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Triplet-to-singlet FRET (TS-FRET) in pure organic phosphors: emerging applications and new opportunities

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Förster resonance energy transfer (FRET) involving spin conversion from the triplet to the singlet state, commonly referred to as triplet-to-singlet FRET (TS-FRET) or phosphorescence energy transfer, has recently emerged as an active area of research in purely organic systems, driven by the development of efficient organic phosphors. This mechanism enables delayed fluorescence with several advantages, including long-lived emission, high quantum yields, large Stokes shifts, and tunable emission profiles, all achieved without the need for complex molecular design strategies. While the growing number of TS-FRET scaffolds has expanded the chemical space of such systems, further progress in this field depends on redirecting the focus toward their practical applications and long-term potential, akin to the well-established singlet-to-singlet FRET systems. With this in mind, this perspective aims to provide a functional and forward-looking interpretation of TS-FRET systems. Alongside a concise overview of their historical development and underlying principles, it highlights key application areas where TS-FRET can make impactful contributions, thereby charting a course for advancing the field beyond current paradigms.

1. Introduction

Triplet-to-singlet Förster resonance energy transfer (TS-FRET), a specialised energy transfer process, involves the nonradiative transfer of energy from an excited triplet donor to a fluorescent acceptor, resulting in the acceptor's excitation to its singlet excited state.[1] The chosen terminology of "TS-FRET" is based on the inductive–resonant mechanism of energy transfer between a triplet and a singlet, as described by Förster,[2] and this term will be employed to illustrate the process throughout the article. The process of TS-FRET is unique because it defies the conventional spin conservation rule in

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typical singlet–singlet FRET (SS-FRET), where the spin angular momentum remains unchanged during energy transfer.[3] The possibility of TS-FRET arises from the fact that the Förster mechanism relies on the coulombic interaction between the transition dipoles of the donor and acceptor, which is inherently independent of spin. However, the efficiency of TS-FRET depends critically on the oscillator strength (f) of the triplet emission in the donor and the spectral overlap between the donor's triplet emission and the acceptor's absorption. A significant oscillator strength for the triplet transition, often facilitated by strong spin–orbit coupling (SOC), is necessary for efficient energy transfer (Fig. 1).

The concept of TS-FRET, albeit initially predicted based on Förster's theory,[2c] remained largely underexplored for several decades. Early studies primarily focused on SS-FRET due to its experimental accessibility and prevalence in various photophysical chromophoric systems. The challenges in experimentally verifying TS-FRET stemmed from the intrinsically low probability of triplet emission from most of the organic molecules at room temperature, owing to competing non-radiative decay pathways. In addition, the understanding of spin angular momentum conservation in energy transfer processes requires further elucidation, as the TS-FRET process inherently involves spin inversion between the initial and final states. However, the potential of TS-FRET in enhancing light extraction from triplet states for various applications prompted further research.[4]

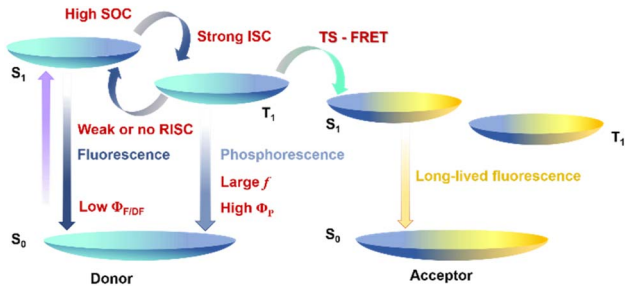

Fig. 1 Perrin–Jablonski diagram and essential criteria for TS-FRET. Efficient TS-FRET requires high SOC between the singlet and triplet of the donor, facilitating strong ISC and significant oscillator strength. Weak RISC and low radiative decay from the singlet prevent the competing pathways. SOC – spin orbit coupling, ISC – intersystem crossing, RISC – reverse intersystem crossing, f – oscillator strength, $\Phi_{F/DF}$ – quantum yield of fluorescence/delayed fluorescence, and Φ_P – quantum yield of phosphorescence.

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The earliest milestone in the understanding of TS-FRET was marked by the work of Ermolaev and Sveshnikova in 1963, which provided the first experimental glimpse of TS-FRET, employing various donors, including *p*-phenylbenzaldehyde, tryptophan, triphenylamine, fluorescein and *N*,*N*-dimethylaniline at cryogenic temperatures (77 K or 90 K) (Fig. 2).[1b] Similar observations at 77 K were made by Bennett and co-workers, with phenanthrene-d_{10} and rhodamine B (RhB) as the donor and acceptor, respectively.[1c] The experiment was conducted at a temperature where the phosphorescence of the donor was significantly enhanced, minimising the influence of competing non-radiative decay pathways from the triplet, enabling the observation of TS-FRET.

The field advanced further with the emergence of organometallic complexes, particularly those of iridium and platinum, as efficient phosphorescent donors.[5] These complexes possess intrinsically high SOC values and large oscillator strengths of the triplet state, resulting in significantly enhanced phosphorescence quantum yields at room temperature. The groundbreaking work by Baldo, Thompson, and Forrest in 2000 demonstrated the use of phosphorescent iridium complexes as donors for TS-FRET in organic light-emitting diodes (OLEDs).[5a] In this study, tris(2-phenylpyridinato)iridium(III) [Ir(ppy)$_3$] acted as the donor, transferring triplet energy to a fluorescent acceptor, resulting in improved device efficiency. This firmly established TS-FRET as a viable pathway for improving the efficiency of OLEDs by harnessing the abundant triplet excitons generated during charge recombination and showcased the practical applications of TS-FRET.

Subsequent studies focused on understanding the mechanistic details of TS-FRET and optimising its efficiency. Wasserberg, Meskers, and Janssen in 2007 explored the competition between Förster and Dexter energy transfer mechanisms in TS-FRET processes involving iridium complexes.[6a] They carefully compared energy transfer to an acceptor that could engage only in Dexter energy transfer *versus* one that also participates in Förster energy transfer. This research provided crucial insights into the role of transition dipole moments and molecular distances in governing the TS-FRET mechanism in both solution and solid-state environments. The experiments demonstrated a shift from diffusion-controlled Dexter energy transfer in solution to a predominantly Förster mechanism in a rigid polymer matrix, highlighting the influence of molecular mobility on energy transfer pathways.

In 2019, Börjesson and co-workers reported the first definitive demonstration of intramolecular TS-FRET, using a rigidly linked organometallic donor–acceptor dyad.[6b] This elegant

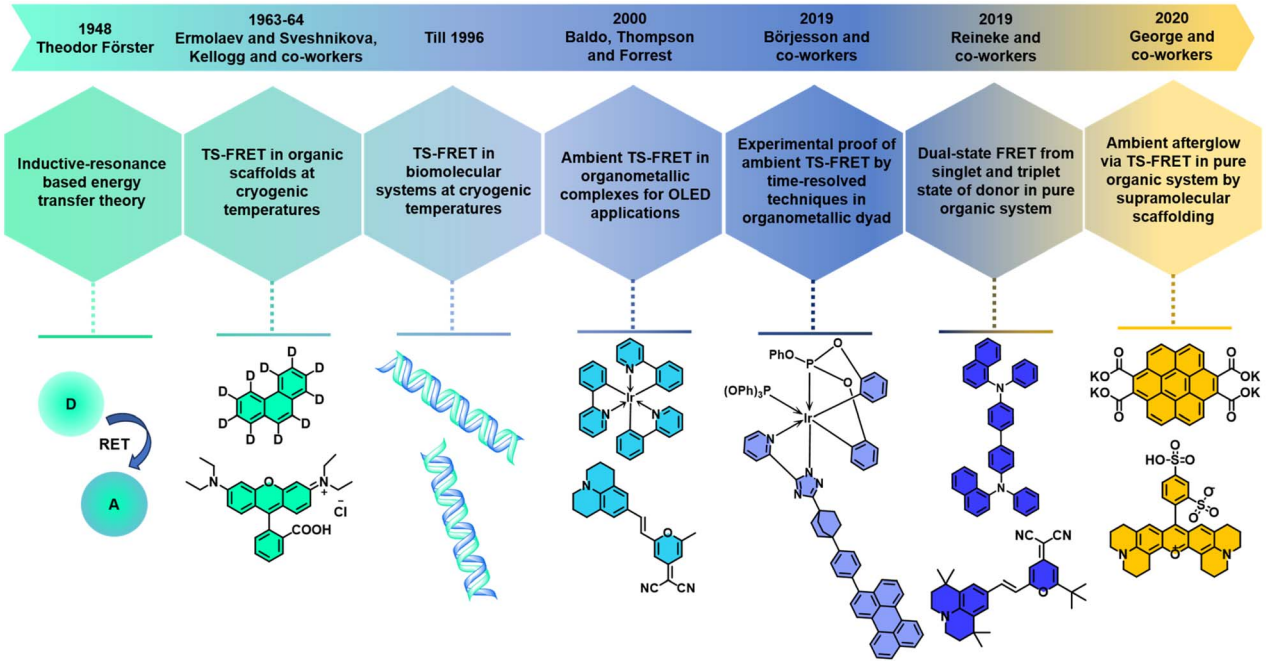

Fig. 2 Historical timeline of the evolution of TS-FRET. RET stands for resonance energy transfer.

study provided conclusive evidence that TS-FRET can occur *via* a dipole–dipole interaction without requiring strict adherence to spin conservation rules. Time-resolved emission spectroscopy was utilised to identify the short and long emission components, confirming the TS-FRET mechanism in the dyad. Moreover, the observed energy transfer rate was remarkably faster than the phosphorescence rate of the isolated donor molecule, reinforcing the potential of TS-FRET for improving the efficiency of light emission from triplet states.

The exploration of TS-FRET was conducted only with either organometallic complexes or at cryogenic temperatures for a long time, without extending to purely organic systems. However, the recent surge in the development of efficient ambient organic phosphors[7,8] and the observation of thermally activated delayed fluorescence (TADF)[9] in organic systems has fuelled the exploration of TS-FRET in organic molecules.[4] Reineke and co-workers in 2019 reported a dual-state FRET system in a purely organic molecular system with simultaneous SS-FRET and TS-FRET exhibiting ambient afterglow.[6c] Nevertheless, the concept of TS-FRET in organic scaffolds hitherto remained a domain of scientific curiosity alone without being driven to applications.

The study by our group in 2020 further extended TS-FRET in purely organic systems to a new regime, developing a method for achieving ambient afterglow fluorescence from a water-processable system.[10] This system utilised coronene tetracarboxylate salt (CS), a long-lived triplet donor developed by our group, and commercially available fluorescent dyes as acceptors, all co-assembled in a polyvinyl alcohol (PVA) matrix. The resulting materials exhibited tunable afterglow colours originating from the singlet states of the acceptors – a phenomenon we termed delayed sensitisation. This highlights the versatile potential of TS-FRET as a universal strategy to obtain afterglow through delayed sensitisation of virtually all fluorescent organic dyes, paving the way for diverse applications. This demonstration marked an explosion in the field of TS-FRET by demonstrating the feasibility of TS-FRET in purely organic, solution-processable materials. Obtaining long-persistent afterglow from commercially available fluorophore dyes using solution processable techniques provided ease of accessibility and steered the direction of TS-FRET towards developing applications.

2. Photophysical insights and analytical approaches in TS-FRET

In order to perceive TS-FRET from an applicative perspective, an understanding of photophysical properties and characterisation of TS-FRET is vital above all. The fundamental photophysical principles governing TS-FRET are rooted in Förster theory, which describes dipole–dipole interactions.[2e] The rate constant (k_{FRET}) for TS-FRET is proportional to the radiative rate constant of the donor (k_r) in its triplet state and the spectral overlap integral ($J(\lambda)$) between the donor's phosphorescence and the acceptor's absorption spectrum. The Förster radius (R_0), which represents the distance at which the energy transfer efficiency is 50%, is crucial in determining the efficiency of TS-FRET.[2d,e] It is determined by the equation:

$$R_0 = \left(\kappa^2 \varphi_r J(\lambda) \frac{9000(\ln 10)}{128\pi^5 N_A n^4}\right)^{\frac{1}{6}} \quad (1)$$

where N_A is Avogadro's number, n is the refractive index of the medium, κ^2 is the orientation factor (2/3 for isotropic systems), and φ_r is the phosphorescence quantum yield of the donor.

The efficiency of TS-FRET (φ_{FRET}) is then given by:

$$\varphi_{FRET} = \frac{{R_0}^6}{{R_0}^6 + {R_{DA}}^6} \quad (2)$$

where R_{DA} is the donor–acceptor distance.

Considering that TS-FRET involves the transfer of excitation energy from the donor triplet to the acceptor singlet, the intensity quenching of the donor phosphorescence with a concomitant increase in the fluorescence intensity of the acceptor is the primary observation for the characterisation of TS-FRET in donor–acceptor composition mixtures, analogous to that for SS-FRET.[1c]

The spectral overlap between the phosphorescence spectrum of the donor and the absorption spectrum of the acceptor serves as a prerequisite energy criterion that significantly influences the efficiency of the energy transfer. Moreover, the decrease in the lifetime of donor phosphorescence with acceptor composition distinguishes FRET from the trivial reabsorption energy transfer process.[2d–f] The majority of reports on TS-FRET rely solely on donor lifetime quenching with varying acceptor compositions to characterise the energy transfer process. However, the dependence of the energy transfer efficiency on the distance between the donor and the acceptor scaffolds is critical in eliminating any contribution from the Dexter energy transfer. As Dexter energy transfer between the donor and acceptor requires physical collisions for orbital overlap, the viscosity of the solution medium tends to have an impact on the energy transfer efficiency and could be utilised to differentiate between the two processes effectively.[2c] The process of TS-FRET pivots on the radiative decay rate of the donor triplet state, and therefore, attenuation of triplet deactivation pathways becomes vital to augment the energy transfer process. Thus, to activate the triplet radiative pathways of the donor, the SOC between the singlet and the triplet has to be large so that inter-system crossing (ISC) and subsequent radiative decay to the ground singlet state can be supported.

The characterisation of the TS-FRET process can be exemplified by a system first reported by our group, where efficient TS-FRET leading to tunable afterglow fluorescence from acceptor molecules was demonstrated.[10] This triplet light-harvesting system comprised a persistent afterglow donor CS, developed in our group, and two commercial fluorescent dyes – sulforhodamine 101 (SR101) and sulforhodamine G (SRG) – selected as acceptors, coassembled within a polymer matrix of PVA (Fig. 3a). The CS–PVA scaffold, stabilised through non-covalent interactions between the carboxylic acid groups of CS and the hydroxyl pendants of PVA, serves as an ideal platform for TS-FRET, owing to the strong oscillator strength of the CS triplet state, as evidenced by its high phosphorescence quantum yield and excellent stability under ambient conditions.[10]

A primary requirement for TS-FRET, similar to conventional FRET from singlet states (SS-FRET), is a significant spectral overlap between the donor emission and the acceptor

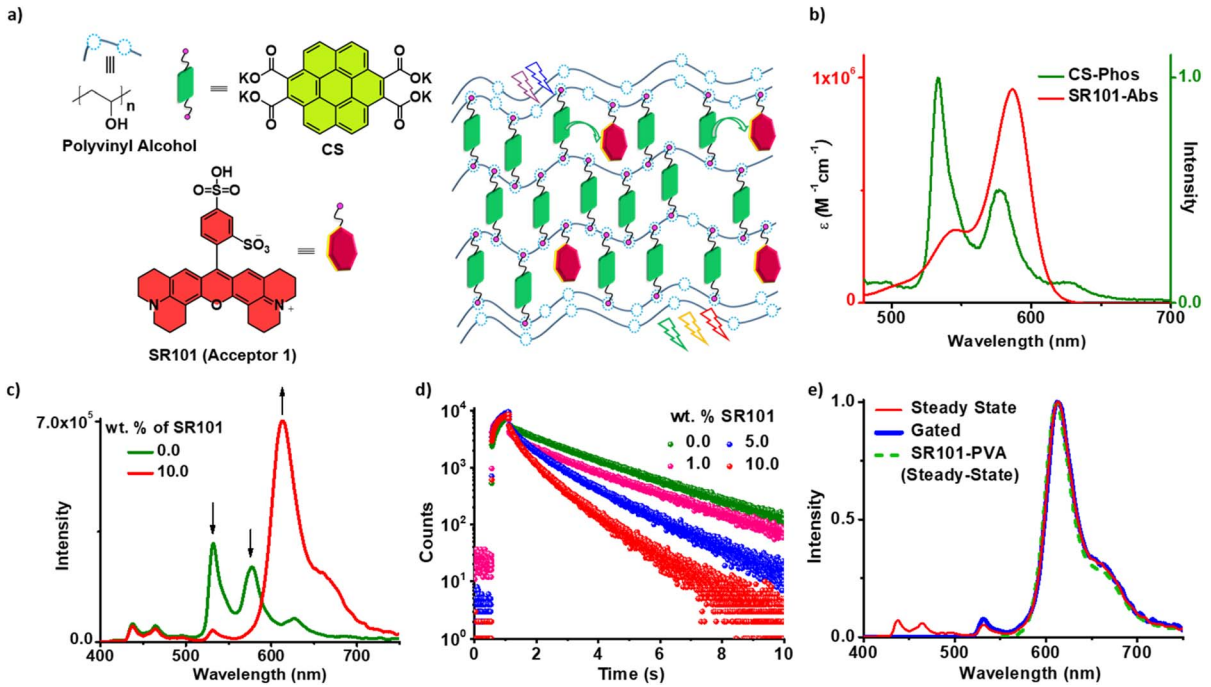

Fig. 3 (a) Molecular structures of the donor and the acceptor with their representative figures and schematic representation of the PVA scaffold with the donor and acceptor anchored *via* hydrogen-bonding and (b) spectral overlap of CS-RTP with the absorption of SR101; Phos: phosphorescence and Abs: absorbance. (c) Steady-state emission spectra ($\lambda_{exc.}$ = 350 nm) of CS–PVA hybrid films in the presence and absence of SR101 and (d) lifetime decay profiles of CS–PVA hybrid films with varying wt% of SR101 ($\lambda_{exc.}$ = 350 nm, $\lambda_{monitored}$ = 532 nm). (e) Normalised steady-state and gated-emission spectra ($\lambda_{exc.}$ = 350 nm and delay time = 5 ms) of 10 wt% SR101 in CS–PVA films along with the steady-state fluorescence spectrum of SR101–PVA film. Reproduced with permission from ref. 10 copyright © 2020 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

absorption, which is efficiently fulfilled in this system (Fig. 3b). Upon selective excitation of the donor (CS–PVA) at 350 nm, a progressive decrease in the phosphorescence intensity of CS–PVA, accompanied by the emergence of a new emission band corresponding to the acceptor (SR101), was observed with increasing donor-to-acceptor ratios (Fig. 3c). This spectral evolution indicates the occurrence of TS-FRET. However, the most critical experiment to conclusively demonstrate that the increase in acceptor emission intensity arises specifically from TS-FRET, and not from SS-FRET or direct excitation of the acceptor, is the gated emission experiment. Gated emission measurements upon excitation of the donor (detection gating time of 5 ms) revealed delayed acceptor emission, indicating sensitisation *via* TS-FRET, since in the absence of triplet sensitisation, the fluorescent acceptor would exhibit only a nanosecond-scale lifetime. Additionally, gated excitation spectra monitored at the acceptor emission wavelength can be used to confirm the origin of the delayed emission as absorption by the donor, providing unambiguous proof of the TS-FRET process and the long-lived nature of the acceptor emission.

Further evidence for TS-FRET is provided by the observed decrease in the donor lifetime along with the appearance of an acceptor lifetime component in the longer timescale regime (milliseconds to seconds) (Fig. 3d). Collectively, these observations provide conclusive evidence for TS-FRET in the CS–PVA–SR101 system and establish a general framework for extending this mechanism to other light-harvesting systems.

This system also establishes the crucial importance of supramolecular interactions in TS-FRET. Confinement effects brought about by non-covalent supramolecular interactions aid in achieving high oscillator strength of the donor by rigidification of the system and reducing oxygen quenching[11] (Fig. 4a). Moreover, these supramolecular interactions assist in bringing the donor and acceptor into close proximity by offering anchor groups, thereby facilitating the energy transfer process. These supramolecular interactions can be brought about by various strategies such as doping in polymer matrices, encapsulation *via* hydrophobic–hydrophilic interactions and micellar formations.[4a] These strategies assist in effectively anchoring the phosphorescent molecules through non-covalent interactions, which suppress the non-radiative decay and increase the oscillator strength of the donor for efficient energy transfer.

3. Towards tailoring TS-FRET for practical applications

The preceding sections have established a comprehensive understanding of the photophysical principles underlying TS-FRET and charted its historical development. This detailed foundation is essential because, while recent reviews have extensively covered design strategies, the diverse range of systems employed, and the intricacies in the photophysical aspects of TS-FRET, a focused perspective on its applications remains conspicuously absent.[4] Existing literature primarily details the "how" of TS-FRET – the synthesis of specific donor–acceptor pairs, the optimisation of photophysical properties, and the mechanistic investigation of energy transfer processes. However, a critical gap exists in detailing the "why" and "where" – the specific application requirements that dictate the design choices and the translation of laboratory-based findings into real-world technologies. This perspective addresses this gap by focusing exclusively on the applications of TS-FRET, thereby providing a much-needed bridge between fundamental research and technological development. By examining the application requirements within diverse technological contexts, we aim to highlight the specific challenges and opportunities associated with each field. This approach allows us to analyse the interplay between the desired functional characteristics of a given application and the design choices for the TS-FRET system. For example, the requirements for a TS-FRET system

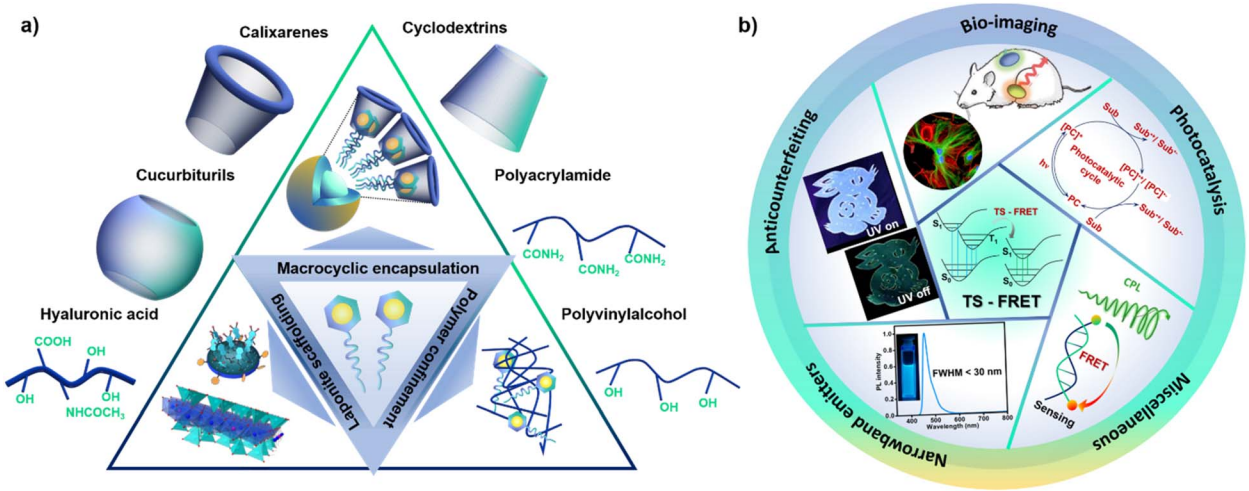

Fig. 4 (a) Various supramolecular strategies employed in TS-FRET systems, stabilising the phosphorescence of the donor. (b) Applications explored in the domain of TS-FRET based organic systems. Part of Fig. 4a reproduced with permission from ref. 12b copyright © 2018 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim and ref. 12c copyright © 2021 Wiley-VCH GmbH. Parts of Fig. 4b reproduced from ref. 22a copyright © 2024 Royal Society of Chemistry and ref. 19b copyright © 2020 Wiley-VCH GmbH and ref. 12a.

in an OLED differ significantly from those in a bioimaging probe or a long-lived luminescent material.

Highlighting these differences allows for a more nuanced and targeted approach, fostering its translation into commercially viable and impactful innovations, beyond its current status as predominantly a phenomenon of scientific curiosity. This application-focused lens not only provides practical insights but also identifies unexplored domains and inspires future research directions (Fig. 4b).

3.1. Afterglow materials based on TS-FRET

One of the most powerful and promising applications of TS-FRET is the realisation of wide-range tunable afterglow, determined by the choice of the acceptor dyes. TS-FRET enables modulation of afterglow emission across a broad chromatic range using purely organic commercial dyes. This concept was first demonstrated by our group, resulting in tunable yellow and red afterglow fluorescence by using the fluorescent dyes, SRG and SR101, respectively, as acceptors (Fig. 5).[10] We envisaged that TS-FRET from an afterglow organic phosphor would impart delayed emission from fluorescent acceptors *via* delayed sensitisation. With this approach, we could obtain afterglow from commercially available fluorophores, which typically emit on the nanosecond timescale. Our pioneering work has sparked significant interest in the scientific community, catalysing deeper exploration of the TS-FRET mechanism and leading to a surge of research articles in this domain. Achieving afterglow from purely organic materials is significantly more challenging than achieving it from their inorganic counterparts; therefore, any demonstration of organic afterglow holds intrinsic merit. While persistent luminescence has been realised using organic room-temperature phosphorescent (RTP) materials,[13] achieving afterglow in organic systems requires specific strategies like exciton traps, which limit the library of molecules that can be utilised.[13e,f] TS-FRET offers a powerful alternative strategy since it enables tunable afterglow fluorescence across a broad spectral range, by the appropriate choice of donors, effectively overcoming the limitations of RTP systems – particularly by achieving high quantum yields in the red and near-infrared (NIR) regions under ambient and even in aqueous conditions.

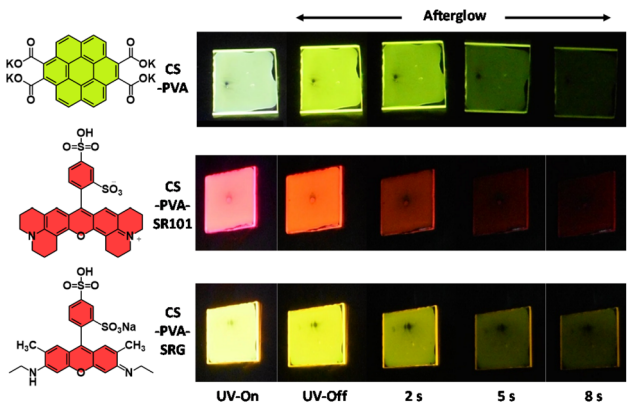

Fig. 5 Images of CS–PVA, CS–SR101–PVA and CS–SRG–PVA hybrid films (with 10 wt% of SR101/SRG) showing afterglow emission over 5 s after turning off the excitation source, a 365 nm UV-lamp. Reproduced with permission from ref. 10 copyright © 2020 Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim.

The energy transfer from a long-lived triplet state of a phosphor to the singlet state of the acceptor enables the generation of persistent, red-shifted delayed fluorescence from the acceptor molecule, effectively extending the emission lifetime and shifting the emission wavelength beyond the limitations of the donor Stokes shift alone. The advantages of utilising TS-FRET in organic afterglow materials are manifold. This approach circumvents the need for complex molecular engineering solely focused on enhancing the inherent RTP properties of the donor molecule. Instead, it enables the use of readily available and highly fluorescent dyes as acceptors, simplifying material design and synthesis and offering a powerful alternative to inorganic phosphors. Furthermore, the ability to tune the emission wavelength by simply selecting different acceptor dyes expands the colour palette of achievable afterglow emissions, offering greater versatility in material applications.

Inorganic afterglow phosphors have long been known for applications in light-emitting diodes, sensing and emergency pavers. However, organic materials, due to their inherently low SOC, do not exhibit high quantum yield phosphorescence with long lifetimes. Afterglow by TS-FRET utilising organometallic complexes is difficult to achieve as the phosphorescence lifetime is extremely short in these heavy metal complexes. Thus, realising TS-FRET in organic materials offers an inherent advantage by offering a universal supramolecular strategy to increase SOC without compromising the lifetime, enabling persistent luminescence.

Afterglow organic materials based on TS-FRET are primarily implemented in biological cell imaging and anticounterfeiting devices. Since the majority of TS-FRET applications involve the afterglow from the acceptor emission achieved *via* energy transfer, we have not elaborated on the applications of afterglow in detail here, as they are covered extensively in the subsequent sections.

3.2. Bio-imaging *via* TS-FRET

The future of live-cell imaging hinges on the development of more effective markers to overcome the existing limitations. Most of the existing fluorescent imaging agents suffer from interference with cellular autofluorescence, reducing contrast and hindering the identification of target regions.[14] This underscores the necessity for biomarkers with prolonged emission, such as phosphorescent biomarkers, which are particularly well-suited for cell imaging as they effectively eliminate background fluorescence from the cell.[15] However, this presents a significant bottleneck, as triplet-state emissions are quenched through collisions with water molecules and dissolved oxygen in the cellular environment.[15]

Consequently, channelling the triplet state energy into another state, non-radiatively, which can sustain the delayed emission can be considered vital for developing superior imaging agents that fully utilise triplet state energy. It is in this context, TS-FRET can act as a promising avenue that can

address the aforementioned challenges by transferring the energy from the triplet of the donor to the acceptor.[10]

Moreover, most of the currently reported phosphorescent molecules emit in the UV or visible region, which limits their imaging depth. In contrast, TS-FRET offers the added advantage of tunable emission at longer wavelengths, due to large Stokes shifts, allowing deeper tissue penetration with less invasive signals while simultaneously mitigating the adverse effects of the energy-gap law.

In order to design a suitable system for achieving efficient TS-FRET emission, one of the crucial deciding factors is the radiation yield from T_1 (φ_r^T), which is in turn decided by k_q, the quenching rate of the triplet of the donor due to its interaction with the host matrix, given by:

$$\varphi_r^T = \frac{k_r^T}{k_r^T + k_{nr}^T + k_q} \quad (3)$$

where k_r^T and k_{nr}^T are the radiative and non-radiative rate constants from the triplet state of the donor.[7a]

Therefore, selecting an appropriate host material to disperse the guest or donor (phosphor) is crucial for minimising triplet quenching. Taking this into account, most reported systems demonstrating TS-FRET, particularly those proposed for potential biomarker applications, employ a secondary confinement design strategy to restrict non-radiative decay pathways effectively. Non-covalent macrocyclic hosts, such as cucurbit[*n*]urils (CB), modified α- and β-cyclodextrins (CD), and sulfonatocalixarenes, have proven effective in this regard.[17] These hosts not only rigidify the system but also provide amphiphilic, multicharged, pre-organised structures to create a hydrophobic environment that facilitates the loading of organic dyes, thereby bringing the donor and acceptor into close proximity for efficient energy transfer.[16]

The initial attempt to develop a supramolecular assembly exhibiting TS-FRET in the solution state was reported by Liu's group, where it was also successfully applied for mitochondrial imaging.[17a] In this system, a 4-(4-bromophenyl)-pyridine-modified β-cyclodextrin (CD-PY) encapsulated in CB[8] functioned as the phosphorescent donor, transferring energy to RhB *via* TS-FRET (Fig. 6). Furthermore, the secondary confinement by adamantane-modified hyaluronic acid (HA-ADA) led to the formation of nanoparticles in solution, resulting in enhanced delayed fluorescence emission from the acceptor. Subsequently, mitochondrial imaging was performed with the nanoparticles inside A549 cancer cells.[17a]

Similarly, a dodecyl-chain-bridged 6-bromoisoquinoline derivative encapsulated within CB[7] and β-cyclodextrin-modified hyaluronic acid (HA-CD) exhibited an 8- and 10-fold increase in triplet state emission and lifetime, respectively *via* the secondary confinement, and the subsequent TS-FRET to nile blue or tetrakis(4-sulfophenyl)porphyrin resulted in delayed emission at 680 nm and 710 nm, respectively. This red-shifted emission from the supramolecular light-harvesting system enabled visualisation of living tumour cells *via* hyaluronic acid receptor-mediated endocytosis.[17b]

Further advancements, particularly in achieving more pronounced red-shifted NIR emission *via* TS-FRET, have been realised through cascade energy transfer. This process involves the use of an intermediate dye positioned between the phosphor and the final acceptor (fluorescent dye), facilitating successive energy transfer steps. The pioneering work in this aspect was reported in 2021 by Liu and co-workers, where a dibromophthalimide phosphor was confined inside CB[7] and sulfonatocalix[4]arene.[17c] The triplet state energy of dibromophthalimide was transferred initially to organic dyes like RhB or dibenzothiadiazole by TS-FRET and subsequently to nile red or cyanine 5 (Cy5) *via* SS-FRET.[17c] This process achieved a large Stokes shift of 365 nm with a long lifetime, making it suitable as an imaging agent for multicolour cell labelling. Various research groups have meticulously exploited this stepwise energy transfer, achieving remarkable results, including an ultra-large Stokes shift of 525 nm at 825 nm, which could result

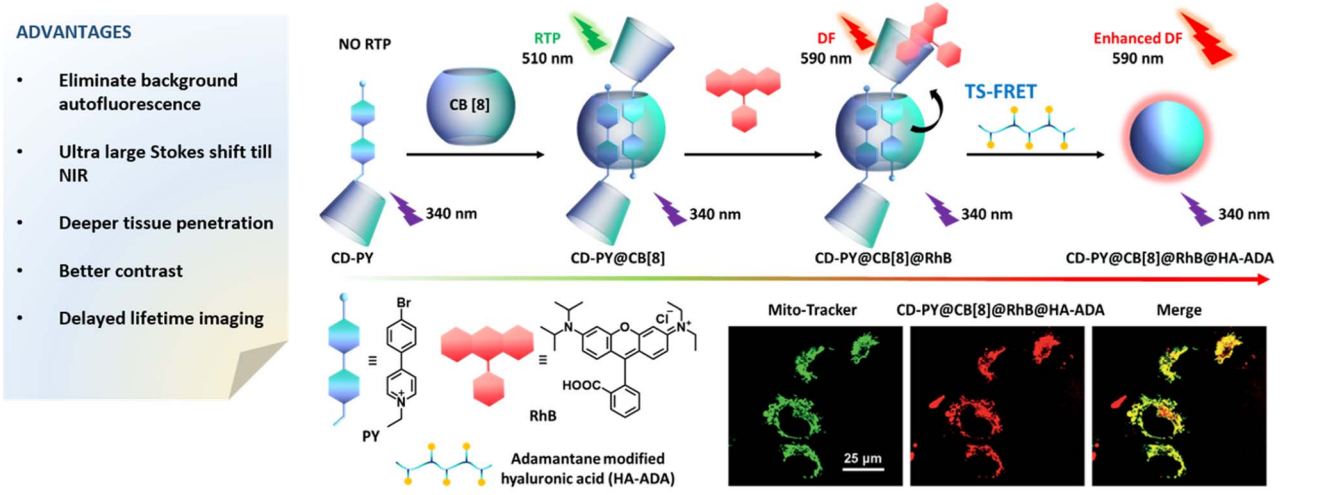

Fig. 6 TS-FRET systems for imaging applications. Molecular structures and schematic representation of CB[8]-confined 4-(4-bromophenyl)-pyridine-modified β-cyclodextrin (CD-PY) assembled with HA-ADA, forming phosphorescent nanoparticles in solution, with their application as mitochondrial imaging agents. Part of Fig. 6 reproduced from ref. 17a copyright © 2021 Royal Society of Chemistry.

in better image contrast during labelling.[17d] Additionally, these advancements demonstrate a significant capacity to penetrate skin tissue over 2 mm thick, as shown by Chi and co-workers, indicating promising future applications in terms of imaging across a range of practical fields (Fig. 7a).[17e] It is noteworthy to mention the role of the intermediate TS-FRET process in the cascaded energy-transfer in achieving delayed luminescence to the final acceptor – an important outcome that would otherwise be unattainable. TS-FRET thus presents a powerful strategy for achieving tunable delayed emission without complex chemical modifications, simply by selecting the appropriate dyes as the acceptors. A significant recent advancement is the transition

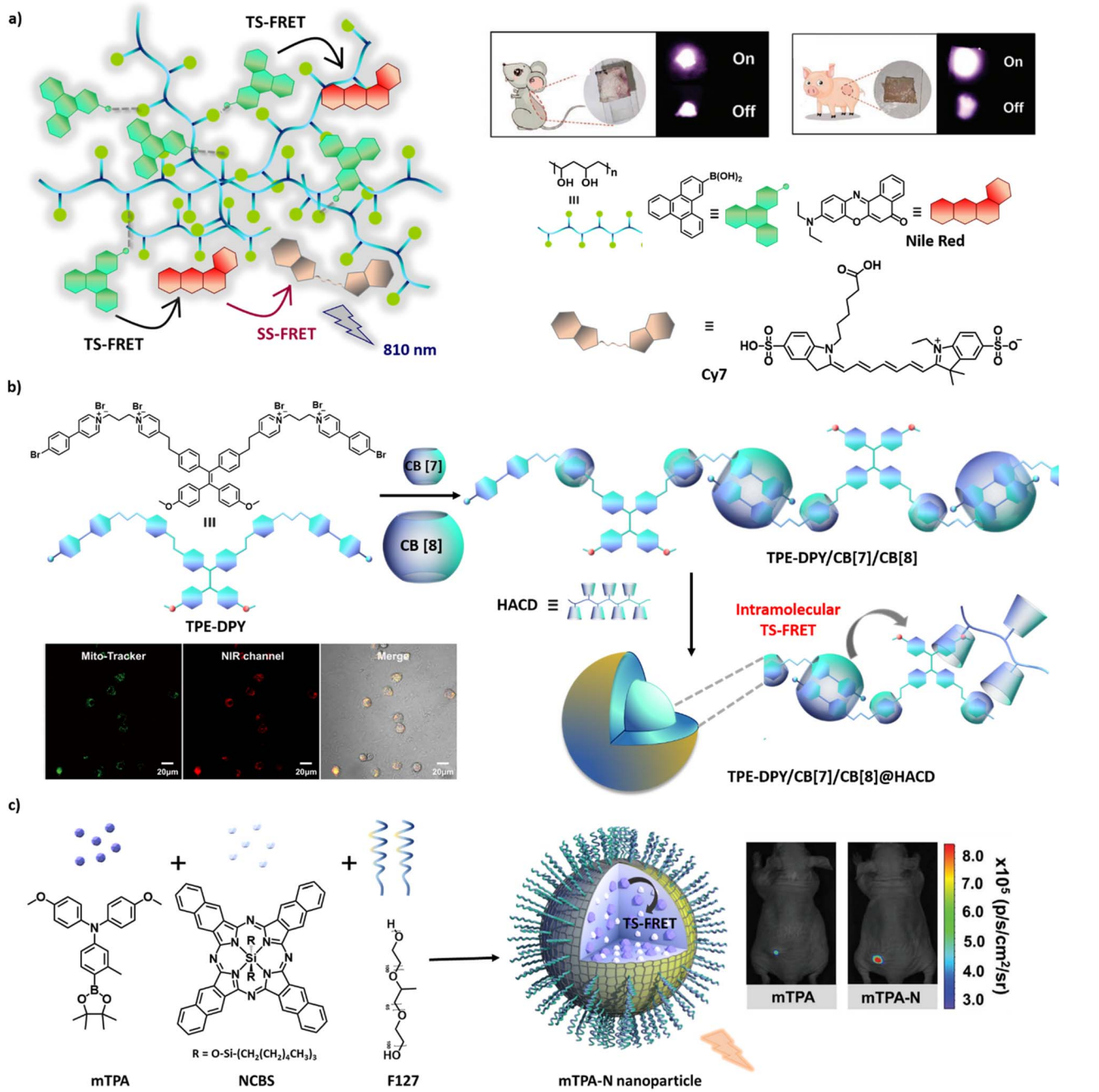

Fig. 7 TS-FRET systems for afterglow imaging. (a) Molecular structures of triphenylene-2-ylboronic acid, nile red, and Cy7, along with a schematic representation of triphenylene-2-ylboronic acid anchored in the presence of PVA. Photographs captured behind a piece of mouse skin and a piece of pork skin demonstrate the system's afterglow imaging and penetrating capability over 2 mm. (b) Molecular structure of TPE–DPY and the schematic illustration of the single-molecule TS-FRET process in assembly, and respective images of mitochondria taken. (c) Schematic showing the mTPA-N nanoparticles formed from mTPA and NCBS, and *in vivo* afterglow imaging of living mice after subcutaneous injection (50 μL) of mTPA and mTPA-N (600 μg mL^{-1}). Afterglow images were acquired after UV irradiation of mice for 5 s. Parts of Fig. 7 reproduced with permission from (a) ref. 17e copyright © 2022 Wiley-VCH GmbH, (b) reproduced from ref. 17f, available under a CC-BY 4.0 license, and reproduced with permission from (c) ref. 19b copyright © 2020 Wiley-VCH GmbH.

from bicomponent systems to a single-component, purely organic intramolecular TS-FRET, achieving an impressive ultra-large Stokes shift of 367 nm, as demonstrated by Liu and co-workers (Fig. 7b).[17f] Interestingly, the topological morphologies of the phosphor, namely, an alkyl-bridged methoxy-tetraphenylethylenebromophenylpyridines derivative, transformed from nanospheres to nanoplates depending on the ratio between CB[7] to CB[8], accompanied by changes in photophysical properties. The subsequent addition of HA-CD created a secondary assembly that further refined the structure, affecting the conformation and facilitating TS-FRET from the bromophenyl-pyridine to the tetraphenylethylene moiety, demonstrating intramolecular TS-FRET. This approach paves the way for future molecular designs based on single-molecule TS-FRET, potentially reducing the complexity of multi-component systems for imaging applications.

The recent surge in time-resolved imaging, driven by its advantage of concentration-independent probing, establishes long-lived emissive systems as a valuable tool for advanced imaging applications. Importantly, longer-lived emissions obviate the need for specialised requirements such as image intensifiers, pulse generators, and ultrafast shutter cameras, thereby rendering afterglow imaging a highly accessible and practically useful time-resolved technique.[18] TS-FRET can be easily identified as one of the most efficient approaches to achieve afterglow from a highly red-shifted state, avoiding the the detrimental effects of the energy gap law. In this context, Hirata and co-workers could selectively achieve TS-FRET alone in the excited state by particularly choosing a near-zero fluorescent phosphor with an afterglow of 5 s and a red-emissive acceptor, which could be used as a red-emissive afterglow imaging agent.[19a] One step ahead, Li and co-workers have developed NIR afterglow imaging agents by integrating the advantages of phosphorescence (ultra-long-lived emission) and NIR emission.[19b] This approach enabled the redshift of phosphorescence emission of the organic phosphorescent molecule, *N*,*N*-bis(4-methoxyphenyl)-3-methyl-4-(4,4,5,5-tetramethyl-1,3,2-dioxaborolan-2-yl)aniline (mTPA) (530 nm) to the NIR region (780 nm) by using the acceptor NCBS, harnessing the benefits of RTP (low background noise) and NIR emission (deep tissue penetration). This system was finally used for the imaging of lymph nodes in living mice with a high-signal to-noise ratio (Fig. 7c). All these exemplars clearly indicate that, without TS-FRET, the afterglow of the phosphors would remain confined in the visible range, negating the benefits of enhanced penetration depth. This combination is crucial for achieving high-sensitivity, deep-tissue afterglow imaging.

Therefore, TS-FRET offers a versatile platform for manipulating complexes to suit specific imaging needs, bridging the gap between fundamental research on long-lived emissive molecules and diverse practical applications.

3.3. Anticounterfeiting applications

Anticounterfeiting materials are essential for protecting industries from the economic and public health threats posed by counterfeit goods.[20] Several anti-counterfeiting technologies have been developed to combat counterfeiting and piracy, including methods based on digital watermarks, laser holograms, barcodes, radio-frequency identification, and luminescence.[21] Luminescence in organic materials is gaining significant attention in this domain due to their unique optical features and scalability over traditional strategies such as watermarks and barcodes, which can be duplicated easily and hence are ineffective. However, traditional luminophores emit static fluorescence, which relies only on the emission colour for storing information. In order to develop efficient anti-counterfeiting devices, a multilevel approach is required with more luminescence parameters that can add complexity to the encryption.[21b] In this context, organic afterglow materials for anticounterfeiting offer spatially and temporally resolved tunable optical characteristics and ease of manufacture, providing a versatile platform for multilevel optical encoding while being cost-effective and scalable.

A tetracarboxylic modified acyclic cucurbituril (ACB-COOH), embedded in a PVA matrix (ACB-COOH@PVA) manifests ultralong phosphorescence emission at 510 nm (Fig. 8a). In the presence of RhB and pyronine Y (PyY) as acceptors, TS-FRET from the donor to the acceptor led to the emergence of an emission peak at 580 nm, with energy transfer efficiencies of 98.4% and 87.4% respectively. This system was implemented for potential optical encoding by creating patterns with RhB⊂ACB-COOH@PVA, where a red afterglow was observed after turning off the 254 nm excitation source.[22a] By using the ink on specific parts of a pattern with pristine RhB on the other parts, a ciphered image could be imprinted, which is decipherable only with the afterglow that follows UV light irradiation, taking advantage of the large Stokes shift provided by TS-FRET.

Anticounterfeiting techniques leverage afterglow materials by employing strategies, *viz.*, orthogonal, dynamic, multichrome, and multimode luminescence.[21b] Orthogonal luminescence occurs when the emission is distinctive with multiple excitation wavelengths due to non-interfering independent chromophores within the system. Dynamic luminescent systems have luminescent intensity or colour that changes reversibly over time. Dual-emissive materials and stimuli-responsive afterglow fall under multichrome and multimode strategies, respectively. TS-FRET allows tuning of the photophysical properties through the choice of acceptors, enabling these various approaches for anticounterfeiting. In addition to emission modification by varying the acceptors, colour tunability can be realised by simply varying the doping content of the acceptors.[22b]

The ultra-long phosphorescence emission from PVA confined 9,10-diaminophenanthrene was used sensitise the acceptor RhB. With increasing doping content of RhB, the emission was gradually red-shifted from yellowish to pure red. Herein, the aggregation effect of the dye leading to dual emission is the key factor responsible for the colour tunability in this system, materialising multichrome luminescence (Fig. 8b).

Moreover, owing to the different lifetimes of the monomer and aggregation peaks, the system shows a variation in colour during and after the irradiation of light. The dynamic

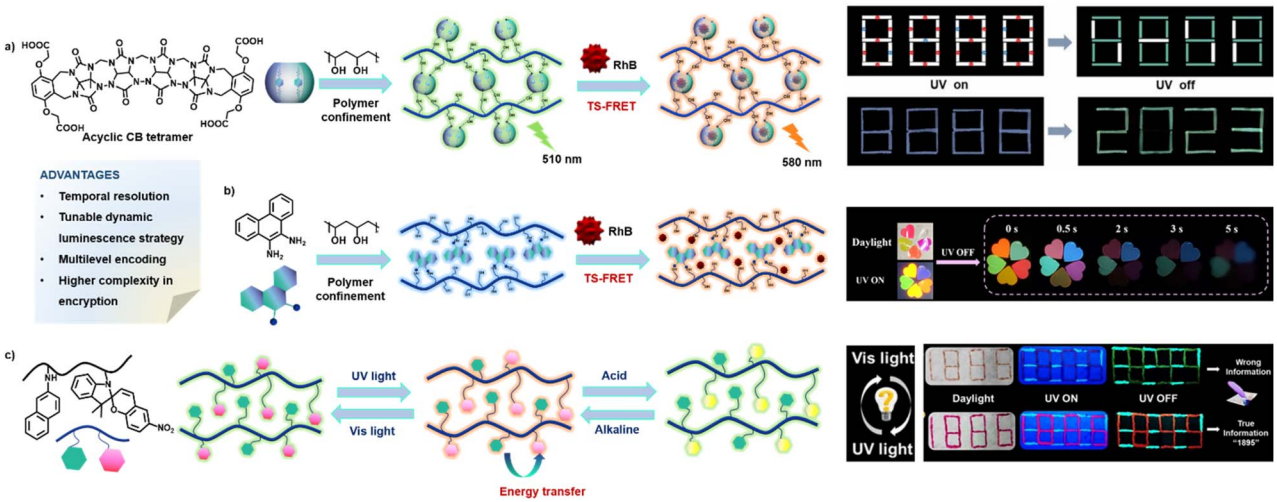

Fig. 8 TS-FRET systems for anticounterfeiting applications. (a) Polymer-confined ACB-COOH acting as a donor for RhB as a potential system for anticounterfeiting. (b) Multichrome luminescence arising from the aggregation effects in the diaminophenanthrene-based TS-FRET system. (c) Multi-stimuli responsive anticounterfeiting system constructed from a co-polymerised TS-FRET system. Part of Fig. 8 reproduced from (a) ref. 22a copyright © 2024 Royal Society of Chemistry, reproduced with permission (b) ref. 22b copyright © 2024 Elsevier and (c) ref. 22c copyright © 2023 Wiley-VCH GmbH.

luminescence enhances the multifaceted nature of the anti-counterfeiting system, adding dimensions to multilevel encoding. The multichrome luminescence strategy by varying the doping content can be extended to other acceptor dyes such as cyanine, rhodamine 6G and fluorescein.

TS-FRET also offers multiple avenues for augmenting the complexity of the encryption. In a molecularly copolymerised system of a naphthylamine and spiropyran (SP) derivative in acrylamide and methylmethacrylate, the TS-FRET process occurs from the naphthylamine derivative to the spiropyran moiety, leading to colour-tunable afterglow emission.[22c] In addition to colour tunability by the D : A ratio in the polymer, the intriguing structural changes of SP enable regulatable FRET channels between SP and merocyanine (MC) forms under continuous UV irradiation, tuning the emission colour from green to red. The process is reversible as the SP form is regenerated by continual visible light irradiation. Subjecting the MC system to an acidic environment results in similar configurational switching to the MCH^+ state, which is again reversible upon exposure to an alkaline environment (Fig. 8c). Anticounterfeiting designs based on these multi-stimuli responsive polymeric systems were developed for programmable encryption and anti-forgery purposes.[22]

A noteworthy feature of TS-FRET in anticounterfeiting applications is the numerous channels it offers for optimising and tuning the optical properties. The multicomponent nature of TS-FRET-based functional designs increases the number of factors that can be controlled, facilitating the exploration of various systems by employing the variety of fluorescent dyes available in the literature. This allows for incorporating elements of tunability, deepening the complexity of the encryption modes.

3.4. Photocatalytic applications of TS-FRET

Photocatalysts offer an inherent advantage over conventional synthetic methodologies due to their ability to facilitate reactions that are thermally inaccessible.[23a] A photocatalyst in its excited state undergoes either photoinduced energy transfer (PEnT) (type-II photosensitiser) or photoinduced electron transfer (PET) (type-I photosensitiser) with the substrate, subsequently leading to the formation of products. To maximise the utilisation of captured energy and direct the absorbed energy toward the desired reaction, multicomponent artificial light-harvesting systems employing excited state energy transfer prove to be advantageous over molecular photocatalysts. For example, a light-harvesting system, based on a supramolecular assembly between pillar[5]arene and a tetraphenylethylene-functionalized dialkylammonium derivative as the donor with eosin Y (EY) and nile red as acceptors, demonstrates significantly enhanced photocatalytic efficiency for the dehalogenation of α-haloacetophenone. The reaction performed in the presence of the light-harvesting system as the photocatalyst led to a product yield of 96%, compared to the 31% yield obtained in the presence of nile red and EY alone.[24a]

However, it is crucial to note that in addition to a large absorption cross-section and a high photoluminescence quantum yield, the excited states of these photocatalysts must be sufficiently long-lived to enable the required diffusion processes required for forming the catalyst–substrate complex.[23b] Short-lived singlet excited states rapidly undergo radiative and non-radiative decay, necessitating the involvement of triplet states to increase the excited state lifetime. In this context, TS-FRET serves as a promising phenomenon for achieving longer excited lifetimes alongside the maximisation of utilising absorbed energy for photocatalytic purposes.

The longer lifetime of the phosphorescence increases the probability of energy transfer from the donor to the acceptor, leading to better energy transfer efficiency and increased catalytic activity in these phosphorescent light-harvesting systems. For example, a supramolecular double-network confined hydrogel based on a CB-encapsulated bicationic vinylbromophenylpyridine derivative, copolymerised with acrylamide and co-assembled with hyaluronic acid modified hydroxypropyl-β-cyclodextrin, exhibits phosphorescence at 510 nm with a lifetime of ∼5 ms. The emission is employed to photosensitise EY, leading to the emergence of delayed emission at 560 nm by TS-FRET (Fig. 9). This light harvesting system, when exploited for the catalysis of the cross-coupling reaction between benzothiazole and diphenylphosphine oxide with a xenon lamp as the light source, shows an improved reaction yield of 62% in comparison with the 32% yield achieved by the EY photocatalyst.[24b] This is attributed to the formation of more active EY species with a long lifetime due to the cumulative effect on EY by direct excitation and photosensitisation through phosphorescence, exemplifying the role of TS-FRET in photocatalytic applications.

Furthermore, phosphorescent light harvesting systems implementing cascade energy transfer from triplet-to-singlet-to-singlet can be used to catalyse multiple reactions of different activation barriers within the same system. This strategy was applied to develop a system with a supramolecular polymer BDBP-CB[8], combining CB[8] and a 4-(4-bromophenyl)-pyridine derivative (BDBP) acting as a donor and SR101 and Cy5 serve as energy acceptors. The phosphorescence of the polymer at 525 nm is channelled to undergo TS-FRET to SR101 ($\lambda_{max} = 625$ nm), followed by subsequent SS-FRET to Cy5 ($\lambda_{max} = 680$ nm). The supramolecular polymer BDBP-CB[8] acts as a type-II photosensitiser, generating singlet oxygen ($^1O_2$) for the photooxidation of styrene derivatives under aqueous conditions. On the other hand, the phosphorescent light harvesting system (BDBP-CB[8] + SR101 + Cy5) serves as a type-I photosensitiser, generating a superoxide radical ($O_2^{\bullet -}$) for cross-dehydrogenative coupling (CDC) of *N*-phenyltetrahydroisoquinoline and indole derivatives.[24c] The reaction catalysed by the TS-FRET based light harvesting system, BDBP-CB[8] + SR101 + Cy5, achieved a three times higher yield (90%) than the reaction yields obtained in the presence of SR101, Cy5 or both SR101 and Cy5 (>30%). The transformation from a type-II to type-I photosensitiser and the various derivatives of the reaction substrates that can be catalysed demonstrate the versatility of TS-FRET in photocatalytic organic conversions.

In these systems, supramolecular assembly and polymer confinement for TS-FRET, along with the prevention of oxygen quenching of triplets and orientation of donor–acceptor for energy transfer, could offer a platform for anchoring the substrates through molecular engineering. This promotes diffusion rates for the formation of substrate–catalyst encounter complexes, thereby improving the overall product yield. Moreover, additional non-covalent interactions between the donor and acceptor can be incorporated by exploring various molecular designs for effective orientation to enhance energy transfer efficiency. The confined microenvironment assists in reducing the photobleaching effects of continual light irradiation.

Despite the various advantages offered by TS-FRET for photocatalytic applications, the domain remains largely unexplored and warrants greater attention for research.

3.5. Narrowband emission employing TS-FRET

Achieving emitters with narrow band emission and high colour purity is of paramount importance in the realm of high-end display electronics and controllable bio-optical imaging. To date, narrow-band emitters based on quantum dots, perovskite materials, and organic chromophores have been reported, with

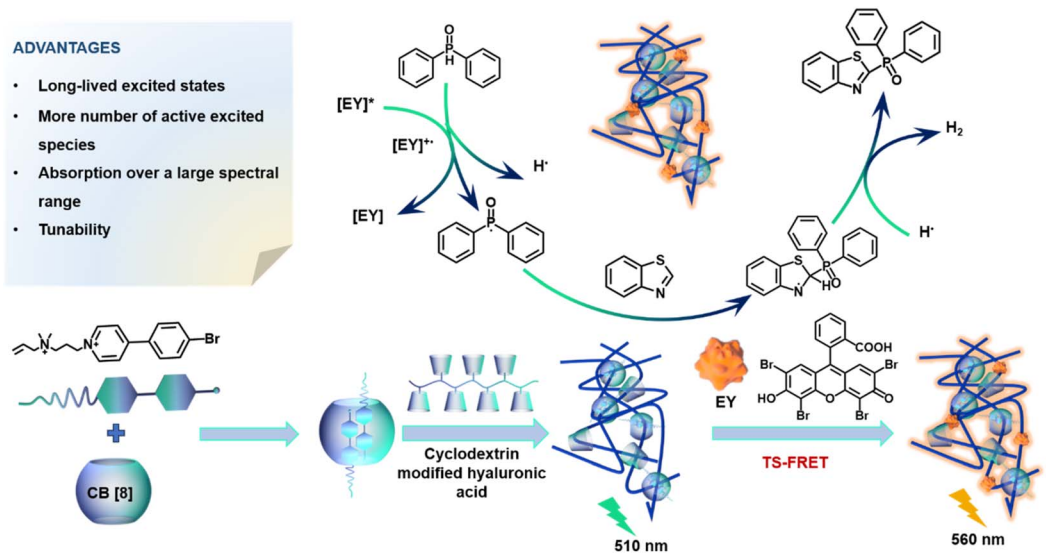

Fig. 9 Photocatalytic applications of TS-FRET systems. CB and CD encapsulated donors for delayed sensitisation of EY dye, enabling the photocatalytic process of cross-coupling between benzothiazole and diphenylphosphine.

the first two exhibiting higher efficiencies.[25] Owing to their superior biocompatibility, ease of structural modification, cost-effectiveness, and suitability for large-area fabrication, organic chromophores have emerged as a preferable alternative to toxic heavy-metal-based luminophores.[26] However, π-conjugated organic chromophores exhibit limited colour purity and pronounced peak broadening in their emission, primarily due to strong intrinsic vibronic coupling between the ground and excited states and structural relaxation occurring in the excited state.[26] Although optical filters and microcavities have been employed to achieve narrow emission, this often comes at the expense of the quantum efficiency of the system. Recently, twisted rigid structures with fused aromatic backbones, which exhibit a lower tendency to form charge transfer (CT) states and aggregates, have demonstrated emissions with very narrow full-width at half-maximum (FWHM), but their synthesis requires complex and tedious strategies. Hence, both phosphorescent and thermally activated delayed fluorescent materials displayed very poor colour purity.[26] To this end, channelling triplet excitons *via* TS-FRET to an acceptor with a narrow FWHM can enable narrow-band emission without concerns of peak broadening.

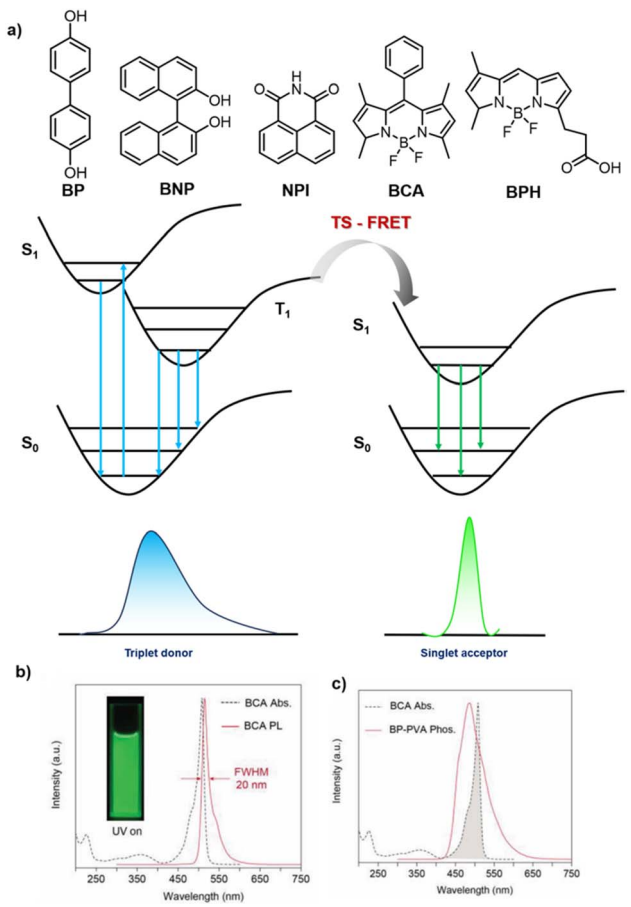

Fig. 10 Narrow-band emitters *via* TS-FRET. (a) Molecular structures of donors (BP, BNP, and NPI) and acceptors (BCA and BPH) and a simplified Jablonski diagram showing the TS-FRET process to produce narrow-band emission. (b) Emission and absorption spectra of acceptor BCA. (c) Spectral overlap of emission of BP (donor) embedded in PVA, and absorption of BCA (acceptor). Adapted with permission from ref. 27a copyright © 2023 Wiley-VCH GmbH.

In this context, tunable narrow-band afterglow can be achieved through TS-FRET by selecting a donor that exhibits afterglow and pairing it with a narrow-band emissive acceptor (BODIPY-based fluorescent dyes) doped in PVA, as demonstrated by An, Huang and collaborators (Fig. 10).[27a] As demonstrated here, since most TS-FRET systems utilise a host–guest strategy, molecular motion can be restricted through rigidification, and the donor and acceptor are spatially separated, thereby preventing the formation of charge-transfer states. The same group achieved tunable narrowband emission by employing multiresonant TADF (MR-TADF) molecules with diverse emission maxima as acceptors, utilising triplet–singlet Förster resonance energy transfer (TS-FRET) from a phosphorescent donor.[27b]

Another strategy, known as hyperfluorescence, first reported by Adachi and co-workers, involves combining a fluorescent emitter dopant with a TADF-assistant dopant to achieve narrowband emission with a delayed lifetime and enhanced external quantum efficiency.[28] In this approach, an SS-FRET occurs from the TADF-assistant dopant to the fluorescent emitter following reverse intersystem crossing (RISC) in the TADF-assistant dopant, resulting in delayed luminescence with high spectral purity.[28] Although TS-FRET does not directly occur in these types of molecules, the triplet state of the TADF-assistant dopant plays a crucial role in facilitating long-lived narrowband emission from the singlet state of fluorescent emitting dopant following the SS-FRET. Thus, by leveraging the properties of both triplet and singlet states, hyperfluorescence emerges as a pivotal photophysical mechanism for achieving narrowband delayed luminescent emitters. There are also systems in which excitons are transferred to the fluorescent emitter both by SS-FRET and TS-FRET simultaneously to enhance the internal quantum efficiency.[28]

3.6. Miscellaneous applications

Circularly Polarised Luminescent (CPL) systems are another class of materials that are gaining widespread attention owing to their spatial resolution and optical sensitivity, finding their way towards applications such as chiroptical devices, 3D displays, sensing and storage of information.[29] However, combining chirality and triplet state characteristics such as afterglow through Circularly Polarised Phosphorescence (CPP) has been relatively less explored.[30] Moreover, tuning the emission colour and lifetime in CPP materials is a formidable challenge. TS-FRET offers a facile route to this limitation without additional synthetic challenges.[31] For instance, Yuan and co-workers designed a TS-FRET based circularly polarised afterglow system with naphthylphosphoric acid derivatives in PVA as phosphorescent and chirality transfer donors, and commercial RhB as an energy acceptor. Tunable CPL afterglow with an enhanced dissymmetry factor was obtained in this system by varying the RhB composition due to energy and chirality transfer between the CPP donor and fluorescent acceptor.[31a] Although several reports have stated an increase in

the dissymmetry factor due to SS- and TS-FRET,[32] the mechanism behind the enhancement remains to be elucidated. Determining whether the energy and chirality transfer processes are concerted and distinguishing their occurrence in the ground and the excited states is a concept under discussion, which could potentially pave the way for future research on CPL/CPP energy transfer systems.[33]

In the context of triplet-based organic materials, an application which cannot be disregarded is OLED devices. Despite the pioneering electroluminescent TS-FRET system with an organometallic complex being reported as early as 2000,[5a] the research on organic TS-FRET systems exhibiting electroluminescence remains scarce. The persistent luminescence of pure organic TS-FRET systems, while advantageous for other applications, is not particularly appealing in the case of electroluminescence, due to interference from secondary phenomena and charge annihilation.[34] Moreover, contemporary studies on TS-FRET in purely organic scaffolds largely depend on supramolecular scaffolding and multicomponent systems to increase the oscillator strength of the donor triplets, which prompts perceptions of its disadvantages for potential implementation in the fabrication of devices. Focussed research on overcoming challenges related to reducing exciton lifetimes, and thereby increasing the device stability, commands greater attention for expanding the scope of TS-FRET towards optoelectronic applications.

4. Conclusions and future outlook

TS-FRET has emerged as a powerful tool for manipulating excited-state energy, offering unique advantages across diverse applications. Its ability to harness long-lived triplet states, often inaccessible *via* traditional fluorescence, coupled with the high emission properties of fluorescent acceptors, makes it particularly well-suited for generating narrow-band emitters, enhancing bioimaging capabilities, and improving photocatalytic efficiencies. The non-radiative nature of TS-FRET, circumventing the limitations of direct triplet emission, enables faster decay pathways and overcomes the constraints of the energy-gap law, enabling red-shifted and NIR emissions crucial for deep-tissue imaging. This is evident from the numerous examples detailed, showcasing the breadth of applications from information encryption and deep-tissue bioimaging to photocatalysis.

However, realising the full potential of TS-FRET requires addressing several key challenges. The majority of contemporary research articles on TS-FRET depend solely on the spectral overlap between donor emission and acceptor absorption for developing molecular designs. Factors such as the orientation and distance between the donor and acceptor need to be focused, along with spectral overlap, for future designs to realise higher FRET efficiency and antenna effects. Moreover, the employment of TS-FRET in domains such as CPL materials is still in its early stages and needs further exploration.

Besides, current strategies often rely on supramolecular assemblies and polymer confinement, which, while enhancing triplet lifetimes, can introduce complexities in synthesis and reduce the stability of the resulting systems, especially within the demanding domains of optoelectronic devices. Furthermore, the potential competition from triplet–triplet energy transfer (TTET) mechanisms remains a significant hurdle to achieving device efficiency in optoelectronics.

Prioritising the development of robust, small-molecule TS-FRET systems that exhibit high stability and extended operational lifetimes is pivotal. This requires a deeper understanding of the interplay between molecular structure, host matrix, and energy transfer dynamics. Computational methods combined with detailed experimental characterisation could provide valuable insights for optimising donor and acceptor properties, minimising TTET, and achieving high TS-FRET efficiencies in solution-processable materials suitable for device fabrication. Ultimately, overcoming these challenges will pave the way for the widespread adoption of TS-FRET in next-generation technologies, from high-performance OLEDs to advanced bioimaging probes capable of offering enhanced penetration depth and spectral resolution.

Author contributions

The manuscript was written by S. N. and A. A. K. under the supervision of S. J. G. S. N. and A. A. K. have contributed equally to the organisation, figure preparation and writing of the manuscript.

Conflicts of interest

There are no conflicts to declare.

Data availability

It is a perspective, and hence, data availability is not applicable for this submission.

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