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Evolution and fabrication of carbon dot-based room temperature phosphorescence materials

Jiurong Li,^a Yongzhong Wu^b and Xiao Gong^{id}*^a

Traditional room temperature phosphorescence (RTP) materials usually include organometallic composites and pure organic compounds, which generally possess the disadvantages of high toxicity, high cost and complicated preparation. Carbon dots (CDs) are a new kind of luminescent material and have attracted widespread attention due to their benefits of excellent tunable emission, nice biocompatibility, cost-effectiveness, facile preparation and environmental friendliness. Since photoluminescence is an important luminescent property of carbon-based fluorescent nanomaterials, CD-based RTP materials have sparked a new research wave due to the properties of extremely long phosphorescence lifetime, large Stokes shift and high environmental sensitivity. In order to construct excellent CD-based RTP materials, many attempts have been made, and the corresponding progress has been achieved. Herein, we summarize the progress in CD-based RTP materials in recent years, mainly focusing on the outstanding contributions over the years, phosphorescence emission, phosphorescence lifetime, preparation and application of CD-based RTP materials. In particular, this review provides a comprehensive summary and analyze the outstanding contributions in the fields of the phosphorescence emission and phosphorescence lifetime of CD-based RTP materials over the years. Finally, several existing challenges and the future outlook of RTP materials based on CDs have been put forward.

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1. Introduction

Since the discovery of carbon dots (CDs) in 2004¹ and after nearly twenty years of development, researchers have put many efforts towards understanding them. As a new class of zero-dimensional fluorescent nanomaterials, CDs have attracted widespread concern and great research interest from a wide range of researchers owing to their distinguished properties such as adjustable photoluminescence (PL), excellent stability, facile surface functionalization, environmental friendliness, low toxicity and good biocompatibility.²⁻⁴ According to previous reports,⁵ CDs can be classified into four types, namely carbon nanodots (CNDs), carbon quantum dots (CQDs), graphene quantum dots (GQDs) and carbonized polymer dots (CPDs). Specifically, GQDs are small graphene fragments less than 20 nm in lateral dimension and less than five graphene flakes (~2.5 nm) in height, exhibiting pre-existing graphitic domains (sp² domains) and edge-rich chemical groups. CQDs are morphologically quasi-spherical carbon nanoparticles with a distinct lattice and chemical groups on their surface, and they have a crystalline core based on a mixture of sp² and sp³

domains. CNDs are also defined as quasi-spherical carbon nanoparticles, which consist mainly of cores with an amorphous structure, *i.e.*, CNDs have a certain degree of carbonization. CPDs have a hybrid polymer/carbon structure formed through the aggregation or cross-linking of linear polymers or monomers, with a surface consisting of abundant functional groups/polymer chains and carbon cores. Up to now, some notable achievements have been obtained in CD-related research, including the enhancement of photoluminescence quantum yield (PLQY), the regulation of multicolor fluorescence, the study of the luminescence mechanism and potential multifunctional applications.⁶⁻¹⁴ More importantly, the selection of different carbon source precursors and synthesis methods will lead to different fluorescence properties. The fluorescence of CDs is only one of the most common phenomena in their luminescent properties. In addition, CDs also possess other types of excellent luminescent properties, such as room temperature phosphorescence (RTP), thermally activated delayed fluorescence (TADF), up-conversion luminescence (UCL), chemiluminescence (CL), electrochemical luminescence (ECL), mechanoluminescence and so on.¹⁵ These luminescent properties have shown significant application prospects in biomedicine, optoelectronic devices, energy storage, sensing, anti-counterfeiting, catalysis, imaging and other fields.¹⁶⁻¹⁹ Particularly, the RTP phenomenon of CDs has afterglow

^aState Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Wuhan 430070, P. R. China. E-mail: xgong@whut.edu.cn

^bSchool of Mechanical Engineering, Suzhou University of Science and Technology, Suzhou 215009, P. R. China



emission performance which is very attractive because of its prolonged emission lifetime, larger Stokes shift and minimized interference from short-lived auto-fluorescence and scattered light. Now it has shown a broad application prospect in anti-counterfeiting, information encryption, sensing and other advanced fields.

In the last twenty years, the research on CDs has been widely carried out, and people have also reviewed and summarized their fluorescence in various aspects, such as the classification, synthesis methods, fluorescence regulation, fluorescence mechanism, different precursor selection, and applications in various fields.^{6,8,10,14,20–34} At the same time, the corresponding prospects for the fluorescence development of CDs are also proposed. Although there are some reviews providing a brief introduction of the afterglow phenomenon (*i.e.*, RTP and TADF) of CDs,^{15–19,35–38} there are few articles specifically describing the afterglow phenomenon related performance in detail.^{39,40} Based on the results previously reported, the radiative transition of triplet excitons is the key factor of the long afterglow phenomenon produced by CDs. The phosphorescence of CDs needs to be generated and enhanced by heteroatom doping or by embedding CDs into host matrices (*e.g.*, boric acid, polyvinyl alcohol, inorganic salts, layered double hydroxides, and zeolites) or by immobilizing CDs on a substrate. These matrices are essential for stabilizing the long-lived ternary state of CD fluorescence by effectively isolating and rigidifying CDs, minimizing the non-radiative leap of the triplet exciton and avoiding collisions of CDs with oxygen. In order to better distinguish the principal difference between fluorescence and phosphorescence, as shown in Fig. 1, the ground state (S_0) transits to the lowest singlet excited state (S_1) after being absorbed by ultraviolet and visible light, and then re-radiates back to S_0 and emits fluorescence. The generated excitons first enter S_1 from S_0 , and then enter the lowest triplet state (T_1)

through the intersystem crossing (ISC) process, and the excitons return to S_0 again and generate phosphorescence through the process of radiation transition. Or the exciton first goes through the process of reverse intersystem crossing (RISC), returns to S_1 from T_1 , and then returns to S_0 to generate delayed fluorescence. Obviously, since the energy of T_1 is lower than that of the singlet state S_1 , the phosphorescent emission peak will be significantly red shifted compared with fluorescence. It is worth noting that the fluorescence phenomenon of CDs is easy to realize, but the RTP of CDs is much harder to achieve. This is mainly the result of the intrinsic properties of the triplet excitons in relation to the spin-forbidden transition. More importantly, the triplet exciton is readily exhausted by vibrational deactivation and oxygen quenching under room temperature or higher. Thus, it can be seen that in order to obtain effective RTP, two important conditions need to be met, namely to improve the rate of ISC by enhancing the spin-orbit coupling of excitons and to stabilize the triplet-excited states *via* using structural confinement. For the first condition, it is generally possible to enhance the n-p* transition by incorporating transition metals, halogens or heterocycle groups to promote the ISC process and further generate triplet excitons. The second condition can be well achieved by encapsulating CDs in various substrates or creating self-protective structures (without substrate immobilization). Up to now, some efforts have been made to realize CD-based RTP materials to further explore their potential applications. Although some achievements have been made in the synthesis, construction and application of CD-based RTP materials, so far no article has systematically and completely summarized and analyzed them. And also, there have been several reviews on CD-based RTP materials in recent years, but they are only summarized and analyzed unilaterally for the synthesis, application or construction of CD-based RTP materials. Therefore, this review

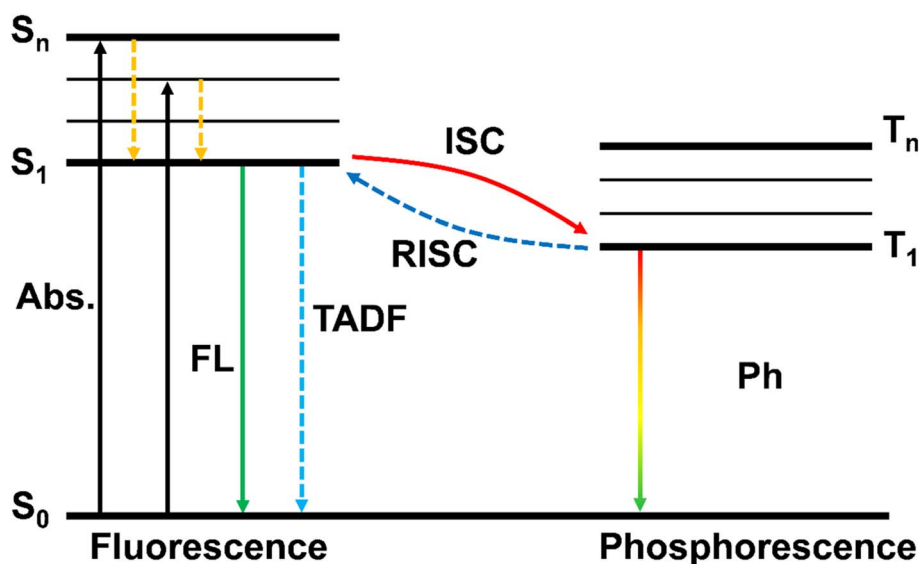


Fig. 1 Schematic illustration of the adsorption and emission processes of fluorescence (FL), phosphorescence (Ph), and delayed fluorescence (DF).



Table 1 Matrix-assisted CD-based RTP materials^a

| Matrix | Precursor | Afterglow mode (298 K) | Afterglow emission (nm) | Afterglow lifetime (τ) ms ⁻¹ (298 K) | PQY (%) | Remarks | Ref. |
|---|---|------------------------|-------------------------|--|----------|--|------|
| PVA | CDs and PVA | RTP | ~500 | 380 | NA | CDs-PVA | 42 |
| | | RTP + TADF | 569 | 1610 | NA | CDs/PVA | 43 |
| | | RTP | 422 | 338.4 | 6.88 | CDs-OH/PVA | 44 |
| | | RTP + TADF | 435 | 567 | 6.24 | CNDs - PVA | 45 |
| | | | 505 | 1387 | 12.32 | | |
| | | | 560 | 726 | 4.32 | | |
| | | | 590 | 311 | 2.03 | | |
| KAl(SO ₄) ₂ ·x(H ₂ O) | CDs and KAl(SO ₄) ₂ ·x(H ₂ O) | RTP | 500 | 655 | NA | CD-KAl(SO ₄) ₂ ·x(H ₂ O) | 46 |
| Urea | CDs and urea | RTP | 490 | 1.06 | 7% | HN powder | 47 |
| Urea | CDs and urea | RTP + DF | 500 | 1020 | 7.23 | NCD1-C | 48 |
| | | | 502 | 1110 | 14.0 | NCD2-C | |
| | | | 625 | 780 | 11.0 | NCD3-C | |
| PU | N-doped CQDs and PU | RTP + DF | 500 | 8.7 | NA | CQD/PU | 49 |
| | | | 525 | 386.8 | 5.99 | CDs-LDHs | 50 |
| LDHs | CDs and LDHs (Zn ²⁺) | RTP | 490 | 800 | 9.44 | Zn-CDs-LDH | 51 |
| | | | 490 | 719.9 | 9.58 | Zn-CDs-LDH | 52 |
| | | | 490 | 719.9 | 9.58 | Zn-CDs-LDH | 52 |
| SiO ₂ | m-CDs and colloidal nanosilica | RTP + TADF | 470 | 703 | NA | m-CDs@nSiO ₂ | 53 |
| | | | 520 | 1640 | NA | CDs@SiO ₂ | 54 |
| | | | 508 | 1760 | NA | CDs@SiO ₂ | 55 |
| | | | 507 | 1620 | NA | CDs@SiO ₂ | 56 |
| | | | 464 | 5720 | 26.36 | CDs@SiO ₂ | 57 |
| | | | 520 | 1860 | 11.6 | WSP-CNDs@silica | 58 |
| | | | 600 | 910 | 3.56 | CNDs-RhB@silica | 59 |
| | | | 520 | 1570 | 12.6 | CNDs@silica | 60 |
| | | | 510 to 610 | 1610 to 1260 | NA | PCDs | 61 |
| | | | 525 | 1800 | NA | CDs in silica gel | 62 |
| | | | Silica gel | CDs and silica gel | RTP | 430 | 350 |
| 500 | 22.32 | 14.1 | | | | CDs@zeolite | 64 |
| 620 | 1.81 | 5.7 | | | | | |
| 525 | 574 | NA | | | | CDs@zeolite | 65 |
| 440 | 153 | | | | | | |
| 530 | 2100 | 24.4 | | | | CDs@zeolite | 66 |
| 516 | 1400 | NA | | | | CDs@EuAPO-5 | 67 |
| NaCl | CDs and NaCl | RTP | 516-520 | 380-2100 | NA | CDs@zeolite | 68 |
| | | | 519 | 314 | NA | CD-NaCl | 69 |
| Cyanuric acid | CDs and cyanuric acid | RTP | 480 | 705 | 14 | CD-CA | 70 |
| | | | 480 | 687 | NA | CD-CA | 71 |
| | | | 550 | 220.74 | NA | CD@CA | 72 |
| | | | 690 | 13.29 | | | |
| | | | 425-510 | >2 hour | NA | m-CDs@CA | 73 |
| ZnAl ₂ O ₄ | CDs and zinc aluminate | RTP | 416 and 550 | 1740 | 23.2 | m, p-CDs@CA | 74 |
| | | | 517-650 | NA | NA | CDs-ZnAl ₂ O ₄ | 75 |
| | | | 490-570 nm | 2.26 | 17.5 | a-CDs/BA | 48 |
| | | | 454-500 | 783 | NA | GQD@BNO | 76 |
| | | | 520 | 1760 | 30 | CDs/BA | 77 |
| Boric acid | Lycorine hydrochloride and boric acid | RTP | 475-555 | 445.9 | 17.61 | CDs/B ₂ O ₃ | 78 |
| | | | 530 | 1670 | 48 | g-t-CD@BA | 79 |
| | | | 478-631 | 85.67-1064.21 | 6.5-18.6 | CDs@MOF | 80 |
| MOF | CDs and MOF | RTP | 530 | 10.48 | NA | CDs@MnAPO-CJ50 | 81 |
| | | | 620 | 10.94 | 9.6 | CDs@MnAPO-tren | |
| Mn-framework | CDs and Mn-framework | RTP | 529 | 664 | 25 | M-CDs | 82 |
| | | | | | | | |



Table 2 Self-protection CD-based RTP materials^a

| Method | Precursor | Afterglow mode (298 K) | Afterglow emission (nm) | Afterglow lifetime (τ) ms ⁻¹ (298 K) | PQY (%) | Remarks | Ref. |
|----------------------------------|---|------------------------|--------------------------|--|--------------------------|-----------------------------|------|
| Hydrothermal | Citric acid and ethylenediamine | RTP | 440 | 0.16 | NA | CDs | 86 |
| Hydrothermal | PVA and ethylene diamine | RTP | 564 | 13.4 | NA | NCD | 87 |
| Hydrothermal | Polyacid and diamine | RTP | 494 | 658.11 | NA | CDs | 88 |
| Microwave-assisted | Ethanolamine and phosphoric acid | RTP | 535 | 1460 | NA | URTP CDs | 89 |
| Heat treatment | Laspartic acid and D-glucose | RTP | 515 | 747 | NA | NCDs | 90 |
| Solvothermal | Glucose and (C ₂ H ₅) ₃ N·3HF | RTP | 455 | 1210 | 3.45 | FNCDs | 91 |
| Microwave-assisted | Triethanolamine and phosphoric acid | RTP | 518 | 820 | 15.85 | P-CDs | 92 |
| Hydrothermal | Acrylamide, urea and citric acid | RTP | 519 | 459 | NA | NCDs | 93 |
| Heat treatment | Urea and phosphoric acid | RTP | 495 | 320 | 23 | SW-CPDs | 94 |
| Hydrothermal | Isophthalic acid and ethylenediamine | RTP | 510 | 769 | NA | N-CDs | 95 |
| Carbonization | Citric acid and 2,4,6-trihydrazinyl-1,3,5-triazine | RTP | 476 | 66.4 | 4.7 | C-dots | 96 |
| Hydrothermal | Citric acid (CA) and basic fuchsine | RTP | 550 | 51.9 | 28 | N-CDs | 97 |
| Solvent-free pyrolysis treatment | Hydroxyurea | RTP | 570 | 175 | 13 | FP-CDs | 98 |
| Solvothermal | 5,5'-Disulfanediylbis(2-nitrobenzoic acid) | RTP | 426/520 | 1.1 | NA | HCDs | 99 |
| Solvothermal | Phenol, cytosine, resorcinol, and phloroglucinol | RTP | 500 505 520 | 65 127 218 | NA | F, O-codoped CDs | 100 |
| Microwave-assisted | Polyethylenimine and phosphoric acid | RTP | 515 | 565.19 | NA | N-CDs | 101 |
| Solvothermal | Fructose and diethylenetriamine | RTP | 500 | 1140 | 8.3 | FNCDs | 102 |
| Hydrothermal | Diethylenetriamine, phosphoric acid and boric acid | RTP | 509 535 567 603 | 481 511 437 426 | 8.7 6.3 3.2 1.5 | CPDs | 103 |
| Hydrothermal | Levofloxacin | RTP | 555 630 | 354 237 | 4.2 | CD@paper | 104 |
| Solvothermal | Urea, HCl, citric acid, DMF | RTP + DF | 500 572 | 314 462 | 5.8 | CD powder | 105 |
| One-step melting | Boric acid | RTP | 512 495 560 | 1740 | 66.13 | (NACA) _{0.01%} /BA | 106 |
| Hydrothermal | Silica and hexamethyleneimine | RTP + TADF | 508 | 451 | NA | CDs@SiO ₂ | 107 |
| Solvent-free catalytic assistant | <i>o</i> -Phenylenediamine and AlCl ₃ ·6H ₂ O | RTP | 554 | 243.2 | 2.8 | M-CDs | 108 |
| Pyrolysis | Citric acid and boric acid | RTP | 466–638 | 113.9–581.76 | 0.42–13.74 | B-CD | 109 |
| Microwave-assisted | 1-[3-(Trimethoxy silyl) propyl]urea and phosphoric acid | RTP | 480–560 | 1000 | 59.41 | CPDs | 110 |

^a NA: not available; PQY: phosphorescence quantum yield; TADF: thermally activated delayed fluorescence; RTP: room temperature phosphorescence; DF: delayed fluorescence; PVA: polyvinyl alcohol.

capable of accommodating luminescent species. As such, it is capable of accommodating CDs to achieve RTP, particularly silica, and is widely applied for the generation of CD RTP.^{53–60,107,118} For example, Li *et al.*⁵⁴ proposed a strategy for achieving ultra-long RTP in air-saturated aqueous media based on carbon dot-based silica composites (CDs@SiO₂) (Fig. 3B). The formation of Si–O–C bonds between CDs and silica during TEOS hydrolysis acts as a scaffold for the nucleation and the growth of the silica framework. The CDs are able to covalently attach to the silica network and the silica acts as a matrix that

contributes to the dispersion of the CDs, providing protection from environmental bursts such as water and oxygen. More importantly, the abundance of silanol groups on the surface of the composites gives the whole hybridized system a good hydrophilic character. And also, Sun *et al.*⁵⁷ designed and developed a strategy to fabricate metal-free multi-confined CDs (CDs@SiO₂) within SiO₂ by generating an effective multi-confinement effect (MCE) to develop room temperature phosphorescent materials with simultaneous ultra-long lifetime, high phosphorescence quantum efficiency and



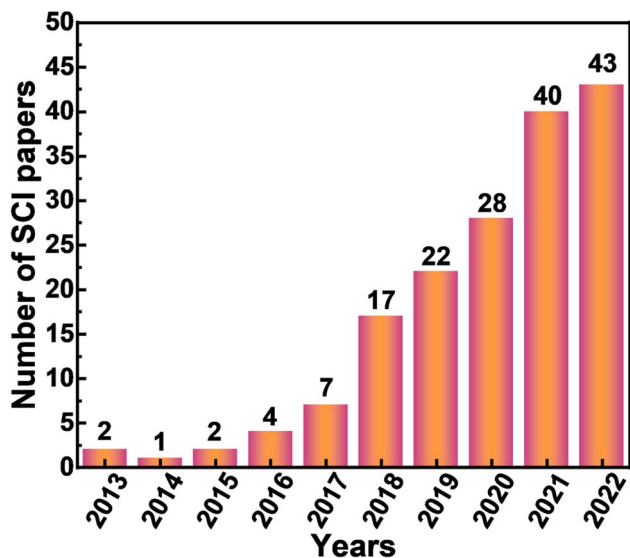


Fig. 2 The number of articles related to CD-based RTP since 2013; search results were obtained from the Web of Science, on 31st August 2022.

excellent stability (Fig. 3C). In addition, zeolites are a class of matrix-assisted materials widely utilized to achieve ultra-long phosphorescence lifetimes, and they are crystalline

aluminosilicates or aluminophosphates with a three-dimensional 4-connected structure and a uniform pore size of less than 2 nm.^{63–68,119} For instance, Wang *et al.*⁶⁴ have achieved a facile strategy to modulate the RTP properties of CDs through donor–acceptor energy transfer in the CDs–zeolite system by introducing heteroatoms into the aluminium phosphate zeolite backbone to construct an efficient donor–acceptor system that promotes exchange coupling between the CD exciton and the dopant in the matrix (Fig. 3D). Furthermore, the confinement effect of the crystal structure not only immobilizes T₁ but also prevents non-radiative relaxation of T₁ and is also a promising method for generating CD-based RTP. Crystal structures can be divided into two categories: organic crystal structures and inorganic crystal structures. The organic crystal structures include urea, cyanuric acid and melamine, while inorganic crystal structures include sodium chloride, sodium hydroxide, boric acid and so on.^{46–48,70,72–76,78,79,82,120–130} For example, Wang *et al.*¹²⁴ proposed a molten salt method for the preparation of CD-based RTP materials by selecting high charge density magnesium salts and phosphates as doping salts, and calcining the carbon source directly in the presence of inorganic salts (Fig. 3E). During the melting and recrystallization process, CDs are formed and incorporated into a matrix with a special crystal structure. As magnesium phosphate is insoluble in water, the solid matrix provides rigid protection for the CDs. As a result, the CDs are non-phosphorescent as monomers, but the RTP

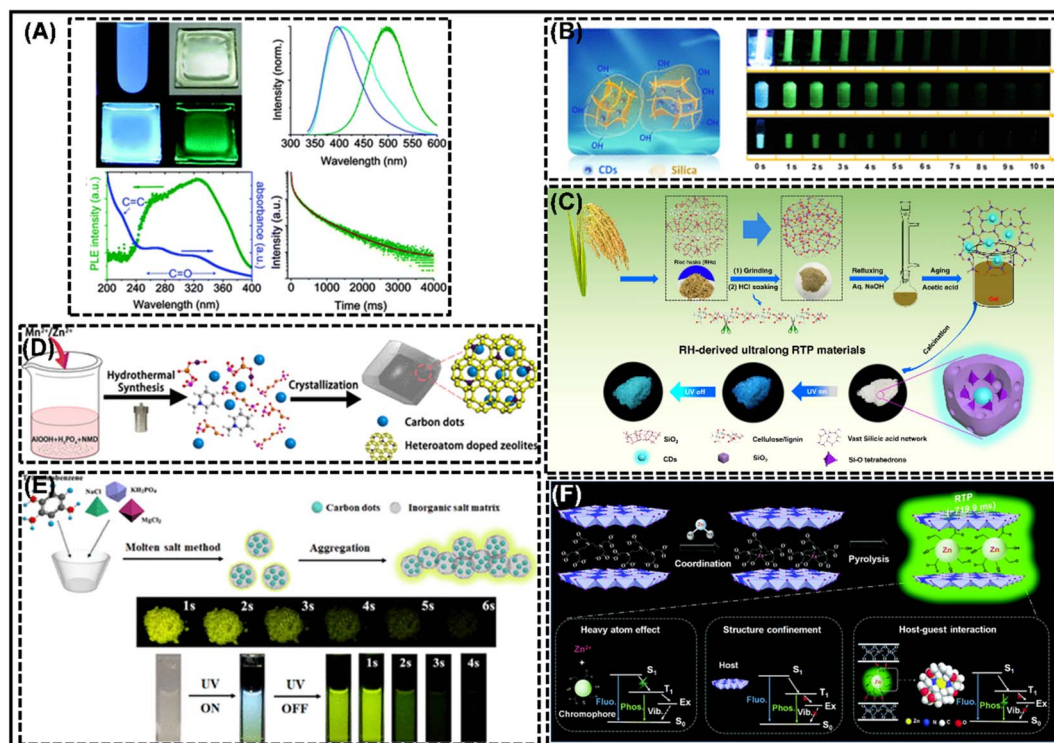


Fig. 3 Matrix-assisted synthesis of CD-based RTP materials. (A) Reprinted with permission from Deng *et al.*⁴² Copyright 2013, Royal Society of Chemistry. (B) Reprinted with permission from Li *et al.*⁵⁴ Copyright 2019, American Chemical Society. (C) Reprinted with permission from Sun *et al.*⁵⁷ Copyright 2019, Springer. (D) Reprinted with permission from Wang *et al.*⁶⁴ Copyright 2019, American Chemical Society. (E) Reprinted with permission from Wang *et al.*¹²⁴ Copyright 2019, American Chemical Society. (F) Reprinted with permission from Kong *et al.*⁵² Copyright 2019, Royal Society of Chemistry.



phenomenon is initiated and enhanced by the aggregation of the salt matrix. Besides this, structural confinement displays a unique advantage by reducing the rate of the radiative relaxation process. A number of special structural matrices have also been used for the realization of CD RTP, such as two-dimensional (2D) layered double hydroxides (LDHs) which have attracted significant interest in the available confinement matrix owing to their versatile chemical composition and layer charge. Significantly, LDHs have a nanoscale space that provides a constrained and rigid environment for a number of chromophores that exhibit enhanced fluorescence properties (intensity, efficiency, lifetime, *etc.*) by suppressing the non-radiative relaxation of single-linear excitons.⁵² Therefore, exploring the relationship between the relaxation paths of triplet-state excitons and the rigidity of LDHs will help to achieve optimal luminescence efficiency and to develop a new class of RTP materials. Kong *et al.*⁵² employed LDHs as a matrix and proposed the design principle of activation to achieve RTP on CDs through three synergistic effects (structure-bound effect, heavy atom effect, and chemical bonding) (Fig. 3F). The confined and rigid environment of LDHs suppressed the non-radiative deactivation of CD triplet excitons and improved the luminescence efficiency. In conclusion, in addition to the above matrix-assisted materials, there are also other appropriate matrix-assisted materials suitable for achieving RTP of CDs. Therefore, the appropriate selection of matrices is key to achieving efficient and ultra-long RTP CDs.

2.2 Self-protection synthesis of CD-based RTP materials

Although the RTP of CDs in solid and water-disperse states has been achieved, the CD-based RTP materials prepared by matrix-assisted methods are mostly heterogeneous systems with poor thermal stability and electrical conductivity, which hinders their practical application. Therefore, to overcome these issues, the development of matrix-free self-protected phosphorescent CDs is an urgent problem to be solved. Aiming to generate self-protected RTP of CDs, the choice of the raw material is crucial. Currently, researchers often choose polymers or their monomers, cross-linkable reactants, molecules doped with heteroatoms and larger conjugated structures as precursors.^{86–96,98–109,131,132} The relevant achievements are shown in Table 2.

The earliest use of self-protection methods to achieve the RTP of CDs dates back to 2014, when Yan *et al.*⁸⁶ employed citric acid and ethylenediamine as carbon and dopant sources to achieve the RTP of CDs, but the phosphorescence lifetime at that time was only 160 ms, and thus this result did not attract widespread attention. Until 2016, Chen *et al.*⁸⁷ achieved the RTP of CDs in one step using PVA and ethylenediamine as reaction precursors, with a phosphorescence lifetime of up to 13.4 ms. At this time, the RTP of CDs gradually came into the limelight, and the research frenzy gradually surged. Later, Jiang *et al.*¹³¹ by simple heat treatment of ethylenediamine and phosphoric acid could produce unexpectedly long room temperature

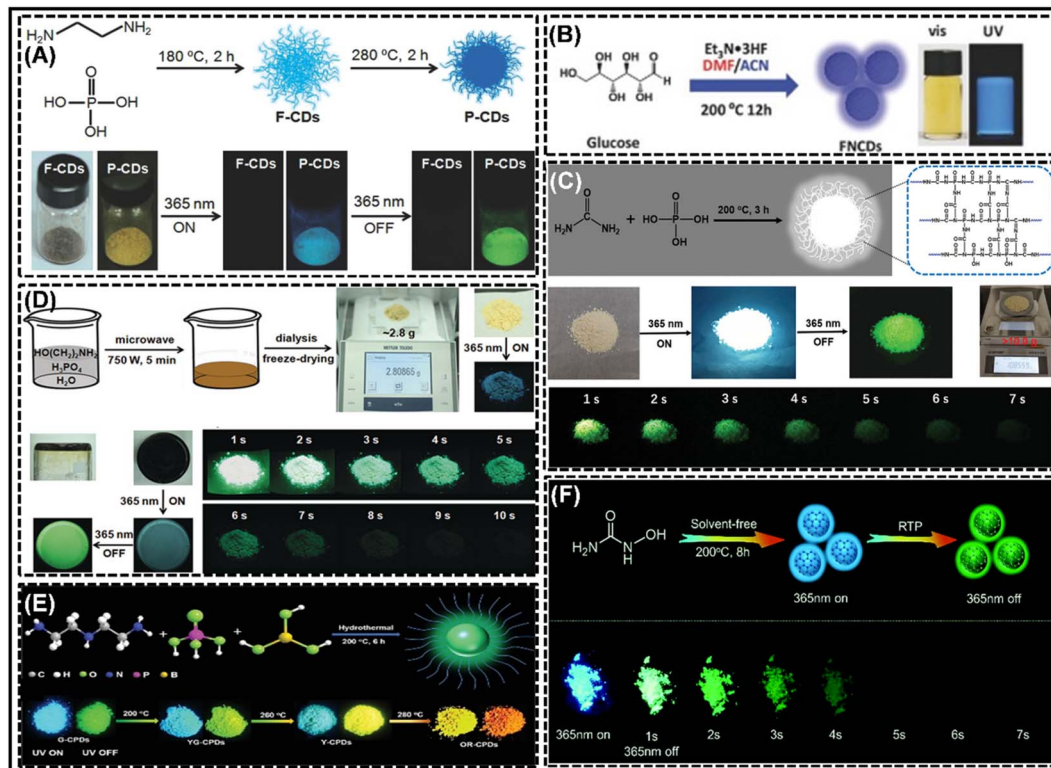


Fig. 4 Self-protection synthesis of CD-based RTP materials. (A) Reprinted with permission from Jiang *et al.*¹³¹ Copyright 2018, Wiley-VCH. (B) Reprinted with permission from Long *et al.*⁹¹ Copyright 2018, Wiley-VCH. (C) Reprinted with permission from Wang *et al.*⁹⁴ Copyright 2020, Wiley-VCH. (D) Reprinted with permission from Jiang *et al.*⁸⁹ Copyright 2018, Wiley-VCH. (E) Reprinted with permission from Wang *et al.*¹⁰³ Copyright 2021, Wiley-VCH. (F) Reprinted with permission from Zhao *et al.*⁹⁸ Copyright 2021, Royal Society of Chemistry.



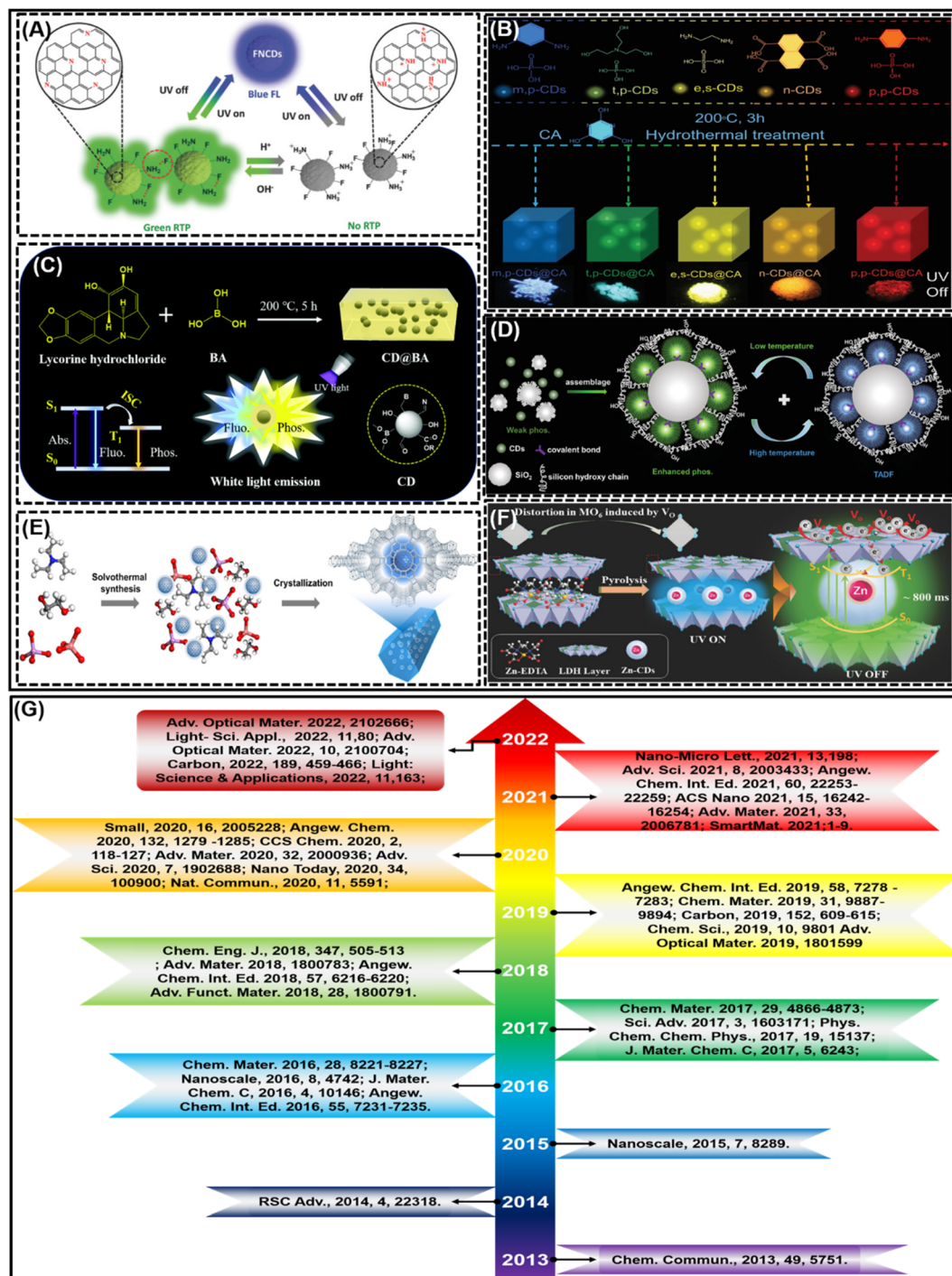


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successfully prepared by Tan *et al.* via treating oil-soluble N-doped carbon quantum dots by a one pot hydrothermal method.¹³⁴ The obtained ACDs may be homogeneously dispersed in PVA and the PU matrix. Furthermore, RTP can be recognized in these ACD based composites. This is the first time that ACDs realize phosphorescent emission in polymers, which

greatly broadens the research and application scope of CQDs. In 2017, Chen *et al.*⁸⁷ reported for the first time the aggregation-induced RTP of self-quenching-resistant nitrogen doped CD powder by structure design *via* using PVA-chains, and the potential application of the temperature sensor is preliminarily prospected. Jiang *et al.*⁵³ also reported a new method for



method, CDs are embedded into the growing zeolite crystals with the maximum extent, and the non-radiative transition of CDs is surpassed by the strong host-guest interaction, and thus composite materials with ultra-long double emission of TADF and RTP are prepared, and have a lifetime of 1.7 s and 2.1 s, respectively, as well as a QY of 90.7% and a PQY of 24.4%, respectively. These values are at the top superiority level among CD-based PL materials, and thus, represent the majority of organic afterglow materials. What's more, in 2021, Li *et al.*⁷⁹ reported a method to prepare highly efficient RTP materials from crystalline heat-annealed CDs and BA composites which can induce amorphous to crystalline transition by grinding. This method can enable CDs to be uniformly embedded in BA crystals to the maximum extent, reduce the non-radiation attenuation of CDs, and promote the cross-connections between systems by suppressing the free vibration of CDs, thus generating strong RTP materials. The reported PQY is the highest (48%). It is well known that water-soluble red afterglow imaging agents have a great penetration depth and nondurable excitation characteristics which have potential application prospects in time-gated afterglow bioimaging. Thus, Liang *et al.*⁵⁹ reported a red afterglow imaging agent composed of Rhodamine B (RhB) and CNDs, which were confined in a hydrophilic silica shell to form CNDs-RhB@silica nanocomposites. CNDs-RhB@silica can achieve a luminescence lifetime and afterglow QY of 0.91 seconds and 3.56%, respectively, which is the best result for the red afterglow region. Liang *et al.*⁶⁰ also reported a time division duplex technology based on environmentally friendly CNDs with controllable luminescence lifetime. It was the first time they demonstrated that the time-division duplexing technique for CNDs and CNDs@silica is independent of the emission color and intensity depending on the phosphorescence lifetime under control. Water has been demonstrated to play an essential role in modulating the luminescence lifetime of CNDs by quenching triple excitons. In addition, Tan *et al.*¹⁰⁴ also reported a method to realize time-dependent phosphorescence colors in CDs, which were synthesized through a one pot hydrothermal method by using levofloxacin as the raw material. They realized a new type of time-dependent phosphorescence color that changed from orange to green in a very short time (1 s). In 2021, Zheng *et al.*¹³⁷ also reported a general approach in which effective radiative energy transfer can be applied to support the reabsorption of upconversion materials (UMs) into CD-based room temperature near-infrared excited multicolor afterglow materials (CDAMs). Please note that this is the first report on the multicolor afterglow of near-infrared excited in CDAMs. More importantly, this work provides a general route for constructing novel room-temperature afterglow materials with tunable excitation wavelengths. For the first time, Zhou *et al.*¹³⁸ proposed an approach for efficient energy transfer mediation to boost the RTP of CDs by incorporating pure phosphorescent CDs into the afterglow matrix. In this system of design, there is a significant increase in the emission intensity, RTP lifetime and emission time of CDs, and all CD-based materials emit visible phosphorescence for more than 20 seconds after UV excitation. In addition, Mo *et al.*¹³⁹ provided

the first example of visible light excited TADF in aqueous solution by confining the co-doping of CDs with fluorine and nitrogen in silica nanoparticles (F, NCDs@SiO₂). Although there have been many reports on RTP materials, it is still a challenging task to realize ultra-long RTP in aqueous media, especially for CD-based materials. In 2022, Jiang *et al.*⁷³ developed a robust organic long persistent luminescence (OLPL) system with hour-level afterglow emissions through simple microwave-assisted heating of a mixture of m-CDs and urea. It is a very uncommon case of an OLPL system displaying hourly afterglow under ambient conditions, even for aqueous media. Further studies showed that the generation of covalent bonds between cyanuric acid and CDs played a key role in the afterglow presentation. Green preparation has always been the synthetic route pursued by everyone. At present, RTP materials are greatly developed. Nevertheless, it is a huge priority to achieve both multicolor and long wavelength RTP emission with favorable stability in CD-based RTP materials. Liang *et al.*¹⁴⁰ proposed a new and general "CDs-in-YOHF" scheme to yield multicolor and long wavelength RTP by confining different CDs to a Y(OH)_xF_{3-x}(YOHF) matrix. It should be noted that the RTP lifetime of the orange emissive CDs-o@YOHF is the longest in the reported single-CD-matrix composite materials with emission above 570 nm. Compared with the common representative matrices, the YOHF matrix is also proved to be more effective in protecting the long-wavelength triplet emission of CDs-o. Similarly, Mo *et al.*⁶¹ reported a newly developed strategy to incorporate phosphorescent CD and fluorescent dyes into monodisperse silica nanoparticles below 20 nm to achieve multi-color long afterglow in aqueous solution. For the first time, CD-based multicolor long afterglow systems (green, yellow, orange and red) were fabricated in aqueous solution by cascaded Förster resonance energy transfer. Especially, under UV excitation, a prolonged red afterglow with a Stokes shift of 255 nm was developed. So far, there have been few studies on thermal stimulated response photoluminescence of CD-based materials. Xu *et al.*¹⁴¹ reported for the first time a polymeric nanocomposite incorporating fluorinated CDs (FCDs) that can be efficiently synthesized in large quantities through the utilization of commercial water-soluble polymer sodium carboxymethylcellulose (CMCNa) as a stable matrix. The synthesized FCDs-CMCNa has bimodal emission, *i.e.*, both solid state fluorescence and in-room RTP. It is more interesting to note that FCDs-CMCNa exhibits special temperature-sensitive optical properties when the temperature is reduced from 300 K to 90 K. It exhibits an increase in fluorescence/phosphorescence intensity with decreasing temperature up to the switching point of 150 K, which then gradually decreases with decreasing temperature to 90 K. Later, Liu *et al.*¹⁴² proposed a series of flexible dynamic ultra-long RTP polymer composites, which are illuminated by halogen doped CDs and have fully programmable emission. They had produced a transparent, flexible and fully programmable dynamic ultra-long RTP composite film with reliable gray-scale display ability from the synthesized RTP material for the first time. As can be seen, each year's contribution has grown year on year, indicating a steady stream of scholars working on CD-based



RTP materials with notable achievements. In each year of the prominent work represented, RTP of CDs was slowly developed in the initial stage, however, three years later, it was springing up. This indicates that as research progresses, a steady stream of findings is discovered that continue to advance the development of RTP for CDs.

To sum up, the outstanding work of CD-based RTP materials is growing every year. According to Fig. 2, we conducted statistical analysis on all the 166 retrieved papers and summarized the corresponding RTP emission wavelengths (it is worth noting that the number of wavelengths counted here is more than the number of papers published because some articles reported multicolor wavelengths), as shown in Fig. 4. According to the statistical results, the emission of CD-based RTP materials is mainly concentrated in the range of 500–550 nm (Fig. 6a), accounting for 40.68% of the total (Fig. 6b). In other words, the current CD-based RTP materials mainly emit green light. The second is blue to green emission with emission wavelengths concentrated between 450 and 500 nm, followed by yellow-orange emission with emission wavelengths concentrated between 550 and 600 nm. It can be seen that the achievable emission of CD-based RTP materials is mainly concentrated in the short wavelength (450–600 nm) region of visible light, while reports of long wavelength emission (>600 nm) are very rare. Certainly, the realization of long-wavelength emission from CD-based RTP materials, even extending into the near-infrared region, is the focus of current research and the key to enhancing and broadening their application areas. This is a challenging research problem which needs to be solved.

4. Annual representative studies of phosphorescence lifetime in CD-based RTP materials

RTP materials have attracted much attention because of their great optical application potential. With the deepening of

research, researchers have made breakthroughs in CD-based RTP materials, such as multi-mode emission RTP materials,^{72,111} multi-color RTP materials,^{48,75,120,140,143} aqueous solution RTP materials,^{55,59,61,86} multifunctional detection application RTP materials,^{43,92,134} biocompatible RTP materials⁵⁴ and high PQY^{79,94,135} and ultra-long phosphorescence life RTP materials.^{46,57,58,73,89,131} However, it is still difficult to obtain RTP materials with simultaneous long-lifetime and high PQY. In addition, phosphorescence lifetime is one of the important parameters to measure CD-based RTP materials. Phosphorescence lifetime is a key feature of RTP materials, and ultra-long phosphorescence lifetime is central to the excellent performance of RTP materials. The key to achieving long phosphorescence lifetimes is the choice of the RTP material structure or the implementation of methods, such as the use of melt co-crystallisation¹²⁵ to immobilize CDs in the form of wrapping to achieve RTP (Fig. 7A), the utilization of rigid structural networks and the coexistence of covalent bonds⁵⁷ to immobilize CDs in a three-dimensional spatially confined way to achieve RTP (Fig. 7B), the adoption of porous space crystalline materials⁶⁶ to immobilize CDs by adsorption and intercalation to achieve RTP (Fig. 7C), the employment of polymers to covalently cross-link CDs⁴⁵ to achieve RTP (Fig. 7D), the employment of metal-organic frameworks with porous structures providing active sites⁸⁰ to immobilize CDs to achieve RTP (Fig. 7E), and the utilization of high strength rigid crystal structures⁷² to immobilize CDs to achieve RTP (Fig. 7F). Each year's work has made corresponding outstanding contributions, from the initial millisecond level to the later second level, and then to the current hour level, which are major breakthroughs one after another. As shown in Fig. 7G, the best phosphorescent life of CD-based phosphorescent materials is shown in each year from the first discovery to the present. In 2013, Deng *et al.*⁴² reported for the first time that the phosphorescence phenomenon of CDs in the PVA matrix was observed, and its average phosphorescence life was as high as ~380 ms. Then, in 2014, Yan *et al.*⁸⁶ reported for the first time

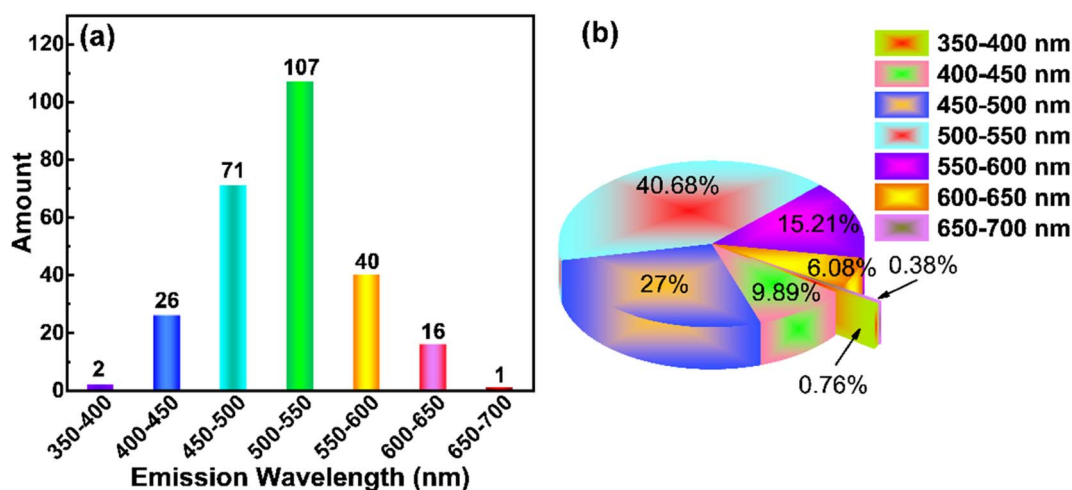


Fig. 6 (a) Distribution of phosphorescence emission of CD-based RTP. (b) The proportion of phosphorescence emission of CD-based RTP in the total number of reported articles.



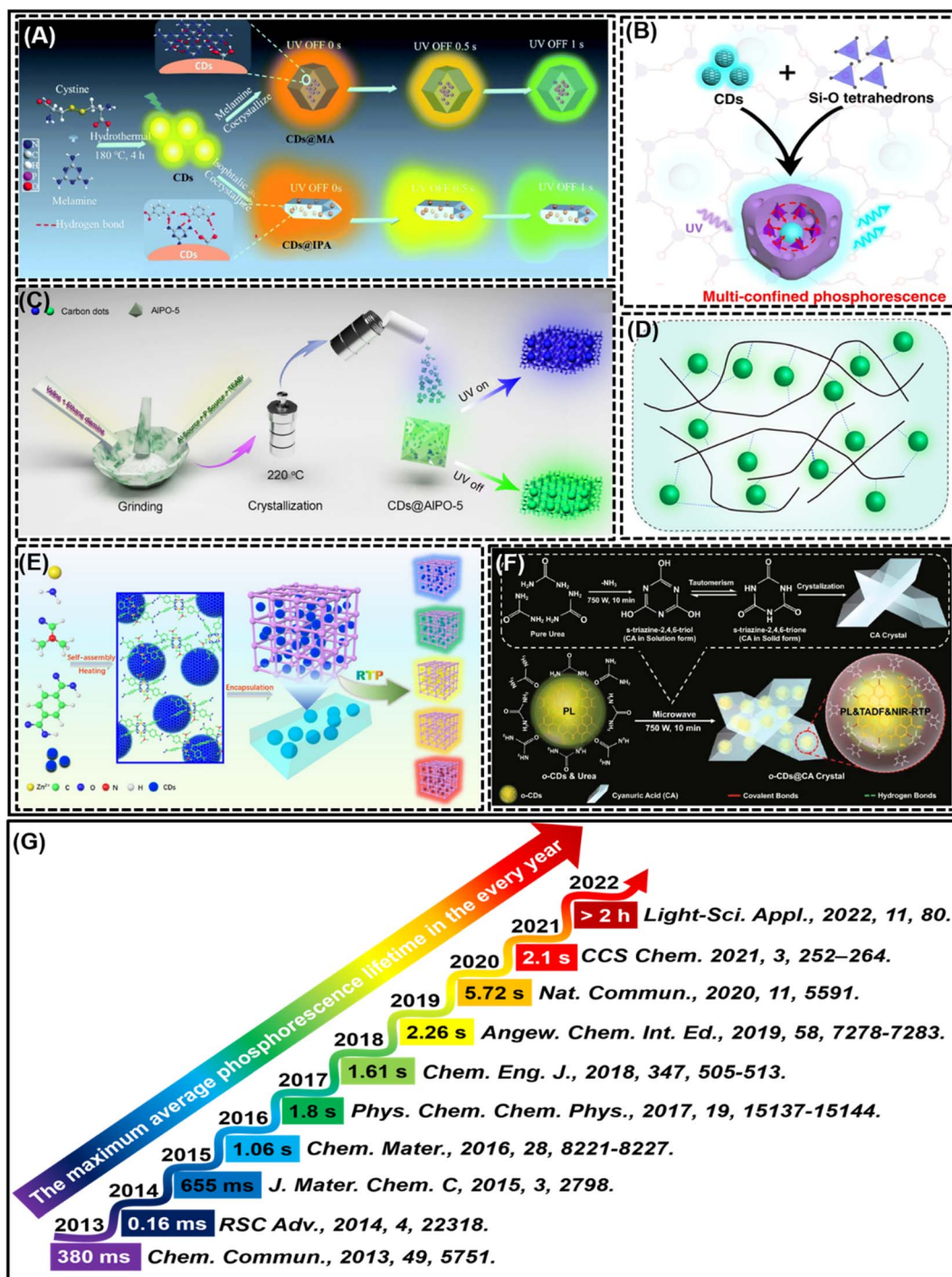


Fig. 7 (A) Reprinted with permission from Qu *et al.*¹²⁵ Copyright 2021, Royal Society of Chemistry. (B) Reprinted with permission from Sun *et al.*⁵⁷ Copyright 2019, Springer. (C) Reprinted with permission from Zhang *et al.*⁶⁶ Copyright 2020, Chinese Chemical Society. (D) Reprinted with permission from Cao *et al.*⁴⁵ Copyright 2022, American Chemical Society. (E) Reprinted with permission from Xu *et al.*⁸⁰ Copyright 2022, American Chemical Society. (F) Reprinted with permission from Wang *et al.*⁷² Copyright 2021, Springer. (G) The longest phosphorescence lifetime of CD-based RTP materials in each year.

that the phosphorescence of CDs in pure aqueous solution was observed, but its average phosphorescence lifetime was only 0.16 ms. In 2015, Deng *et al.*⁴⁶ embedded CDs into a KAl(SO₄)₂·x(H₂O) matrix to prepare composite powder of CDs, and presented long-lived RTP with an average phosphorescence life

of up to 655 ms. In 2016, Li *et al.*⁴⁷ prepared an efficient CD-based phosphorescent material, that is, NCDs were heated in one pot by treating a mixture of urea and NCDs, and then they were incorporated into the composite matrix. The resulting material had an ultra-long phosphorescent life of 1.06 s under



the excitation of 280 nm. In 2017, Joseph *et al.*⁶² embedded CDs in a silica gel matrix to obtain an RTP material. The CDs showed strong blue fluorescence in aqueous dispersion and showed green afterglow when combined with silica gel. Under the excitation of 380 nm, the phosphorescent emission lifetime of the CDs is about 1.8 s, which is the highest value of CDs in a solid matrix. In 2018, He *et al.*⁴³ combined CDs with PVA *via* the help of electrospinning technology, and realized RTP and TADF at the same time. In addition, these nanofibers showed a longer average afterglow lifetime of 1.61 s and a visual recognition period of 9 s. In 2019, Li *et al.*⁴³ realized the preparation of RTP of CDs with or without heteroatom doping through a single-step thermal treatment of CDs and boric acid. This composite exhibits the highest phosphorescence lifetime of 2.26 s and a PQY of 17.5%, and this is the maximum record for CD-based RTP materials to date. In 2020, Sun *et al.*⁵⁷ obtained RTP materials with ultra-long life, high-level PQY and superior stability by rational design and fabrication of multi-constrained CDs. The designed multi-constrained CDs possess an ultra-long lifetime of 5.72 seconds, 26.36% PQY, and outstanding stability to strong oxidants, acids, bases, and polar solvents. In 2021, Yu *et al.*⁶⁸ systematically adapted the reaction parameters and host-guest interactions to achieve nine green RTP CDs-in-zeolite (CDs@zeolite) composites with engineering lifetimes ranging from 0.38 to 2.1 s under solvent-free conditions. In 2022, Jiang *et al.*⁷³ developed the first CD-based organic long persistent luminescence system with a duration of more than 1 h. A system based on CDs (named m-CDs@CA) is reported, which can be conveniently and efficiently fabricated through the utilization of a household microwave oven. What is even more impressive is that its long-term sustained luminescence may be noticed under ambient conditions, even in aqueous media. In summary, the extension of phosphorescence lifetimes has always been at the heart of scholars' pursuits, and shows that phosphorescence lifetimes occupy an irreplaceable and central position in the RTP of CDs, and are

a sign that CDs exhibit RTP properties. Progress in the field of research can be seen in the representative work each year, and in the advancement of scientific capabilities. Following on from the previous research, the phosphorescence lifetime of CD RTP has made a qualitative breakthrough, extending from the initial millisecond level to the hourly level, which provides favorable preconditions for the practical application of CDs RTP.

Phosphorescence lifetime is an important index to measure RTP materials, and the phosphorescence lifetime of CD-based RTP materials is also constantly developing. According to Fig. 2, the phosphorescence lifetime of the published CD-based RTP materials was statistically analyzed. As shown in Fig. 8a, CD-based RTP materials with different phosphorescence lifetimes have been reported one after another, but the main phosphorescence lifetimes are between 1 and 1000 ms. More obviously, the phosphorescence lifetime of most CD-based RTP materials is still at the millisecond level. Of course, there are also some outstanding performances, such as the existence of phosphorescence lifetime at the level of hours, but this is rare. According to the analysis of statistical data, the ratio of phosphorescence lifetime between 1 and 1000 ms accounts for 67.76%, which is a very high percentage, while the ratio between 1000 and 5000 ms reaches 25.66% (Fig. 8b). It can be seen that the preparation of CD-based phosphor materials with second phosphorescence lifetime is still in the stage of development. It is worth noting that CD-based RTP materials with all second phosphorescence lifetimes account for less than half of all reported CD-based RTP materials. This is a very terrible existence. Therefore, developing CD-based RTP materials with ultra-long phosphorescence lifetime is a hot spot and a difficult problem at present.

5. Applications

Realizing the final application is the value of a material. As new luminescent materials, CD-based RTP materials are no

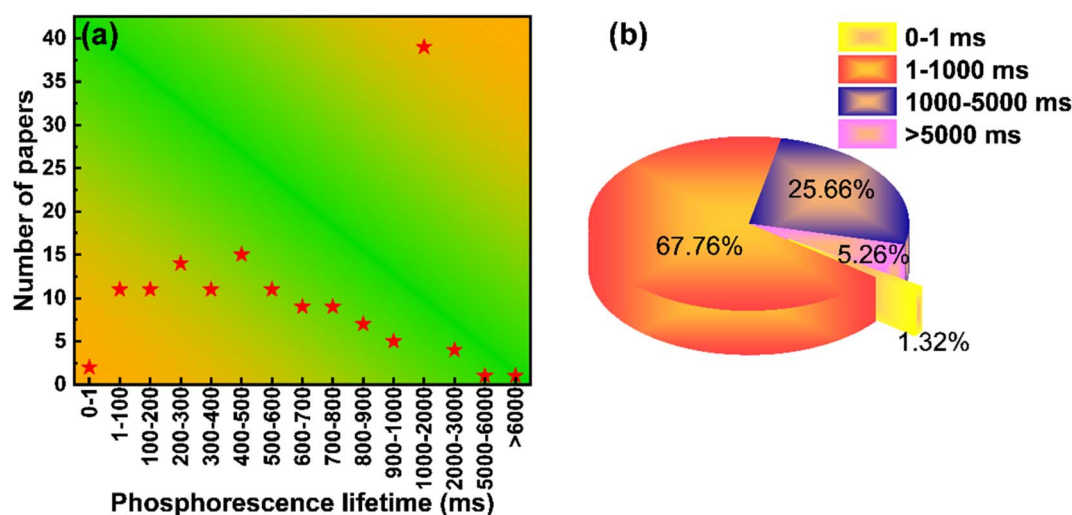


Fig. 8 (a) Distribution of phosphorescence lifetime of CD-based RTP. (b) The proportion of phosphorescence lifetime of CD based-RTP in the total number of reported articles.



Table 3 Applications of CD-based RTP materials

| Application | Method | Afterglow emission (nm) | Afterglow lifetime (τ) ms ⁻¹ (298 K) | Remarks | Ref. | | |
|-------------------------|---|---|---|--------------------------|--------------|------------|----|
| Anti-counterfeiting | A composite film of m-CDs (CDs prepared from <i>m</i> -phenylenediamine) and polyvinyl alcohol (PVA) | 500 | 456 | CDs-PVA | 111 | | |
| | Microwave-assisted heating of ethanolamine and phosphoric acid aqueous solution | 535 | 1460 | URTP CDs | 89 | | |
| | <i>In situ</i> in the hydrothermal system by using an organic amine as the template and the precursor of the CDs and an inorganic crystalline framework as the matrix | 620 | 10.94 | CDs@MnAPO-CJ50 | 81 | | |
| | By one-step heat treatment of CDs and boric acid (BA) | 490–570 | 2260 | a-CDs/BA | 120 | | |
| | One-step solvothermal treatment | 455 | 1210 | FNCDS | 91 | | |
| | Hydrothermal treatment of trimellitic acid | 560 | 183.6 | TA-CDs | 136 | | |
| | A hydrothermal reaction of succinic acid and diethylenetriamine | 500–575 | 880 | MP-CDs | 144 | | |
| Information encryption | Thermal-treated CDs into a PVA matrix | 420–440 | >100 | CD@PVA | 113 | | |
| | A hydrothermal treatment of nSiO ₂ solution mixed with m-CD solution | 470 | 703 | m-CDs@nSiO ₂ | 53 | | |
| | Using polyacid and diamine to synthesize PCDs through a one-step hydrothermal treatment | 494 | 658.11 | PCDs | 88 | | |
| | A further heat treatment of the F-CD powder at a higher temperature | 538 | 1390 | F-CDs | 131 | | |
| | By <i>in situ</i> embedding CDs within a series of zeolitic crystalline matrices under solvothermal/hydrothermal conditions | 430 | 350 | CDs@zeolite | 63 | | |
| | By utilizing CDs and melamine to construct hydrogen-bonded networks to form a polymer | 529 | 664 | M-CDs | 82 | | |
| | By pyrolysis of citric acid and boric acid precursors | 466–638 | 113.9–581.76 | B-CD | 109 | | |
| Biological imaging | One-pot hydrothermal treatment of levofloxacin | 555/630 | 354/237 | CD@paper | 104 | | |
| | By confining CNDs in a silica encapsulation layer | 520 | 1860 | WSP-CNDs @ silica | 58 | | |
| | Neutral red, KNO ₃ , MgCl ₂ and KH ₂ PO ₄ as precursors <i>via</i> the molten salt method | 518/520 | 546/616 | CD71 | 180 | | |
| | By confining the fluorine and nitrogen co-doped CDs in silica nanoparticles | 500 | 480 | F, NCDs@SiO ₂ | 139 | | |
| | One-step solvent-free catalytic assisted strategy | 554 | 243.2 | M-CDs | 108 | | |
| | Light-emitting diodes | WLED | A one-pot heat treatment of a mixture of urea and NCD | 490 | 1060 | HN powders | 47 |
| | | LEDs | CD-based composites were prepared by simply mixing CD solution and NaOH under constant stirring | 480 | 705 | CD/CA | 70 |
| LEDs | | <i>In situ</i> solvent-free thermal crystallization | 530 | 2100 | CDs@AlPO-5 | 66 | |
| WLED | | One-step heat treatment of lycorine hydrochloride and boric acid | 520 | 1760 | CD@BA | 77 | |
| WLED | | A one-step heat treatment process of urea and phosphoric acid aqueous solutions | 495 | 320 | SW-CPDs | 94 | |
| WLED | | Encapsulating multicolor lignin-derived CDs and PVA into a delignified wood framework | 505 | 208.83 | LTW | 176 | |
| WLED | | Through the solvothermal reaction of polyvinylpyrrolidone (PVP), urea, and seed CDs | 501–596 | 419 | CD powder | 143 | |
| pH and Fe ³⁺ | | One step hydrothermal | 440 | 0.16 | CD-based RTP | 86 | |



Table 3 (Contd.)

| Application | | Method | Afterglow emission (nm) | Afterglow lifetime (τ) ms ⁻¹ (298 K) | Remarks | Ref. |
|---------------------|------------------|---|-------------------------|--|---------------|------|
| Sensing detection | pH | Microwave strategy to synthesize P-CDs by using triethanolamine serving as the carbon source with phosphoric acid | 518 | 820 | P-CDs | 92 |
| | Fe ³⁺ | <i>In situ</i> green synthesis of CDs by using Schisandra chinensis polysaccharide as the only carbon source | 510 | 271.2 | CD/PVA | 114 |
| | Humidity | By combining chelate and hydrothermal methods | 525 | 1.34 | Eu-CDs/PV | 186 |
| | Humidity | By embedding biomass derived CDs into cellulose fibrils | 560 | 167.31 | CDs@Cellulose | 85 |
| | Fe ³⁺ | The CD solution mixed with CA under constant stirring | 480 | 687 | CD-CA | 71 |
| | O ₂ | By intercalating the CD precursor (EDTA) into the LDH interlayer | 525 | 386.8 | CDs-LDHs | 50 |
| Temperature sensing | | Hydrothermally synthesized using PVA and ethylene diamine | 564 | 13.4 | NCD | 87 |
| | | By direct calcination of carbon sources (1,2,4-triaminobenzene) with inorganic salts | 506 | 1280 | CDs@MP | 124 |
| | | By incorporating CDs into poly(vinyl alcohol) (PVA) with the assistance of electrospinning technology | 569 | 1610 | CDs/PVA | 43 |

exception. First of all, some CD-based materials have the characteristics of DF, which have the property of long life and can be used in the field of information security without prompt excitation. Secondly, some CD-based materials with DF characteristics have also been accompanied by RTP performances, and such multiple afterglow emission offers additional possibilities for information encryption and anti-counterfeiting applications. In addition, RTP materials based on CDs have strong sensitivity characteristics in some special detection, and are also used for response detection of ions, pH, temperature, time, fingerprint and so on. It is worth noting that CD-based RTP materials also have low toxicity and good biocompatibility, and also have great potential and development in the field of biological imaging. More importantly, CD-based RTP materials fully embody the characteristics of light-emitting materials, have good tunability, luminescence, stability and other optical properties, and have been greatly developed in the field of light-emitting diodes (LEDs). Table 3 summarizes some main applications of CD-based RTP materials. In addition, the special properties of CD based-RTP materials, such as excitation-related, temperature-related and afterglow color adjustable properties, enable them to build multi-level security defense in the field of information encryption and anti-counterfeiting. The long afterglow life of CD based-RTP provides new possibilities for easier identification of authenticity and information content. It allows significantly increased time to visualize cell states, enables speedy detection, and also facilitates bioimaging and detection. Generally speaking, the RTP material based on CDs possesses huge application prospects in the field of anti-counterfeiting,^{110,126,127,144–150} information

encryption,^{113,115,118,119,121,123,128,151–160} sensor detection,^{114,124,129,130,147,161–174} LEDs,^{175–179} biological imaging,^{134,180,181} logic gates,¹³³ fluorescent ink^{112,182–184} and other fields^{116,117,122,185–195} with the properties of DF, TADF, long afterglow life and adjustable afterglow color.

However, the application of CD RTP is also limited by its own deficiencies. First, the clarity in the luminescence mechanism of CDs is lacking. Different types of CDs have been reported one after another since the development of CDs, but equally inconsistent types of luminescence mechanisms have emerged, which seriously affects the unified definition and future application of CDs and prevents a systematic unification. Secondly, the process for the preparation of RTPs for CDs is too harsh. The current implementation of RTP for CDs requires the assistance of a matrix, high temperature or multi-step synthesis, and the process requirements are too demanding and costly, limiting their preparation and application for large scale production. Then, the RTP emission wavelengths of CDs are too short-term. According to the reported articles, most of the RTP emission wavelengths of CDs are concentrated in the range of 480–550 nm, while there is relatively little RTP emission for long wavelengths (>550 nm), which severely limits the application of RTP characteristics of CDs for advanced information security and encryption. Finally, the PLQYs for CDs RTP are relatively low. There is an urgent need for green preparation of RTP CDs with high quality and high PLQY, especially with long wavelength emission. In the field of optoelectronic devices, especially for WLED applications, red emission is the key to achieving efficient WLEDs and strong luminescence is also the key factor to enhance device



performance, and the emission of current RTP CDs greatly hinders their application in optoelectronics. Thus, the current low PLQY and the emission of RTP CDs have greatly hindered their application in the field of optoelectronics.

6. Challenges and prospects

Although significant achievements in the synthesis and diversified application investigations of CD-based RTP composites have been made, there remain enormous challenges to face. The specific aspects are as follows:

First, since the discovery and development of CDs, the luminescence mechanism of CDs has always been questioned since it does not have a unified structure. At present, we can accept two main categories, namely, carbon core luminescence and surface state luminescence. Similarly, the luminescence of CD-based RTP materials, as a special property of CD luminescence, has not yet been clearly explained and defined. For the luminescence mechanism of CD-based RTP, there are different opinions, each with its own words. For example, it has been reported that CD-based RTP originates from C=N, C=O, C=F, hydrogen bonds, polymer matrices and so on. However, these remarks are all unilateral. Thus, it is essential to address systematically their mutual and distinctive features, as well as their structural evolution patterns during CD synthesis. At the same time, the formation process and luminescence mechanism of CD-based RTP are explored and summarized by combining some more advanced characterization technologies and theoretical calculations, so that future generations can better understand the luminescence mechanism of CD-based RTP materials.

Second, luminescence quantum efficiency is one of the important indicators to measure luminescent materials. As a new emerging carbon based fluorescent nanomaterial, the PLQY of CDs has always been a hot and difficult research topic, and improving the PLQYs of CDs has always been our goal. More importantly, the PQY of CD-based RTP materials is much lower than the PLQY of CD fluorescence, which makes improving the PQY of CD-based RTP materials an urgent need. In addition, compared with CD-based RTP materials, the DF characteristics of CDs require a smaller energy difference (between the excited singlet state and the excited triplet state), which will make the preparation of CD-based DF materials more difficult. Because this also requires that the spatial overlap between the HOMO and LOMO be very small, which in turn reduces the radiation transition rate and ultimately leads to a low PQY. However, reasonable structural design, purification and appropriate matrix selection are expected to solve the pressing problem of low PQY of CD-based RTP materials.

Third, the recognition ability of human eyes is limited. In the visible light region, the luminescence of CDs is mainly concentrated in the short wavelength region (<600 nm), and even the luminescence of CD-based RTP materials is mainly concentrated in the blue to green light range. These short wavelength lights are harmful to the human body. In addition, the luminescence of CD-based RTP materials is mainly concentrated in the solid state, and the luminescence in

solution is relatively less. These limitations have seriously hindered the application of CD-based RTP materials in multi-color displays and bio-imaging. Therefore, the development of CD-based RTP materials with a long wavelength and liquid phase luminescence has also become a hot spot and an unsolved problem in current research. But, it is expected to solve the problem of luminescence at long wavelengths and in the liquid phase by choosing suitable hydrophilic matrices and surface modification.

Fourth, the phosphorescence lifetime is also one of the important indicators to measure the performance of CD-based RTP materials. Currently, the phosphorescence lifetime of CD-based RTP materials is mainly concentrated at the second level, and it is only a few seconds to several tens of seconds. Regarding the long-lifetime research, there are still few reports on minute-level, and even hour-level phosphorescence lifetime researches. In addition, CD-based RTP materials with an ultra-long phosphorescence lifetime are expected to show application value in new fields, such as solar cells. In order to realize CD-based RTP materials with ultra-long phosphorescence lifetime, an important matrix selection is the key to solving the problem.

Finally, advanced synthesis methods are one of the key technologies for preparing high-quality CD-based RTP materials. At present, the synthesis route of CD-based RTP materials mainly relies on a two-step method, that is, a matrix-assisted synthesis route is adopted, CDs are synthesized first, and then encapsulated into a matrix to realize RTP. This synthesis method is complicated and inefficient. In addition, the stability of the synthesized CD-based RTP material is also not good. More importantly, in addition to cumbersome synthesis methods, complicated purification processes are also required in the process of realizing CD-based RTP, and the low repetition rate of these synthesis routes greatly limits its large-scale commercial application. Therefore, in order to solve a series of problems in the preparation process, one kind of excellent matrix with a simple and fast preparation method and high stability properties is the only choice.

In summary, CD-based RTP materials have many excellent properties such as low cost, low toxicity and tunable luminescence. It has some advantages unmatched by other RTP materials, and has become a rising star in the fields of anti-counterfeiting, information security, LEDs, sensor detection and so on. However, there are still some problems, such as unclear luminescence mechanism, low PQY, short luminescence wavelength, insufficient phosphorescence lifetime, and simple and convenient preparation methods. Therefore, as a new CD-based RTP material in the future, it will have advantages such as economy, liquid phase stability, long wavelength emission, multi-color emission, high PQY and environmental friendliness. At the same time, CD-based RTP materials can also be combined with other high-performance materials to obtain new composite materials and realize application value in new fields. It is hoped that through the continuous attempts and efforts of the broad masses of people, in modern society where opportunities and challenges coexist, CD-based RTP materials are expected to popularize people's



lives in the future and become the next generation of newly applied luminescent materials.

Author contributions

All authors contributed to writing and revision of the manuscript.

Conflicts of interest

The authors declare no competing interests.

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