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ILMIY AXBOROTNOMA

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M U N D A R I J A
C O N T E N T S

№	Maqola nomi	Article title	Bet/ page
1.	$CsPbBr_3$ kvant nuqtalariga asoslangan lyuminissent quyosh konsentratorlarining optik xossalarini optimallashtirish **Nurimov A.D., Davurov Sh.M., Sherqulov E.S., Juraboyev L.A., Zakhidov A.**	Optimising optical performance of $CsPbBr_3$ quantum dots-based luminescent solar concentrators **Nurimov A.D., Davurov Sh.M., Sherqulov E.S., Juraboyev L.A., Zakhidov A.**	**5-10**
2.	Impuils-davriyli lazer nuri ta'sirida rux oksidi va rux selenid mikrokukunlarida uyg'otilgan fotolyuminesensiya **Rakhmatullaev I.A., Khaydarov Kh.S., Hoshimov Q.H., Abdusattorov B.A.**	Photoluminescence in zinc oxide and zinc selenide micropowders under pulsed-periodic laser excitation **Rakhmatullaev I.A., Khaydarov Kh.S., Hoshimov Q.H., Abdusattorov B.A.**	**11-15**
3.	Perovskit–polimer kompozitiga asoslangan ko'k bir qatlamli yorug'lik chiqaruvchi diodlar **Rahmonova M.O., Muxamadiyev N.Q.**	Blue single-layer light-emitting diodes based on a perovskite–polymer composite **Rahmonova M.O., Muxamadiyev N.Q.**	**16-23**
4.	Chegaraviy oqim bilan berilgan chiziqsiz diffuziya sistemasining sonli tadqiqoti **Yarmetova D.**	Numerical study of a nonlinear diffusion system with boundary fluxes **Yarmetova D.**	**24-28**
5.	Iqlim o'zgarishi sharoitida samarali haroratlar yig'indisi taqsimoti (Samarqand viloyati misolida) **Kholbaev G., Eshmirzaev D., Egamberdiev Kh., Shermukhammedov U., Arzikulov M., Yuldasheva Sh.**	Spectral properties of schrödinger operator for a two-fermion system on a lattice: eigenvalues and resonances **Kholbaev G., Eshmirzaev D., Egamberdiev Kh., Shermukhammedov U., Arzikulov M., Yuldasheva Sh.**	**29-36**
6.	Janus kinaza ingibitorlari sifatida 4,6-dialmashgan xinazolin hosilalarini dizayni va sintezi **Gaybullaev Sh., Takhirov Y., Zohidov K., Chao Niu, Bozorov Kh.**	Design and synthesis of 4,6-disubstituted quinazoline derivatives as janus kinase inhibitors **Gaybullaev Sh., Takhirov Y., Zohidov K., Chao Niu, Bozorov Kh.**	**37-43**
7.	Suzuki-miyaura usulida 3-almashingan-pirido[2,3-*d*]pirimidinonlarning sintezi **Khushnazarov Z., Turgunov D., Bozorov Kh.**	Synthesis of 3-substituted-pyrido[2,3-*d*]pyrimidinones by the suzuki-miyaura method **Khushnazarov Z., Turgunov D., Bozorov Kh.**	**44-48**
8.	2-metil-pirazolo[3,4-*d*]pirimidinonlarning sintezi **Ruzieva F., Turgunov D., Bozorov Kh.**	Synthesis of 2-methyl-pyrazolo[3,4-d]pyrimidinones **Ruzieva F., Turgunov D., Bozorov Kh.**	**49-53**
9.	Organo/noorganik ms-schiff gibrid sorbentning metall kationlariga nisbatan sorbsiya xossalarini baholash **Kuliboyeva N., Uzokov J., Mukhamadiyev A., Ruziyev I., Mukhamadiyev N.**	Evaluation of the sorption properties of organo/inorganic ms-schiff hybrid sorbents towards metal cations **Kuliboyeva N., Uzokov J., Mukhamadiyev A., Ruziyev I., Mukhamadiyev N.**	**54-64**

№	Maqola nomi	Article title	Bet/ page
10.	Qaror daraxtlariga asoslangan anjuman usullari yordamida ma'lumotlarni oldindan qayta ishlash va tasniflash algoritmlari **Akhatov A., Nazarov F.**	Data preprocessing and classification algorithms using ensemble methods based on decision trees **Akhatov A., Nazarov F.**	**65-74**
11.	Fotokatalitik faollikka ega TiO_2 nanoklasterlarini struktur va elektron xossalarini kvant kimyoviy usullarda baholash **Mamaziyayeva S., Uzokov J., Toshpulatova L., Mukhamadiyev N., Mukhamadiyeva Kh.**	Evaluation of the structure and electronic properties of photocatalytic tio2 nanoclusters using quantum chemical methods **Mamaziyayeva S., Uzokov J., Toshpulatova L., Mukhamadiev N., Mukhamadieva Kh.**	**75-81**
12.	Cu(II) ionlarining Piren sorbsiyasini sinergetik ta'sirini Langmuir-Freundlich modellashtirish asosida baholash **Vokhidov A., Kosimov S., Khalilov S., Mukhammadiyev N.**	Evaluation of the synergetic effect of pyrenee sorption of cu(ii) ions based on langmuir-freundlic modeling **Vokhidov A., Kosimov S., Khalilov S., Mukhammadiev N.**	**82-90**
13.	Magnit suyuqliklarining magneto-optik yorug'lik o'tkazuvchanligi **Kuvandiqov O., Kirgizov S., Mamasoliyev B., Kirgizova M.**	Evaluation of the synergetic effect of pyrenee sorption of cu(ii) ions based on langmuir-freundlic modeling **Kuvandikov O., Kirgizov S., Mamasoliev B., Kirgizova M.**	**91-97**
14.	Sun'iy intellekt mexanizmlari asosida suv tarkibini aqlli tahlil qilish usullari **Salimova M., Nazarov F.**	Methods for intelligent analysis of water composition based on artificial intelligence mechanisms **Salimova M., Nazarov F.**	**98-107**
15.	Glukoza asosida uglerodli kvant nuqtalarining fotolyuminestentsiya xossalari **Sobirova F., Ishankulov A., Khalilov K., Galyametdinov Y.**	Photoluminescence properties of glucose-based carbon quantum dots **Sobirova F., Ishankulov A., Khalilov K., Galyametdinov Y.**	**108-112**
16.	Bir o'lchamli kvadrat panjara zinasida diskret Shredinger operatorining xos qiymatlari **Muminov M.I., Sadullaeva M.S.**	On eigenvalues of the discrete schrödinger operator on a ladder of one-dimensional square lattice **Muminov M.I., Sadullaeva M.S.**	**113-120**
17.	O'zbekistonda aholi hududiy harakatining mintaqaviy xususuyatlari **Mukhamedov O.L.**	Regional characteristics of population territorial movement in Uzbekistan **Mukhamedov O.L.**	**121-126**

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OPTIMISING OPTICAL PERFORMANCE OF CsPbBr3 QUANTUM DOTS-BASED LUMINESCENT SOLAR CONCENTRATORS

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Abstract. This work reports the synthesis of high-quality $CsPbBr_3$ quantum dots (QDs) via an octylammonium bromide (OABr)-assisted ligand-assisted reprecipitation (LARP) method and the systematic tuning of their optical properties. The as-prepared QDs exhibit clear quantum-confinement features, with absorption edges located at ~510–520 nm and narrow photoluminescence (PL) peaks in the range of ~491–518 nm. By varying the ligand-to-precursor ratio from 3:5 to 12:10:5:2000, the polydispersity and emission intensity of the QDs can be effectively controlled. Increased ligand content improves surface passivation, resulting in smoother absorption spectra and enhanced optical stability. Furthermore, embedding the QDs into a PMMA matrix induces pronounced blue shifts (~14–26 nm), which are attributed to reduced aggregation and interfacial interactions between the nanocrystals and the polymer host. The optimised $CsPbBr_3$ QD/PMMA composites exhibit high photoluminescence efficiency and tunable emission characteristics, highlighting their strong potential as luminescent materials for luminescent solar concentrator (LSC) applications.

Keywords. Quantum dots, ligand, ligand-assisted reprecipitation method, PMMA, luminescent solar concentrators, photoluminescence, absorption.

$CsPbBr_3$ kvant nuqtalariga asoslangan lyuminissent quyosh konsentratorlarining optik xossalarini optimallashtirish

Annotatsiya. Ushbu ishda $CsPbBr_3$ kvant nuqtalari oktillammoniy bromid (OABr) yordamida ligand-assisted reprecipitation (LARP) usuli orqali sintez qilinib, ularning optik xossalari tizimli ravishda sozlandi. Olingan kvant nuqtalar yaqqol kvant-chegaralanish xususiyatlarini namoyon etib, yutilish chetlari ~510–520 nm oralig‘ida va tor fotolyuminestsensiya (PL) cho‘qqilari ~491–518 nm diapazonida joylashgan. Ligand va prekursorlar nisbati 3:5 dan 12:10:5:2000 gacha o‘zgartirilishi orqali kvant nuqtalarning polidispersligi va nurlanish intensivligi samarali boshqarildi. Ligand miqdorining oshirilishi sirt passivatsiyasini yaxshilab, yutilish spektrlarining silliqlashishiga va optik barqarorlikning ortishiga olib keladi. Bundan tashqari, kvant nuqtalarni PMMA matritsasiga kiritish sezilarli ko‘k siljishga (~14–26 nm) sabab bo‘lib, bu nanokristallarning agregatsiyasining kamayishi hamda polimer muhit bilan yuzalararo o‘zaro ta’sirlar bilan izohlanadi. Optimallashtirilgan $CsPbBr_3$ QD/PMMA kompozitlari yuqori fotolyuminestsensiya samaradorligi va sozlanadigan emissiya xususiyatlarini namoyon etib, ularning lyuminissent quyosh konsentratorlari (LSC) uchun istiqbolli materiallar ekanligini ko‘rsatadi.

Kalit so‘zlar: Kvant nuqtalar, ligand, LARP usuli, PMMA, lyuminissent quyosh konsentratori, fotolyuminestsensiya, yutilish

spektri.

Оптимизация оптических свойств люминесцентных солнечных концентраторов на основе квантовых точек $CsPbBr_3$

Аннотатция. В данной работе представлено получение высококачественных квантовых точек $CsPbBr_3$ методом лиганд-ассистированной репреципитации (LARP) с использованием бромида октилламмония (OABr) и систематическая настройка их оптических свойств. Полученные квантовые точки демонстрируют выраженные признаки квантового ограничения с краями поглощения в области ~510–520 нм и узкими полосами фотолюминесценции в диапазоне ~491–518 нм. Путём изменения соотношения лигандов к прекурсорам от 3:5 до 12:10:5:2000 эффективно контролируются полидисперсность и интенсивность излучения квантовых точек. Увеличение содержания лигандов улучшает пассивацию поверхности, что приводит к сглаживанию спектров поглощения и повышению оптической стабильности. Кроме того, внедрение квантовых точек в матрицу PMMA вызывает выраженный голубой сдвиг (~14–26 нм), обусловленный уменьшением агрегации и межфазных взаимодействий между нанокристаллами и полимерной матрицей. Оптимизированные композиты $CsPbBr_3$ QD/PMMA демонстрируют высокую эффективность фотолюминесценции и настраиваемые спектральные характеристики, что подтверждает их высокий потенциал в качестве люминесцентных материалов для люминесцентных солнечных концентраторов (LSC).

Ключевые слова: Квантовые точки, лиганды, метод LARP, PMMA, люминесцентные солнечные концентраторы, фотолюминесценция, спектр поглощения.

Introduction. All-inorganic lead halide perovskites have recently attracted considerable attention as promising materials for nanotechnology and next-generation photovoltaic applications due to their superior thermal and chemical stability compared with their organic–inorganic counterparts [1–3]. Among them, caesium lead bromide QDs exhibit outstanding optoelectronic properties, including high absorption coefficients across the visible spectrum, strong photoluminescence, efficient charge-carrier separation, long carrier diffusion lengths, high carrier mobility, and extended carrier lifetimes [4]. These advantageous characteristics have stimulated rapid progress in perovskite-based optoelectronic devices, particularly in solar energy harvesting technologies [5]. Although power conversion efficiencies of perovskite solar cells have increased dramatically over the past decade, their long-term operational stability remains a critical bottleneck for large-scale deployment, as they are vulnerable to degradation under thermal stress, moisture, and ultraviolet irradiation [6–8]. A promising strategy to address these limitations is the incorporation of a perovskite-based spectral conversion layer onto conventional silicon solar cells, where it can function as an LSC. In this approach, thin films composed of inorganic $CsPbBr_3$ QDs embedded in a polymer matrix act as down-shifting layers that convert high-energy photons, which would otherwise be lost as heat due to exceeding the silicon bandgap, into lower-energy photons that can be more efficiently harvested by the underlying silicon device.

Experimental details. A 0.1 M precursor solution was prepared by dissolving 21.2 mg of CsBr and 36.7 mg of $PbBr_2$ in 1 mL of dimethyl sulfoxide at room temperature, with magnetic stirring applied for 1 hour. Oleylamine and oleic acid served as the ligand. For the ligand solution, 0.1 ml oleylamine and 0.3 ml oleic acid were added to the precursor solution and stirred for 1 hour with

magnetic stirring. Separately, 5 mL of toluene was used as an antisolvent and stirred at 700 rpm to create a stable vortex. Then, 100 µL of the ligand solution was quickly injected into the vigorously stirring toluene [9]. A polymer matrix was formed by dissolving 10 g of PMMA in 20 mL of toluene, resulting in a 0.5 g/mL solution. Next, 85 µL of the QDs dispersion was incorporated into this polymer solution, and the mixture was thoroughly mixed to achieve even QD distribution. The final QD–PMMA mixture was poured into a tray-shaped mould and left to dry under ambient conditions for 48 hours [10]. Once the solvent had fully evaporated, a top glass layer was bonded using the same polymer solution to enhance mechanical stability. Silicon-based photovoltaic cells were affixed to the edges of the fabricated triplex structure using a thermally conductive adhesive. The edge-mounted Si-PVs were electrically interconnected in a series configuration to enable efficient charge collection [11]. Photoluminescence (PL) and optical absorption spectra were acquired using a FluoTime 300 spectrofluorometer and an RF-6000 UV–Vis spectrophotometer, respectively. The current–voltage characteristics of the luminescent solar windows were evaluated under standard AM 1.5G illumination conditions employing a calibrated solar simulator.

Results and Discussions. The synthesis mechanism of $CsPbBr_3$ perovskite quantum dots (PQDs) is illustrated in Figure 1a. The ligand-assisted reprecipitation (LARP) technique was employed for the synthesis of these PQDs. This room-temperature method promotes rapid supersaturation upon injection of the precursor solution into a poor solvent under vigorous stirring, leading to simultaneous nucleation of numerous crystallites. This burst nucleation process, coupled with effective surface passivation, facilitates the formation of highly monodisperse nanocrystals with narrow size distributions. Octylammonium bromide (OABr) molecules serve as capping ligands, dynamically binding to the surface of the PQDs. This passivation reduces the overall surface free energy, thereby stabilizing the nanocrystals in the thermodynamically favored cubic phase while mitigating contributions from elevated surface energy that could otherwise drive phase transitions or aggregation [12].

As depicted in Figure 1b, the colloidal PQDs exhibit intense green photoluminescence (PL) under UV irradiation, indicative of efficient radiative recombination and high quantum yield. In contrast, Figure 1c shows the transparent appearance of the QD dispersion under ambient white light, highlighting their nanoscale dimensions and excellent colloidal stability.

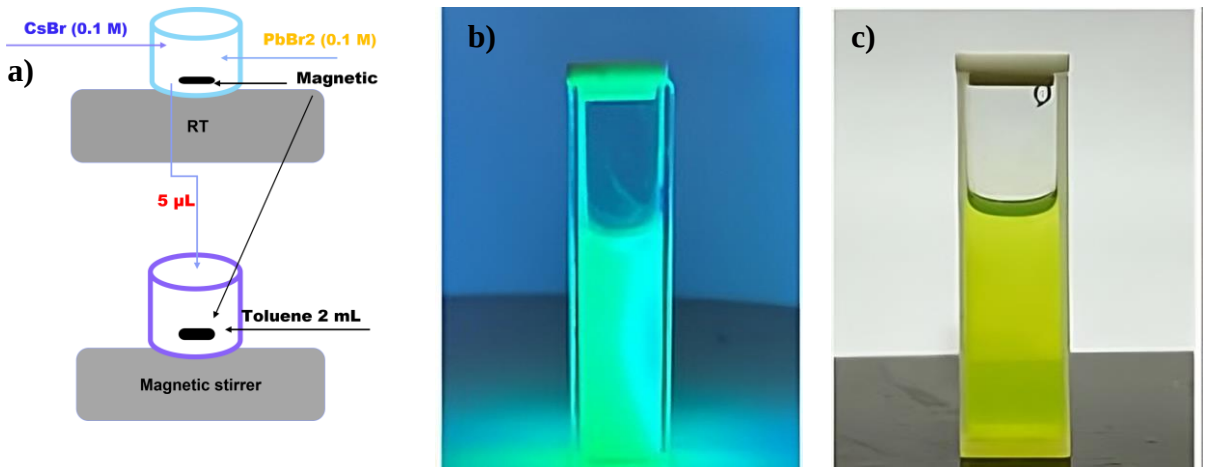

Figure 1. (a) Schematic illustration of the LARP synthesis procedure for $CsPbBr_3$ quantum dots. (b) Photoluminescence of the as-prepared $CsPbBr_3$ QD solution under UV excitation. (c) Corresponding appearance of the $CsPbBr_3$ QD solution under visible light illumination.

The size, monodispersity, and optical properties of the $CsPbBr_3$ PQDs are strongly influenced by the ligand-to-precursor ratio. Higher ligand concentrations enhance surface capping efficiency, restricting nanocrystal growth and yielding smaller particle sizes due to increased steric hindrance and kinetic control during nucleation and growth stages. This quantum confinement effect manifests in the PL spectra presented in Figure 2a, where varying ligand ratios result in tunable emission characteristics. Specifically, doubling the ligand amount (from a 3:5 to 6:5 ratio relative to precursors) induces a pronounced blue-shift in the PL peak position, attributable to the reduced nanocrystal dimensions and widened bandgap. Synthesis at ambient temperature predominantly yields PQDs with a cubic crystal structure, though morphological variations—such as cubic nanocubes or two-dimensional nanoplatelets—may occur depending on precise ligand dynamics. The symmetric profile of the observed PL spectra suggests the predominance of cubic-shaped nanocrystals in the present samples. In contrast, nanoplatelet morphologies typically exhibit asymmetric or bimodal PL features arising from thickness-dependent confinement and edge effects, as reported in sterically controlled syntheses of $CsPbBr_3$ nanoplatelets [4].

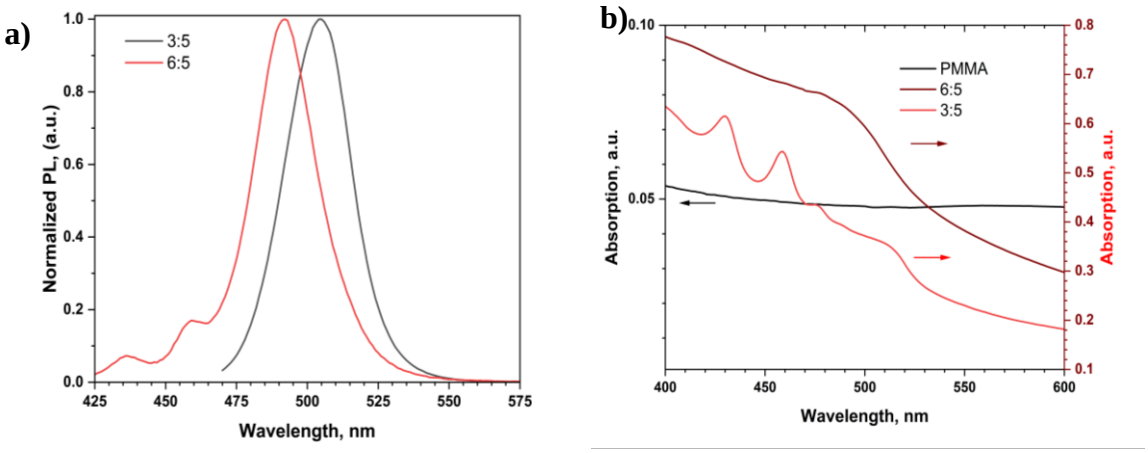

Figure 2. a) PL spectra of the CsPbBr3 QDs with different ratios and b) Absorption spectra of the PMMA as reference, QDs with different ratios.

The absorption spectra of $CsPbBr_3$ quantum dots (QDs) synthesised with OABr:$CsPbBr_3$ precursor ratios of 6:5 and 3:5, and subsequently embedded in PMMA films, reveal that the PMMA matrix itself is nearly optically transparent, while the QDs exhibit distinct absorption edges in the range of 500–520 nm, characteristic of quantum-confined $CsPbBr_3$ nanocrystals with an effective bandgap of approximately 2.4 eV. The sample prepared with a ligand ratio of 6:5 exhibits a markedly higher absorption intensity and a smoother spectral profile compared to the 3:5 sample, which displays pronounced oscillatory features in the 450–500 nm region. These differences originate from variations in ligand concentration, which critically influence nucleation dynamics, growth kinetics, and the final size distribution of the QDs. A higher ligand content enhances the nucleation rate and surface passivation, leading to more controlled crystal growth, reduced polydispersity, and a slightly smaller average particle size. In contrast, the 3:5 sample exhibits spectral artefacts attributable to a broader size distribution and increased inhomogeneous broadening, resulting in a less steep and more diffuse absorption edge. Overall, the 6:5 sample displays a sharper absorption onset around 500 nm, a narrower linewidth, and the highest absorbance (≈0.6–0.7). The 3:5 sample shows a slight red shift, consistent with inferior surface passivation and increased polydispersity. The absorption spectra of PMMA films exhibit a monotonic decrease from ~400 nm to 600 nm, with an onset around 510–520 nm.

Collectively, these results confirm that both samples exhibit strong quantum confinement, and high-quality $CsPbBr_3$ QDs were successfully synthesised, with the ligand ratio playing a decisive role in determining their optical uniformity and spectral characteristics.

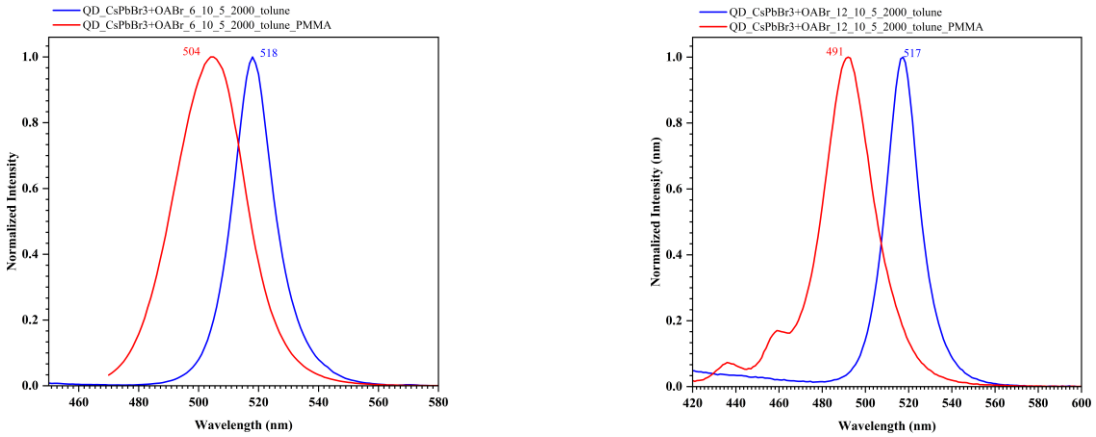

Figure 3. a) PL spectra of the 3:5 QDs and PMMA film with these QDs and b) PL spectra of the 6:5 QDs and PMMA film with these QDs

The PL spectra of CsPbBr3 QDs in toluene solution and PMMA-embedded films are shown in Figure 3. For the lower precursor ratio (6:10:5:2000), the colloidal QDs in toluene exhibit a narrow emission peak at 518 nm (FWHM ≈ 22 nm), corresponding to an effective band gap of ~2.39 eV and consistent with moderately confined nanocrystals (~8–10 nm estimated from sizing relations). Upon embedding in PMMA, the emission blue-shifts to 504 nm ($\Delta\lambda$ = 14 nm, $\Delta E \approx 0.07$ eV), with preserved FWHM (~22 nm). At the higher ratio (12:10:5:2000), toluene solution emission peaks at 517 nm, while PMMA embedding induces a larger blue-shift to 491 nm ($\Delta\lambda$ = 26 nm, $\Delta E \approx 0.13$ eV). These consistent blue-shifts are atypical compared to common red-shifts in higher-dielectric polymer matrices, where dielectric screening typically reduces exciton binding energy. The observed blue-shifts are primarily attributed to improved QD dispersion in the rigid PMMA matrix, which mitigates potential aggregation in toluene solution and restores stronger quantum confinement in isolated particles. Additionally, interactions between OABr ligands and PMMA's ester groups may reorganise the surface passivation layer or induce compressive strain, further widening the band gap. The larger shift at a higher precursor ratio suggests optimised ligand density and QD uniformity, enhancing matrix compatibility. Narrow FWHMs in both media confirm high monodispersity and minimal defect introduction during embedding. Complementary absorption spectra (Figure 3) show consistent trends in onset, supporting an intrinsic origin for the shift rather than reabsorption artefacts. This matrix-dependent blue-tuning highlights the potential of OABr-assisted LARP QDs in polymer composites for tunable green-blue emission in optoelectronic devices, with PMMA providing enhanced environmental stability.

Conclusion. In conclusion, this study demonstrates that OABr-assisted ligand-assisted reprecipitation (LARP) synthesis enables the fabrication of high-quality $CsPbBr_3$ quantum dots (QDs) with finely tunable optical properties. The QDs exhibit pronounced quantum-confinement behaviour, as reflected by well-defined absorption onsets in the ~510–520 nm range and narrow photoluminescence (PL) emissions spanning ~491–518 nm. Systematic variation of the ligand-to-precursor ratio (from 3:5 to 12:10:5:2000) significantly influences the polydispersity and optical intensity of the nanocrystals. Higher ligand contents lead to smoother absorption profiles due to improved surface passivation and reduced aggregation. Moreover, incorporation of the QDs into PMMA matrices induces notable blue shifts (~14–26 nm), which are attributed to de-aggregation

effects and interfacial interactions between the QD surfaces and the polymer host. These results indicate that the optimised $CsPbBr_3$ QD/PMMA composites possess high photoluminescence efficiency and spectral tunability, making them promising luminescent materials for application in luminescent solar concentrators (LSCs).

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