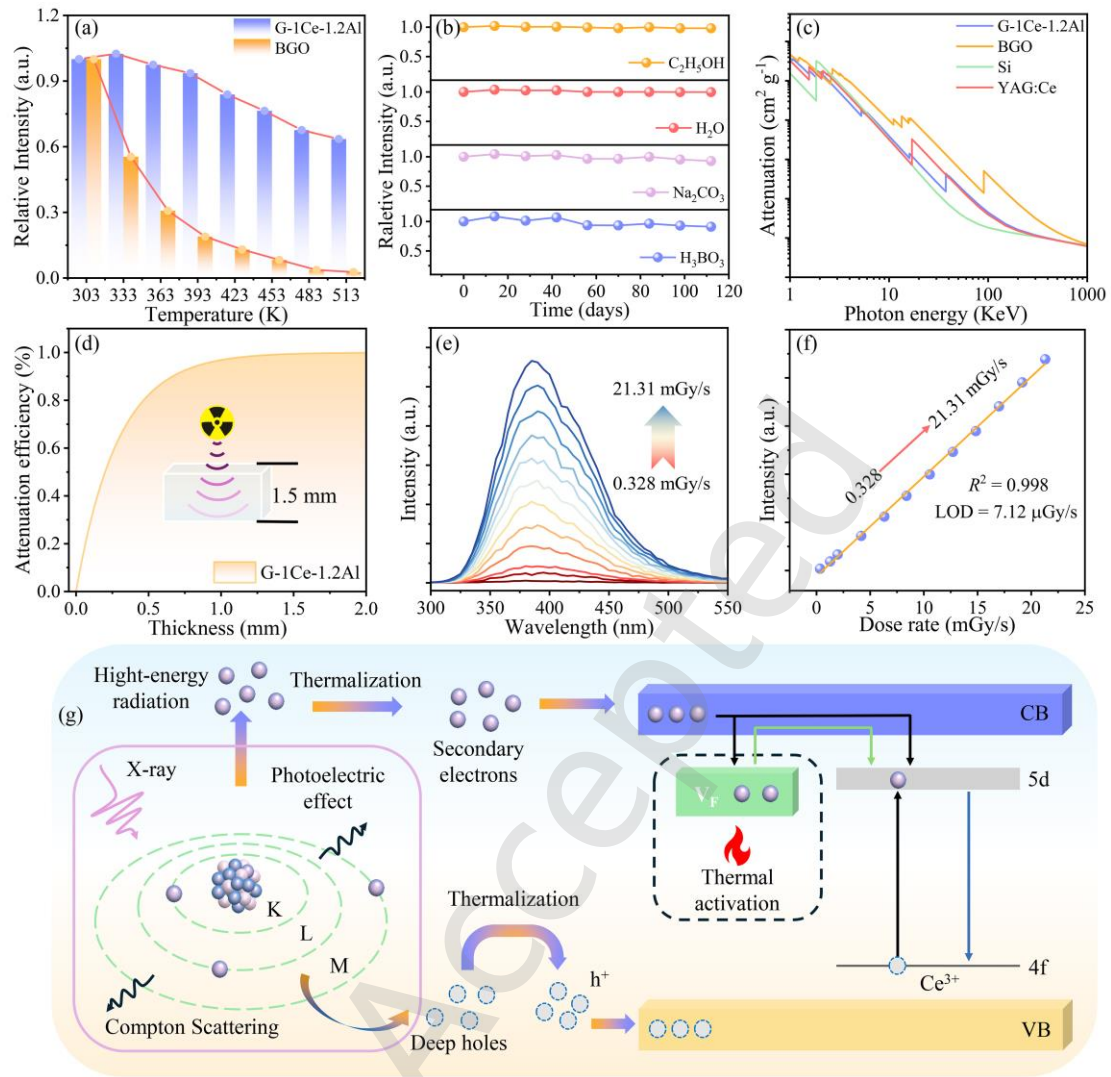


Fig. 4 G-1Ce-1.2Al: (a) Temperature-dependent variation of XEL intensity relative to BGO; (b) Temporal evolution of XEL spectra in water, ethanol, weak alkali (PH = 10), and weak acid (PH = 4) environments; (c) Attenuation coefficients of BGO, YAG:Ce, Si and G-1Ce-1.2Al; (d) X-ray attenuation efficiency as a function of sample thickness; (e) XEL spectra under X-ray radiation with different dose rates ranging from 0.33 to 21.31 mGy/s; (f) Corresponding linear fitting graph; (g) Schematic illustration of the XEL mechanism.

Thermal stability is a crucial performance metric for scintillators operation under

elevated-temperature scenarios, such as industrial non-destructive testing and energy exploration. As presented in Fig. 4(a) and S18, The XEL intensity of G-1Ce-1.2Al undergoes an initial improvement before gradually attenuating with rising temperature. In comparison with BGO, G-1Ce-1.2Al demonstrates markedly thermal robustness, with the integrated XEL intensity reaching 105% of its room-temperature value at 333 K and retaining 64% even at 513 K. In contrast, its PL intensity decreases monotonically without showing anti-thermal-quenching behavior (Fig. S19). This discrepancy can be rationalized by defect-engineered carrier regulation under X ray excitation. Thermally released carriers are promoted from trap states to the conduction band (CB) and subsequently captured by Ce^{3+} centers, partially compensating for thermally induced non-radiative losses.[55] Therefore, such a thermally assisted carrier recycling process significantly reinforces scintillation stability and stabilizes imaging output at elevated temperatures.

Practical scintillators are frequently exposed to humid, and chemically aggressive conditions. Accordingly, chemical stability is critical for long-term reliability. Benefiting from the unique glass composition and robust network architecture, G-1Ce-1.2Al sample features remarkable chemical corrosion resistance. Even after 112 days of immersion in water, ethanol, and weakly acidic and alkaline media (Fig. 4b and S20), more than 92% of the integrated XEL intensity is retained. Furthermore, the XEL profiles, XRD patterns, FTIR spectra, and optical transmittance remain essentially unchanged after prolonged exposure (Fig. S21 and S22), manifesting its strong applicability for long-term operation in underwater environments.

The density (ρ) and effective atomic number (Z_{eff}) of G- y Ce- x Al samples are listed in Table S1. Attributed to the incorporation of high- Z_{eff} Ba, these glasses display relatively high ρ and Z_{eff} (calculated by Eq. S5), offering a favorable basis for efficient X-ray absorption.[26] To quantitatively assess this property, the X-ray attenuation coefficient of G-1Ce-1.2Al was calculated and systematically benchmarked against those of BGO, YAG:Ce and Si across the photon energy range of 1–10³ KeV (Fig. 4(c)). According to data retrieved from the XCOM database, G-1Ce-1.2Al shows a high attenuation coefficient comparable to that of YAG:Ce, exceeding that of Si while remaining moderately lower than that of BGO. Moreover, near-complete X-ray attenuation is achieved at a thickness of merely 1.5 mm (Fig. 4(d)), which facilitates device miniaturization without sacrificing detection efficiency.

The X-ray sensitivity of G-1Ce-1.2Al was assessed by quantifying its RL response under varying irradiation dose rates (Fig. 4(e)). Across the investigated range of 0.328–21.31 mGy/s, the RL output appears an excellent linear dependence with a correlation coefficient of $R^2 = 0.998$ (Fig. 4(f)). This pronounced dose rate linearity enables sensitive X-ray detection, affording a low limit of detection (LOD) of 7.12 μ Gy/s, as calculated from Eq. (S6-S7). This property derives from the proportional generation of charge carriers (electron-hole pairs) with increasing X-ray flux, sustaining highly efficient energy conversion unimpeded by carrier saturation or quenching throughout the examined dose regime. Such a prominent dose linearity effectively eliminates signal distortion during radiation quantification, thereby supporting accurate and reliable measurement over a wide dynamic range for practical detection applications.

The RL mechanism of G-1Ce-1.2Al can be delineated into three sequential stages: energy conversion, carrier transport, and radiative emission (Fig. 4(g)).[59, 60] High-energy photons interact with the material through the photoelectric and Compton effects, producing a cascade of energetic electron-hole pairs. Following rapid thermalization, the resulting carriers efficiently funnel their energy into Ce^{3+} activators via resonant energy transfer, populating the $5d$ excited states. Subsequent radiative relaxation from $5d$ to $4f$ generates the characteristic visible scintillation emission.[61, 62] Simultaneously, a portion of the carriers are captured by Frenkel defects, forming metastable energy-storage states. Their thermally activated release and migration toward Ce^{3+} centers subsequently trigger delayed radiative recombination, thereby accounting for the observed afterglow phenomenon.

3.3 Multifunctional imaging applications

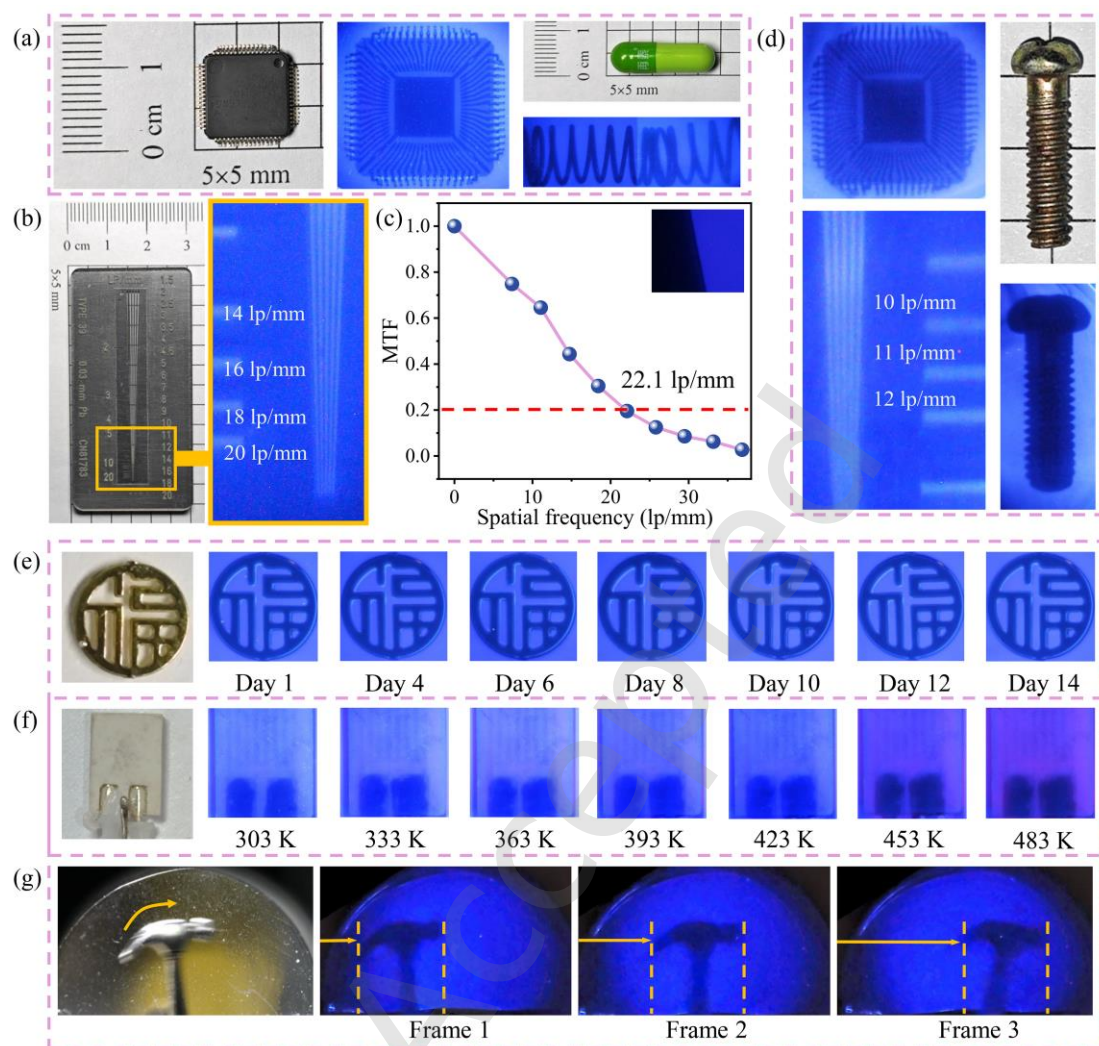


Fig. 5 Demonstration of X-ray-imaging capability of G-1Ce-1.2Al: Photographs and corresponding real-time X-ray images of (a) a chip, a spring and (b) a standard X-ray resolution test pattern plate; (c) MTF analysis; (d) Time-lapse X-ray imaging of a chip, a screw and a standard X-ray resolution test pattern plate; (e) Underwater X-ray imaging of a metal pattern; (f) High-temperature X-ray imaging of heated plates; (g) Real-time dynamic X-ray imaging of a hammer.

Building on the foregoing analyses, G-1Ce-1.2Al glass combines efficient scintillation output, rapid decay kinetics, abundant deep traps, and superior stability,

making it a promising candidate for versatile X-ray imaging applications. Its high instantaneous XEL intensity (135% of that of BGO) and fast scintillation response are conducive to high-quality real-time and dynamic imaging, while the irradiation-induced “bright-burning effect”, which enhances the XEL intensity to 190% of that of BGO after continuous X-ray irradiation, coupled with defect-mediated energy storage, offers strong potential for time-lapse imaging. Meanwhile, the robust water resistance and thermal tolerance provide opportunities for imaging under harsh conditions. These functionalities are subsequently validated employing self-built imaging systems (Fig. S23-S27). As portrayed in Fig. 5(a), the internal electronic components of a chip and the springs encapsulated within a capsule are distinctly resolved with high contrast and excellent structural fidelity. The spatial resolution, determined using a standard X-ray line-pair test pattern, reaches 20 lp/mm (Fig. 5(b)), with the alternating light and dark stripes remaining clearly discernible (Fig. S28). Quantitative modulation transfer function (MTF) analysis further substantiates the superior imaging characteristics, yielding a spatial frequency of 22.1 lp/mm at $MTF = 0.2$ (Fig. 5(c)). This value closely matches the visually evaluated resolution and, to the best of our knowledge, ranks among the highest real-time imaging spatial resolutions reported to date for Ce^{3+} -doped glass and glass-ceramic scintillators (Fig. 3(h) and Table S5).

Following 1 min of X-ray exposure, thermal stimulation enables time-lapse reconstruction of the internal features of both a chip and a screw, attaining a spatial resolution of 12 lp/mm (Fig. 5(d)). Notably, this resolution even surpasses the reported real-time imaging performance of several representative Ce^{3+} -doped scintillators (Table

S5). The imaging robustness is further examined under aqueous and elevated-temperature environments. X-ray images of a metallic pattern immersed in water and heated plates maintain clear structural features and stable contrast over storage periods of 1-14 days and across operating temperatures spanning 303-483 K, respectively (Fig. 5(e-f)), with minimal degradation in luminescence intensity and imaging quality.

The combination of negligible room-temperature afterglow and fast scintillation kinetics further confers high temporal fidelity. During dynamic X-ray imaging of a hammer moving at 12 cm/s, the G-1Ce-1.2Al material supports real-time visualization at 60 fps and captures three consecutive frames without observable motion blur while maintaining high image sharpness and spatial precision (Fig. 5(g) and Movie S1). The preserved image integrity during continuous motion highlights the broad prospects of the as-prepared glass for real-time tracking of rapidly moving objects and transient processes.

Collectively, these results establish the resulting glass as a multifunctional scintillation platform integrating high-resolution real-time imaging, delayed image readout, high-speed dynamic visualization, and exceptional adaptability to diverse imaging environments, underscoring its versatility for advanced X-ray imaging technologies.

4 Conclusion

A defect-engineered Ce^{3+} -activated borosilicate glass scintillator was developed to enable multifunctional X-ray imaging within a single amorphous platform. The material

features high scintillation efficiency, fast decay kinetics, robust structural and luminescence stability, offering a reliable foundation for X-ray detection under diverse operating conditions. The identification of irradiation-induced Frenkel defects elucidates the microscopic origin of delayed imaging behavior and highlights the capability of amorphous host to accommodate defect-mediated energy storage and release processes. Benefiting from the intrinsic environmental adaptability of glass and the synergistic regulation of luminescence and defect states, this work expands glass scintillators beyond conventional single-mode detection toward multimodal and environment-adaptive X-ray imaging. Consequently, high-resolution X-ray imaging, including real-time (22.1 lp/mm), time-lapse (12 lp/mm), dynamic, high-temperature and underwater modes, is achieved. Overall, these findings deepen the understanding of defect physics in amorphous scintillation materials and establishes a general design principle for developing next-generation high-performance composite glass scintillators and intelligent radiation detection systems.

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Author contributions

Chao Tang: Investigation, Writing - original draft, Formal analysis, Methodology

Siyu Guo: Methodology, Investigation, Formal analysis

Rongfei Wei: Conceptualization, Supervision, Resources, Writing - review & editing

Xiangling Tian: Methodology, Investigation, Resources

Fangfang Hu: Methodology, Resources

Hai Guo: Methodology, Conceptualization, Resources

Availability of data and materials

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

Competing interests

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

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