

Pulse-radiolysis study of radioluminescence in colloidal lead halide perovskite quantum dots: comparison with photoexcitation

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ABSTRACT

Lead halide perovskite quantum dots (QDs) have emerged as promising scintillator materials, however, their emission behavior under ionizing radiation in solution remains insufficiently characterized. In this study, we investigated the radiation-induced luminescence of commercially sourced colloidal lead halide perovskite QDs in toluene (CsPbCl₂Br, CsPbBr₃ and FAPbBr₃) using pulse radiolysis with high-energy electron beams.

The radioluminescence spectra closely coincides with the steady-state photoluminescence spectra, indicating that the same band-edge emissive states are populated under both optical and ionizing radiation excitation. Despite significant difference in photoluminescence quantum yields (0.44-0.83), the relative radioluminescence intensities show no direct correlation with photoluminescence efficiencies. This result is consistent with the contribution of solution-phase radiation processes, in which excitation is generated not only within the nanocrystals but also through radiolytically produced solvent-derived species. In addition, the emission spectrum, linewidths and intensities remain unchanged after gamma-ray irradiation up to 17 Gy, indicating that these colloidal QDs retain their photophysical characteristics within this dose range.

These results provide an experimental basis for distinguishing optical excitation from ionizing-radiation-induced excitation in colloidal perovskite QDs and demonstrate the utility of pulse radiolysis for evaluating radioluminescence processes in luminescent nanomaterials. Rather than introducing a new scintillator composition, this study provides a controlled experimental comparison of optical and ionizing-radiation excitation in a solution-phase perovskite QD platform.

1. Introduction

The design of organic/inorganic hybrid materials has become an important strategy for tuning photophysical and electronic properties through control of composition, local structure, and interfacial interactions (Chen et al., 2016, 2017, 2018, 2019; Zhou et al., 2024; Zheng

et al., 2023; Zhong et al., 2025; Gu et al., 2024; Wang et al., 2025; Hu et al., 2023; Guo et al., 2024; Zhang et al., 2021). Among these hybrid systems, metal halide perovskites are particularly attractive because of their outstanding and composition-tunable optoelectronic properties. In recent years, metal halide perovskite materials have attracted much attention as promising components for solar cells (Wen et al., 2023; Pan

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et al., 2022). In recent years, metal halide perovskite materials consisting of organic, metal and halogen ions have attracted much attention as a component material for solar cells (Kojima et al., 2009). In addition to photovoltaic applications, lead halide perovskites have recently emerged as promising materials for scintillators, which convert high-energy radiation such as X-rays, gamma-rays into visible light and are widely used in medical imaging, radiation therapy and high-energy physics (Wibowo et al., 2023; Onoda et al., 2022). However, lead halide perovskite scintillators are still in the early stages of research, and further studies are required not only to improve their performance and stability but also to clarify the fundamental photophysical processes governing radioluminescence (Moseley et al., 2021).

While a number of studies have reported scintillation from lead halide perovskite materials under X-ray irradiation, the excitation pathways and relaxation dynamics induced by other ionizing radiation such as high-energy electron beams and gamma-rays, remain insufficiently explored (Xu et al., 2021; Lian et al., 2023). These radiation sources are particularly relevant to modern radiotherapy and accelerator-based technologies, where understanding the interaction between ionizing radiation and luminescent materials is essential.

Under ionizing radiation, excitation occurs through complex cascades of ionization and differs fundamentally from those induced by photoexcitation. In solution system, these processes may involve solvent-mediated energy or charge transfer prior to population of the emissive states of the nanocrystals. Therefore, elucidating the relationship between photoluminescence and radioluminescence is crucial for understanding the excitation mechanisms of perovskite-based scintillators.

Pulse radiolysis provides a powerful experimental approach to probe such radiation-induced luminescence processes, as it enables controlled high-energy electron excitation. In the present context, pulse radiolysis enables direct comparison between optical excitation and ionizing-radiation excitation within the same colloidal perovskite QD platform. Such a comparison is valuable not because commercially available QDs are structurally novel, but because they offer a standardized model set with different compositions and photoluminescence efficiencies, allowing the relationship between photoluminescence and radioluminescence to be examined in a consistent manner.

In this study, we investigated the luminescence properties of lead halide perovskite QDs under high-energy electron beam excitation using pulse radiolysis. By comparing radioluminescence spectra with steady-state photoluminescence measurements and examining the effects of gamma-ray irradiation up to 17 Gy, we discuss how ionizing-radiation excitation in solution-phase perovskite QDs differs from conventional photoexcitation.

2. Experimental

2.1. Materials and methods

Lead halide perovskite QDs (λ_{em} 450 nm, 510 nm, 530 nm) were purchased from Sigma-Aldrich. Ultra-dehydrated toluene was purchased from FUJIFILM Wako (Osaka, Japan). The reagents were used without further purification.

The shape of the perovskite quantum dots was observed using a JEM-ARM200F (JEOL, Japan) analytical transmission electron microscope (TEM) with ultra-high resolution. 20 μ L of the sample solution, diluted 10 times with toluene, was dropped onto a Cu grid, allowed to dry completely, and then measured by TEM. The particle size was analyzed by extracting N particles ($N = 100$ or 200) from the obtained TEM image and analyzing them using ImageJ software.

Absorption spectra were measured at room temperature using a V630 UV-visible spectrophotometer (JASCO, Japan). The emission spectra were measured at room temperature using a FP-8200 fluorescence spectrometer (JASCO, Japan). Measurements were taken under aerobic and anaerobic conditions. Anaerobic conditions were achieved

by bubbling argon gas for 10 min. The emission quantum yields were measured at an excitation wavelength of 350 nm using the Quantaaurus-QY absolute PL quantum yield measurement device (HAMAMATSU, Japan). The measurement was carried out at room temperature under aerobic conditions.

The luminescence lifetimes were measured at room temperature using the Quantaaurus-Tau compact fluorescence lifetime measurement system C11367 (HAMAMATSU, Japan) with the photon counting method. Measurements were carried out under aerobic and anaerobic conditions. Anaerobic conditions were achieved by argon bubbling for 10 min. Analysis of the decay profile of the emission was performed using the software of the device. The lifetimes were determined by fitting two-component exponential curves ($y = y_0 + A_1 \cdot \exp(-x_1/\tau_1) + A_2 \cdot \exp(-x_2/\tau_2)$, $\Sigma A_i \tau_i^2 / \Sigma A_i \tau_i$). Electron-beam induced emission spectra were measured at room temperature using a multi-channel spectrometer, with a 28 MeV electron beam pulse accelerated by an L-band linear accelerator (L-band LINAC) or a 32 MeV electron beam by S-band RF gun based LINAC (S-band LINAC) irradiated onto the sample as the excitation source at SANKEN, the university of Osaka (Asanuma et al., 2023; Liu et al., 2024). In the gamma-ray irradiation experiment, the samples were placed at a distance of 1 m from the cobalt source (17.26 Gy/h).

3. Results and discussion

3.1. Structural and photophysical properties

The composition of the perovskite quantum dots used in this study is shown in Fig. 1a. While perovskites are represented by an ABX_3 -type structure (A: univalent cation, B: divalent cation, X: halide ion), perovskites with lead as the B-site ion were selected as examples. The univalent cations at the A site are Cs^+ or formamidinium (FA), and the halides are perovskites with Cl or Br. These perovskite quantum dots were coated with oleic acid and oleylamine. The emission maxima vary depending on the combination of ions used, each with an excitation wavelength of 355 nm and each with an emission maximum wavelength of 450, 509 and 532 nm for 1, 2 and 3, respectively (Fig. 1b). Their emission colors ranged from blue to green.

TEM and AFM to determine its size, which was 7 and 9 nm by TEM, with a thickness of 2 and 4 nm by AFM for 1 and 2, respectively (Fig. S1 and S2, and Table S1). 3 with FA at the A site, their size slightly varied and a distribution of around 12 nm in size and 21 nm in height was also observed. These results indicate that these perovskite materials are large or small enough to be called QDs.

Secondary, the photophysical properties of the perovskites used were examined. Measurements of the UV-vis absorption spectra show a broad absorption in the visible region, which also shows that, depending on the component, the broad region continues to longer wavelengths as it goes from 1 to 3, which is the one used in this study (Fig. S3). The quantum

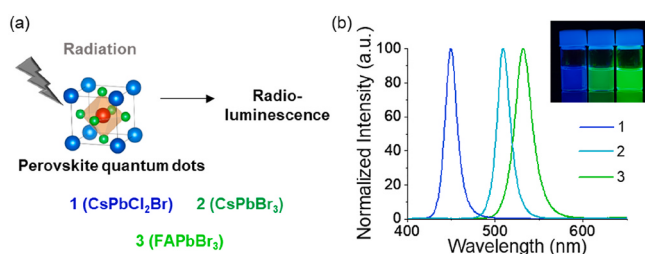


Fig. 1. (a) Schematic diagram of the radio-luminescence from perovskite quantum dots (1: $CsPbCl_2Br$, 2: $CsPbBr_3$ and 3: $FAPbBr_3$) when excited by radiation. Perovskite crystal structure was illustrated by VESTA 3 software (Momma and Izumi, 2011). (b) Photoluminescence spectra for perovskite quantum dots. The inset shows the photograph of samples upon UV light irradiation.

yields of photoluminescence upon excitation of 350 nm UV light were 0.44, 0.72 and 0.83 for 1, 2 and 3, respectively (Table S2). We also checked whether quenching by molecular oxygen in the emission spectrum was observed (Fig. 2). The results showed no significant oxygen-induced quenching in the emission spectrum. Similar results were obtained in photoluminescence lifetime measurements, where no oxygen quenching was observed (Fig. S4), indicating the low reactivity of the perovskite quantum dots used in this study with oxygen. Incidentally, the luminescent lifetimes upon 365 nm LED UV light were about 9.2, 6.3 and 59 ns for 1, 2 and 3 under Ar condition. As the luminescence lifetime at 3 is relatively long, it was considered that these variations in the emission lifetime may originate from defective sites. The results showed that these quantum dots could be excited by radiation and function as a luminescent material.

3.2. Radioluminescence under electron beam excitation

The luminescence after L-band (Fig. 3) and S-band (Fig. 4) LINAC electron beam excitation was then observed. To comprehensively investigate the scintillation processes in perovskite materials under electron-beam irradiation, both S-band and L-band pulse radiolysis systems were employed in this study, as they provide complementary experimental conditions. The S-band system, operating at lower electron energies and higher repetition rates, is suitable for high-sensitivity detection. In contrast, the L-band system, which delivers higher-energy electron pulses, allows us to evaluate scintillation behavior and radiation stability under high-energy irradiation conditions that are more relevant to practical radiation environments, such as those encountered in radiotherapy and radiation detection applications.

Toluene was used as the solvent and the luminescence was observed by electron beam from linear accelerator excitation (Fujitsuka and Majima, 2011). The emission during electron-beam irradiation was directly visualized using a 4K camera (Figs. 3a and 4), clearly revealing distinct luminescence colors corresponding to each perovskite QD sample. The emission spectra recorded under electron-beam excitation are shown in Fig. 3b and 4. The observed emission maxima appeared at approximately at 450, ~510 and 530–540 nm for samples 1, 2 and 3, respectively. Importantly, these emission wavelengths closely match those observed in the steady-state photoluminescence spectra (Fig. 1b), indicating that identical emissive states are populated under both photo- and ionizing radiation excitation. The similar radioluminescence spectral features observed with the L-band and S-band LINAC systems

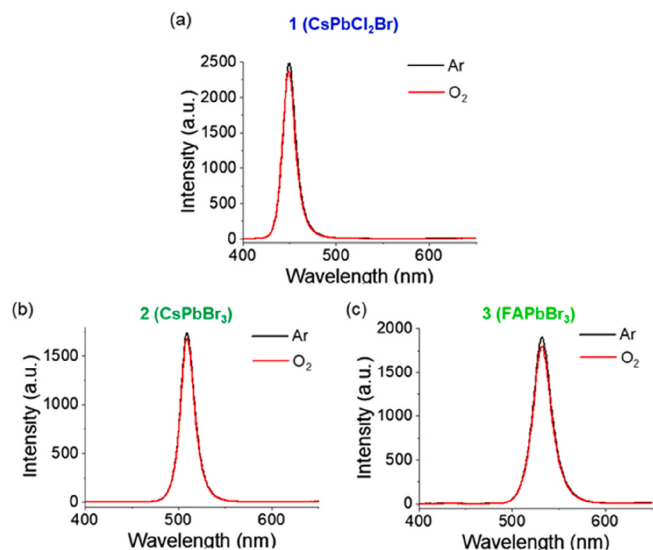


Fig. 2. Photoluminescence spectra of perovskite quantum dots under Ar or aerobic condition for 1 (a), 2 (b) and 3 (c).

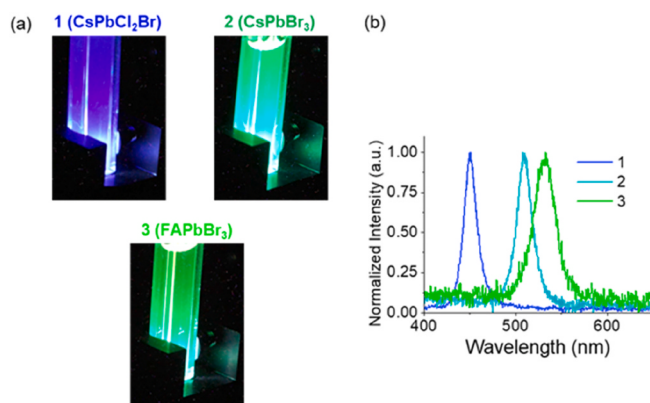


Fig. 3. (a) Photograph of emission under L-band LINAC electron beam excitation. (b) Luminescence spectra for 1, 2 and 3 in toluene under electron beam excitation. The concentrations of perovskite dispersion were 0.01, 0.1, 0.4 mg/mL for 1, 2, and 3, respectively.

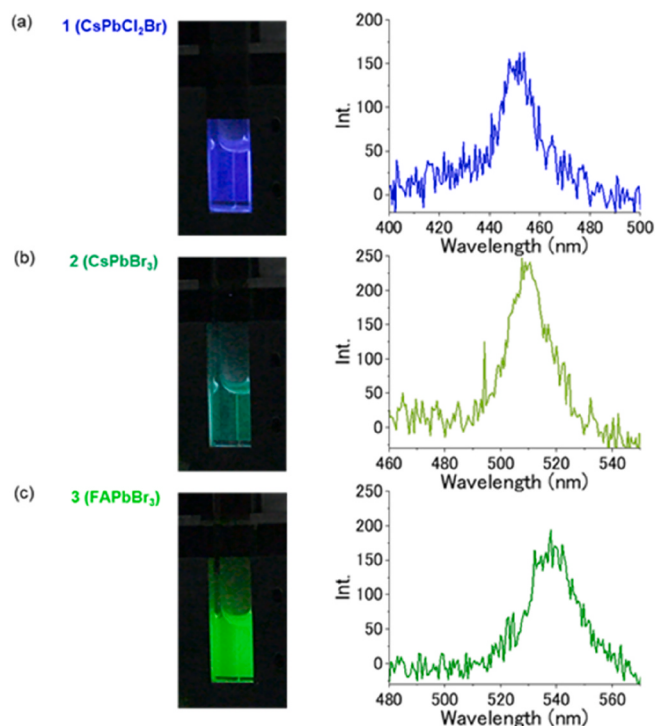


Fig. 4. Photograph of emission under S-band LINAC electron beam excitation (left panel), luminescence spectra for 1 (a), 2 (b) and 3 (c) in toluene under S-band LINAC electron beam excitation (right panel). The concentrations of perovskite dispersion were 0.03, 0.3, 0.8 mg/mL for 1, 2, and 3, respectively.

suggest that the emissive pathway is largely unchanged under the different irradiation conditions used here. In contrast, the radioluminescence intensity cannot be directly attributed to beam energy alone, because other beam-related parameters may also contribute.

Although a quantitative comparison of emission intensities was not performed, qualitative observation suggests that sample 1 exhibited relatively strong radioluminescence, taking weight concentration into account, despite possessing the lowest photoluminescence quantum yield among three samples. Radioluminescence was detected across the investigated QD concentration range, although the observed intensity is likely influenced by both the number of emissive centers and concentration-related optical losses. Thus, concentration optimization is expected to require a balance between signal output and optical

transparency. This lack of direct correlation between photoluminescence quantum yield and radioluminescence intensity implies that carrier generation and recombination processes under high-energy electron excitation differ from those under photo excitation. The absence of a simple correlation between radioluminescence intensity and photoluminescence quantum yield suggests that radioluminescence is influenced not only by intrinsic emissive efficiency but also by additional processes associated with ionizing-radiation excitation, such as carrier generation, trapping, relaxation, and non-radiative recombination.

Under pulse radiolysis conditions in toluene, high-energy electrons primarily interact with the solvent molecules, generating excited solvent states, radical cations, and secondary electrons. Energy and/or charge transfer from these radiolytically generated species to the perovskite QDs can subsequently populate the emissive states of the nanocrystals. Therefore, the observed radioluminescence likely involves solvent-mediated excitation processes in addition to intrinsic recombination within the perovskite QDs. Such processes are characteristic of radiation chemistry in solution systems, where luminescence can arise both from direct excitation of the solute and from recombination of radiation-generated charge carriers (Tabata, 1991; Yamaji et al., 2025; Thomas, 1969). The radioluminescence process may involve not only direct excitation of the QDs but also indirect solvent-mediated pathways arising from radiation-induced solvent ionization and excitation. However, the detailed microscopic mechanism remains unclear from the present data.

3.3. Stability under gamma-ray irradiation

The effects of gamma-ray irradiation on luminescence were also investigated. To evaluate the radiation stability of the perovskite QDs as potential scintillator materials, their photophysical properties after gamma-ray exposure were systematically examined. Gamma-rays represent a typical form of high-penetration ionizing radiation encountered in medical imaging, radiotherapy, and radiation monitoring environments, where scintillators are required to operate under continuous or repeated irradiation. Therefore, assessing the stability of luminescent materials under gamma-ray exposure is important for evaluating their durability in practical radiation environments.

The radiation stability of the samples was examined by placing the solutions at a distance of 1 m from a ^{60}Co source. The dose rate under these conditions was 17.26 Gy/h, and the samples were irradiated for 30 and 60 min, corresponding to accumulated doses of approximately 8.6 and 17.26 Gy, respectively. The UV-vis absorption and photoluminescence spectra before and after gamma-ray irradiation are shown in Fig. 5.

No noticeable changes in the spectral shape, peak position, or emission intensity were observed after gamma-ray irradiation for any of the perovskite QDs, indicating that the emissive states of the QDs were preserved after irradiation. No significant change was observed in the optical properties of the QDs after gamma-ray irradiation within the dose range examined in this study, indicating that no major degradation was detectable by the present photophysical measurements under the investigated conditions. However, subtle structural or surface-chemical changes cannot be excluded. These results suggest that the photophysical properties of the perovskite QDs remain stable under gamma-ray exposure up to 17 Gy. Such spectral stability indicates that no significant degradation, defect formation, or nonradiative quenching pathways were introduced within this irradiation range.

4. Conclusion

In summary, we investigated the luminescence behavior of colloidal lead halide perovskite QDs under high-energy electron-beam excitation by pulse radiolysis and compared the results with steady-state photoluminescence. For all QD compositions examined, the

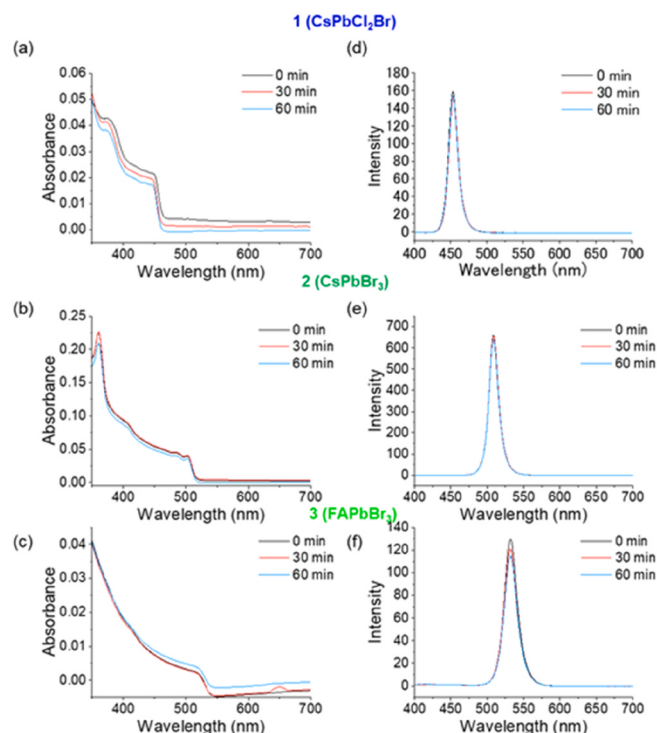


Fig. 5. Stability test after gamma-ray irradiation. Absorption and photoluminescence spectra of perovskite quantum dots under aerobic condition for 1 (a and d), 2 (b and e) and 3 (c and f) after 0, 30 and 60 min gamma-ray irradiation (17.45 Gy/h).

radioluminescence maxima closely matched the photoluminescence maxima, indicating that both excitation modes ultimately populate the same emissive band-edge states.

At the same time, the relative radioluminescence responses did not follow the trend in photoluminescence quantum yield across the samples. Although this observation does not by itself establish the full excitation mechanism, it provides experimental evidence that the processes leading to emissive-state population under ionizing radiation are not simply equivalent to those under optical excitation. In solution, such differences are reasonably attributed to the involvement of solvent-mediated radiation chemistry in addition to intrinsic recombination within the QDs.

We also found that the absorption and emission characteristics of the colloidal QDs were maintained after gamma-ray irradiation up to 17 Gy, indicating preserved photophysical properties within this dose range. Overall, this study establishes a comparative experimental framework for examining radioluminescence in colloidal perovskite QDs and shows that pulse radiolysis is a useful approach for probing how ionizing-radiation excitation differs from conventional photoexcitation in luminescent nanomaterials.

CRediT authorship contribution statement

Zheming Su: Methodology, Investigation. **Setsuka Homma:** Methodology, Investigation. **Quanlin Yu:** Methodology, Investigation. **Yoshio Mizuta:** Methodology, Investigation. **Yusa Muroya:** Methodology, Investigation. **Toshiya Muto:** Methodology, Investigation. **Jinfeng Yang:** Methodology, Investigation. **Tomonao Hosokai:** Methodology, Investigation. **Minoru Yamaji:** Methodology, Investigation. **Tomohiro Sakatani:** Methodology, Investigation. **Hajime Shigemitsu:** Methodology, Investigation. **Tadashi Mori:** Methodology, Investigation. **Toshiyuki Kida:** Methodology, Investigation. **Mamoru Fujitsuka:** Supervision, Methodology, Investigation, Funding acquisition. **Yasuko Osakada:** Writing – review & editing, Writing – original

draft, Methodology, Investigation, Funding acquisition, Conceptualization.

Ethical approval

The author states that the research work carried out in this manuscript does not contain any experiments involving human and/or animal tissue following the ethical guidelines.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.radphyschem.2026.113989>.

Data availability

Data will be made available on request.

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