Chemiluminescence

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Chemiluminescence Resonance Energy Transfer Efficiency and Donor–Acceptor Distance: from Qualitative to Quantitative

Jinhui Lou+, Xiaofang Tang+, Haoke Zhang, Weijiang Guan, and Chao Lu**

Abstract: *Since its birth in 1967, the utilization of chemiluminescence resonance energy transfer (CRET) has made substantial progress in a variety of fields for its unique features. However, the quantitative relationship between CRET efficiency and donor–acceptor distance has not yet been determined owing to the difficulty in designing the variable lengths between chemiluminescent donors and acceptors. Herein, we synthesized three kinds of tetraphenylethene (TPE)-anchored cationic surfactants with aggregation-induced emission (AIE) characteristics. For the first time, it is quantitatively demonstrated that the CRET efficiency is inversely proportional to the sixth power of distance between luminol donors and TPE acceptors. The details disclosed in this contribute have provided the solid evidence that CRET follows Förster resonance theory. Our strategy would build a promising platform to control donor–acceptor distance, allowing to the interdisciplinary applications of CRET.*

Introduction

In 1967, chemiluminescence resonance energy transfer (CRET) was first proposed by simulating fluorescent donors in fluorescence resonance energy transfer (FRET) with chemiluminescence (CL).[1,2] Since then, CRET is subjectively assumed to follow Förster resonance theory, namely, the energy transfer efficiency is inversely proportional to the sixth power of the donor–acceptor distance.[3–5] In comparison to FRET, CRET offers several inherent advantages, including elimination of external photoexcitation, absence of autofluorescence and photobleaching, and thus it could provide excellent performances in immunoassays, macromolecular analysis, small molecule detection, bioimaging, and therapeutic applications.[6–16] Unfortunately, there is no experimental evidence to support the statement that CRET efficiency is proportional to the inverse sixth power of the donor–acceptor distance. Accordingly, CRET is limited to be qualitative, rather than quantitative. Such a gap would greatly limit the in-depth understanding and multidisciplinary applications of CRET. Therefore, it is essential to pursue a feasible strategy to measure the quantitative relationship between CRET efficiency and donor–acceptor distance.

Currently, luminol donors have been attempted to covalently link to suitable fluorescent acceptors to construct intramolecular CRET, where the donors and acceptors were separated by a rigid spacer with a fixed length.[7,8] However, the variable lengths have not yet been achieved due to two major difficulties: 1) as the spacer length increases, the poor water solubility of luminol molecule would make its luminescence efficiency decrease;[7] 2) intermolecular CRET inevitably occurs owing to Brownian motion in solution, affecting the efficiency of intramolecular CRET.[17] To solve these obstacles, it is highly required to maintain the water solubility of luminol as well as to confine the luminol donors and the acceptors in separated regions.

Cationic micelles in aqueous solution can provide the ideal separated regions, where anions and organic fluorescent molecules are confined at the micelle interface and inside the micelle, respectively.[18] In principle, when the luminol anions in a basic solution are electrostatically attracted to the micelle interface, the distances between the luminol donors and the fluorescent acceptors are able to be controlled by tuning the position of the fluorescent acceptors inside the micelle. Accordingly, the corresponding CRET efficiencies are quantitatively measured. However, the conventional acceptors inside the micelle often suffer from aggregation-caused quenching (ACQ) problem, leading to a dramatic drop in their luminescence performances.[19–21] Supposing that if aggregation-induced emission (AIE)-active acceptors can be fixed at different positions inside the micelles, their intense luminescence properties in the aggregated state could be exhibited without ACQ problem.

In this work, three kinds of AIE-active cationic surfactants are designed and synthesized by covalently incorporating tetraphenylethylene (TPE) into the 4th, 8th, and 12th methyl groups adjacent to the cationic headgroup of dodecyl trimethylammonium bromide (C_{12}TAB), namely C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB (Scheme 1). The as-prepared three kinds of cationic micelles in aqueous solution own almost the same critical micelle concentration (CMC) and micelle size. Luminol anions are electrostatically anchored at the micelle interface. As the distance between luminol donors and TPE acceptors increases from 11.9 to 22.4 Å, the CRET efficiency decreases from 99.14% to

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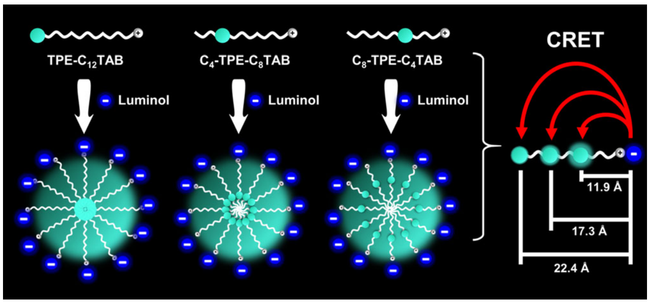

Scheme 1. Preparation of three kinds of AIE-active cationic micelles for the quantitative relationship between CRET efficiency and donor–acceptor distance.

72.39%. The measured CRET efficiency is inversely proportional to the sixth power of distance. These data are in excellent agreement with Förster resonance theory. Our strategy provides the first experimental evidence for determining the quantitative relationship between CRET efficiency and donor–acceptor distance.

Results and Discussion

The chemical structures and synthetic routes of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB were shown in Figure 1 and the Supporting Information, Figure S1, respectively. In detail, under basic conditions, 4-hydroxybenzophenone (BP-OH) was converted into BP-C_4Br, BP-C_8, BP-C_8Br, and BP-C_4 by reacting with 1,4-dibromobutane, 1-bromooctane, 1,8-dibromooctane, and 1-bromobutane, respectively. Similarly, para-hydroxy-TPE (TPE-OH) reacted with 1,12-dibromododecane to form TPE-C_{12}Br.[22] In the presence of $TiCl_4$ and zinc dust, McMurry coupling of BP-C_4Br and BP-C_8, and BP-C_8Br and BP-C_4 could produce C_8-TPE-C_4Br and C_4-TPE-C_8Br.[23] Their molecular structures were verified using ^{1}H nuclear magnetic resonance (^{1}H NMR) spectra (Supporting Information, Figures S2–S4). Thereafter, three bromine-functionalized intermediates (C_8-TPE-C_4Br, C_4-TPE-C_8Br, and TPE-C_{12}Br) were reacted with excess trimethylamine to afford three target products: C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB.[22] The ^{1}H NMR spectra (Figure 1) indicated that all hydrogen atoms were in the correct positions and the corresponding peak integrations were consistent with their structures. Moreover, the ^{13}C NMR spectra (Supporting Information, Figures S5–S7) and positive-ion mass spectra (Supporting Information, Figures S8–S10) gave further validation of the successful synthesis of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB.

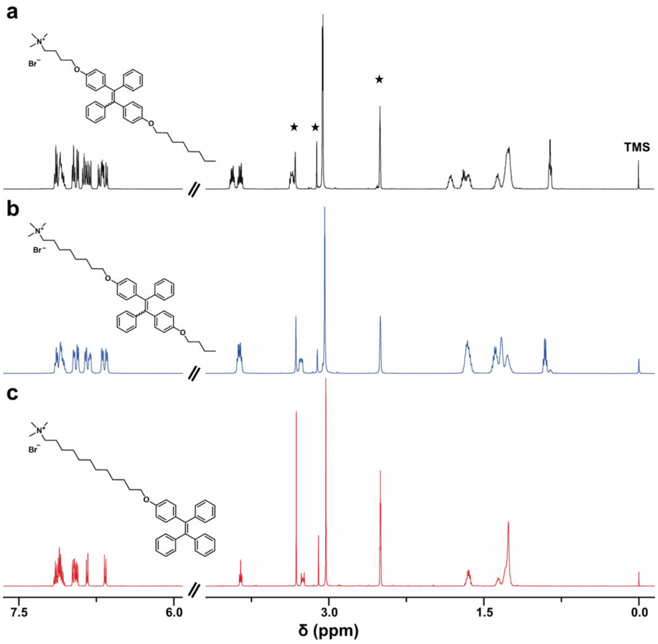

Figure 1. Chemical structures and the corresponding ^{1}H NMR spectra of a) C_8-TPE-C_4TAB, b) C_4-TPE-C_8TAB, and c) TPE-C_{12}TAB. Asterisks indicate solvent peaks, namely, water, methanol, and DMSO. TMS = tetramethylsilane.

The as-prepared C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB were typical amphiphilic molecules consisting of the same quaternary ammonium headgroup and hydrophobic tail (TPE and an alkyl chain with 12 carbon atoms). Their log *P* (octane/water partition coefficient) values were calculated to be 5.96, 5.96, and 6.57, indicating their similar amphiphilicity.[24] Moreover, the amphiphilic properties were also compared by experimentally determining the CMC.[24,25] The conductivities (κ) of the aqueous solutions of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB at different concentrations were plotted and shown in the Supporting Information, Figure S11. Their conductivities increased linearly with increasing their concentrations, with breaks at about 46, 44, and 45 μM, respectively, above which the linear slope became smaller. The break indicated the CMC, which was due to the binding of counterions to the formed ionic micelles.[26] Obviously, C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB had a similar CMC, allowing them to form micelles and perform CRET measurements at the same concentrations above CMC.

To enable that effective CRET could occur between luminol donors and TPE acceptors in the micelle solution, we studied ultraviolet-visible (UV/Vis) absorption and fluorescence properties of the aqueous solutions of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB at different concentrations, as well as CL spectrum of luminol in water. As shown in Figure 2a–c, UV-vis absorption wavelengths of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB were about 250 and 317 nm, corresponding to the absorption of the benzene and TPE groups, respectively.[22] Their absorbances at 317 nm increased linearly with increasing their concentrations from 10 to 100 μM (Supporting Information, Figure S12), indicating no change in the conjugation degree of the TPE groups before and after micelle formation.[22,24] On the other hand, their fluorescence emission wavelengths were around 485 nm under the same conditions (Figure 2a–c). Interestingly, the relationship between fluorescence intensity and concentration was consistent with the relationship between conductivity and concentration, showing two different linear relationships below and above CMC due to changes in the aggregation state of TPE groups (Supporting Information, Fig-

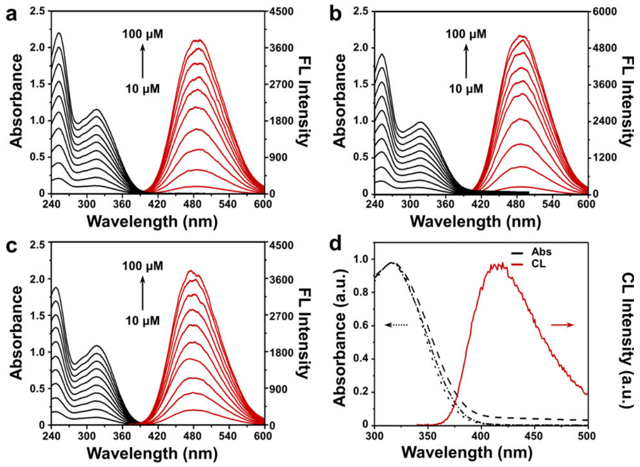

Figure 2. Absorption (black lines) and fluorescence emission (red lines) spectra of the aqueous solutions of a) C_8-TPE-C_4TAB, b) C_4-TPE-C_8TAB, and c) TPE-C_{12}TAB at concentrations of 10–100 μM. d) Normalized absorption spectra of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB (black dotted lines), and normalized CL spectrum of luminol (red solid line).

ure S13).[22,24] The fluorescence quantum yields of the three micelle solutions (80 μM) were 8.45 %, 15.19 %, and 9.10 %, indicating that the TPE groups could intensely emit in the micelles. In addition, the spectral overlap between donor emission and acceptor absorption was essential for RET to occur.[27–29] As shown in Figure 2d, the CL spectrum of luminol was recorded in the presence of 5 mM of H_2O_2 and 10 μM of Co^{2+}, which could overlap with the absorption spectra of the micelle solutions of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB.[2,16] It was worth noting that high CRET efficiency can still be obtained by shortening the donor–acceptor distance in the presence of the small overlap between the emission of donor and the absorption of acceptor.[27–29] Therefore, C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB could act as the high-performance CRET acceptors in the micelles.

To ensure that the TPE acceptors were anchored at different positions of the micelles, transmission electron microscope (TEM), dynamic light scattering (DLS) assays and theoretical calculations were carried out for the three micelle solutions.[30–35] As shown in Figure 3a–c, the TEM images showed that the three kinds of micelles had a spherical morphology with the diameters ranging from 5.0 to 5.5 nm. The DLS data (Figure 3d–f) indicated that the hydrodynamic diameters of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB micelles were about 6.19, 6.25, and 5.85 nm, respectively. Their hydrodynamic radii were further estimated by

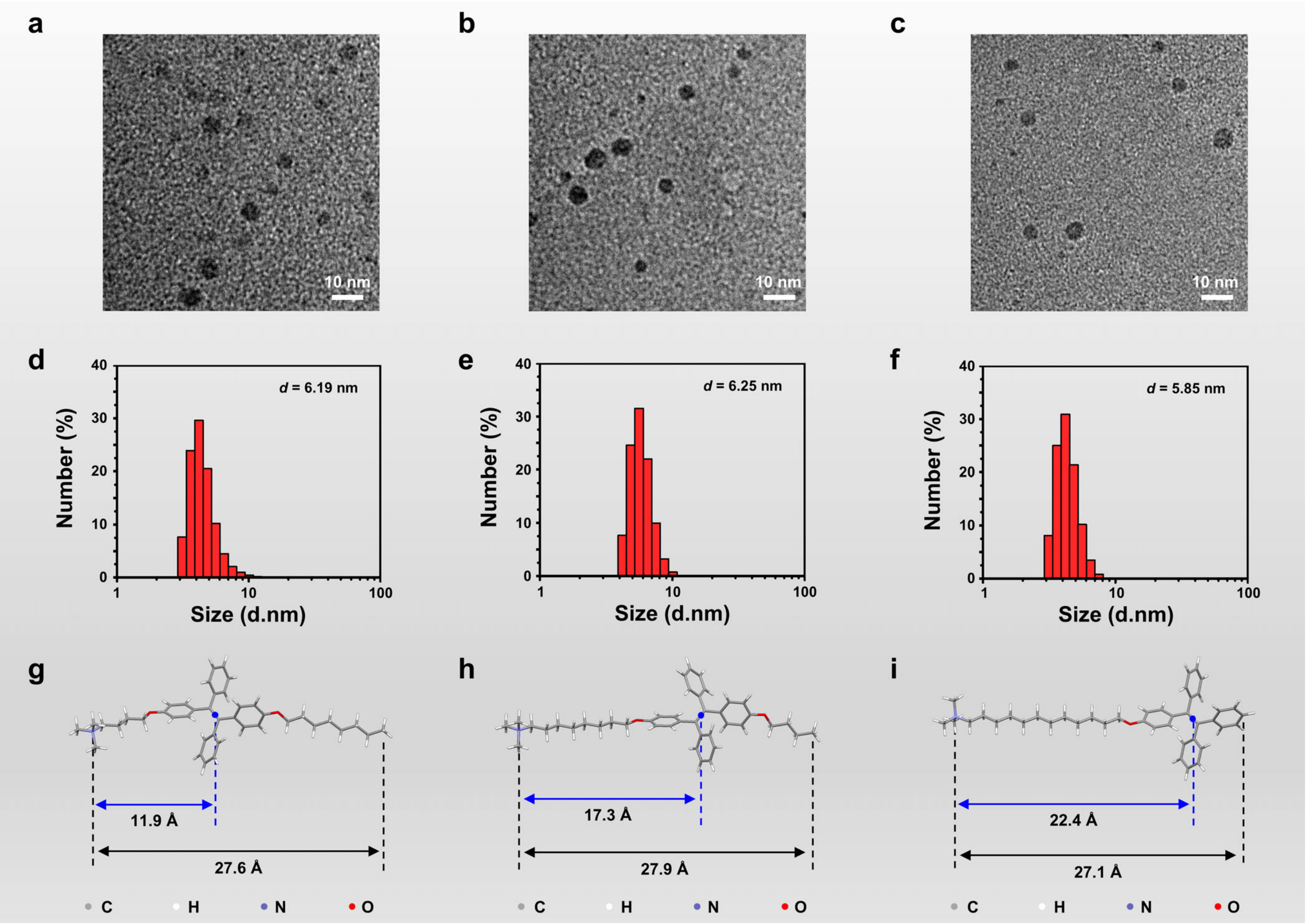

Figure 3. TEM images and DLS data of micelle solutions, and energy-minimized molecular structures of a),d),g) C_8-TPE-C_4TAB, b),e),h) C_4-TPE-C_8TAB, and c),f),i) TPE-C_{12}TAB.

theoretical calculations. The energy-minimized molecular structures (Figure 3 g–i) were optimized using the density functional theory (DFT) method B3LYP with the base 6-31G(d,p).[33,34] The distances between the C atom of the end group and the N atom were calculated to be 27.6, 27.9, and 27.1 Å, respectively, while the distance between the C atom of the headgroup and the N atom was calculated to be 1.5 Å. The thickness of solvation layer was about 2 Å.[31] By summing the molecular length, the headgroup radius, and the thickness of the solvation layer,[30–32] the estimated hydrodynamic radii of the three micelles were 3.11, 3.14, and 3.06 nm, which approximately equaled those obtained by DLS. It was worth noting that the neighboring TPE molecules were packed with overlapping benzyl fragments (ca. 4.3 Å) in the crystal lattice.[35] The hydrodynamic diameter of TPE-C_{12}TAB obtained by DLS (5.85 nm) fell within the estimated range (5.69–6.12 nm). Moreover, the effects of the presence of $Co(NO_3)_2$ on the micelle aggregates and photophysical properties were investigated. As shown in the Supporting Information, Figure S14, the hydrodynamic diameters of the three micelles remained unchanged after the addition of $Co(NO_3)_2$ because low concentrations of neutral electrolytes caused no change in the micelle size.[36] The fluorescence spectra of the micelle solutions of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB in the absence and presence of $Co(NO_3)_2$ are given in the Supporting Information, Figure S15. There was a slight increase in the fluorescence intensity with the addition of 10 μM of $Co(NO_3)_2$. The fluorescence enhancement is because the shielding effect of neutral electrolytes can reduce the electrical repulsion between the charged headgroups, allowing the tighter packing to further restrict the intramolecular motions of TPE groups.[36] Therefore, the anchored positions of the TPE acceptors in the three micelles were at 11.9, 17.3, and 22.4 Å from the cationic headgroups, respectively.

Luminol has a pK_a value of ca. 6.7, and it easily loses a proton in basic solution to form the luminol monoanion.[37–39] By physically mixing an alkaline solution of luminol with an aqueous solution of cationic micelles, the luminol monoanions could be electrostatically adsorbed onto the cationic headgroups of micelles.[40] The strong electrostatic attraction between luminol monoanions and headgroups of cationic micelles ensures that the luminol monoanions could be anchored at the micelle interface, rather than diffuse inside the micelles.[41–43] Accordingly, the distance between the luminol anions and the TPE acceptors was equal to the distance between the cationic headgroups and the center of the TPE acceptors in the C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB micelles (i.e., 11.9 Å, 17.3 Å, and 22.4 Å). Figure 4 and the Supporting Information, Figures S16–S18 show the CL spectra of different concentrations of luminol ranging from 40 to 110 μM in the presence of 80 μM of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB, respectively. Luminol could deprotonate to form luminol anion as the pH values of luminol solution and the final solutions containing micelles, luminol, and cobalt ions (Supporting Information, Table S1) were higher than the pK_a of luminol. The CRET efficiency was determined through dividing the integral area of the TPE emission spectrum by the integral area of the whole spectrum of the luminol and TPE.[8] It was found that the CRET efficiency was almost constant with increasing luminol concentrations until 60 μM, above which the CRET efficiency decreased sharply. These results indicated that the adsorption of the micelle interface was saturated when the luminol concentration reached 60 μM. Further increases in the luminol concentrations could make them dissolve in the bulk solution rather than localize on the micelle interface, resulting in the longer donor–acceptor distances. To maintain the luminol donor-TPE acceptor distance constant, the luminol concentrations in this work were employed within 60 μM. In addition, the electrostatic interaction between the luminol anions and the three cationic micelles was examined by determining the zeta potentials of the three micelle solutions in the absence and presence of 60 μM of luminol. Figures 4 c, f, and i show that the zeta potentials of the C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB micelles were 53.7, 53.4, and 55.5 mV. The addition of luminol could reduce their values to 40.3, 40.1, and 39.0 mV. The partial neutralization of zeta potentials clearly indicated that the luminol anions were adsorbed at the micelle interface through electrostatic interaction. Accordingly, 11.9 Å, 17.3 Å, and 22.4 Å can be used as the CRET donor–acceptor distances when the luminol concentrations were controlled within 60 μM.

When the luminol concentrations were 40, 50, and 60 μM, the average values of the corresponding CRET efficiencies were $99.14 \pm 0.07\%$, $93.41 \pm 0.10\%$, and $72.39 \pm 0.22\%$, respectively. To ensure that micelle formation was critical to the calculations, the energy transfer behavior was further performed at lower concentration than CMC. The CL spectra of luminol at 60 μM in the presence of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB at 20 μM are shown in the Supporting Information, Figure S19. The corresponding CRET efficiencies were calculated to be 64.06 %, 81.85 %, and 60.73 %, respectively. These results indicated that luminol anions could bind to positively charged C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB molecules to shorten their distance for CRET to occur. However, in the absence of micelle, the donor–acceptor distance was unknown. Therefore, the quantitative relationship between CRET efficiency and donor–acceptor distance must be performed in the presence of micelle.

With knowing the CRET efficiency and the distance between luminol donors and TPE acceptors, their Förster distances were calculated to be 26.3 ± 0.3, 26.9 ± 0.1, and 26.3 ± 0.1 Å, respectively ($E = R_0^{6}/(R_0^{6} + r^{6})$, where E was the CRET efficiency, and R_0 and r corresponded to the Förster distance and the donor–acceptor distance, respectively).[3–5] Apparently, they had nearly the same Förster distance. Moreover, the efficiency of resonance energy transfer as a function of distance was given by $E = (R_0/r)^j/[(R_0/r)^j + 1]$, where R_0 was the distance corresponding to 50 % transfer and j was the exponent of the distance dependence.[3–5] In the current micelle systems of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB, the calculated j was 6.0 ± 0.1.[3–5] Therefore, it was concluded that the CRET efficiency was proportional to the inverse sixth power of the donor–acceptor distance, which agreed well with Förster resonance theory.

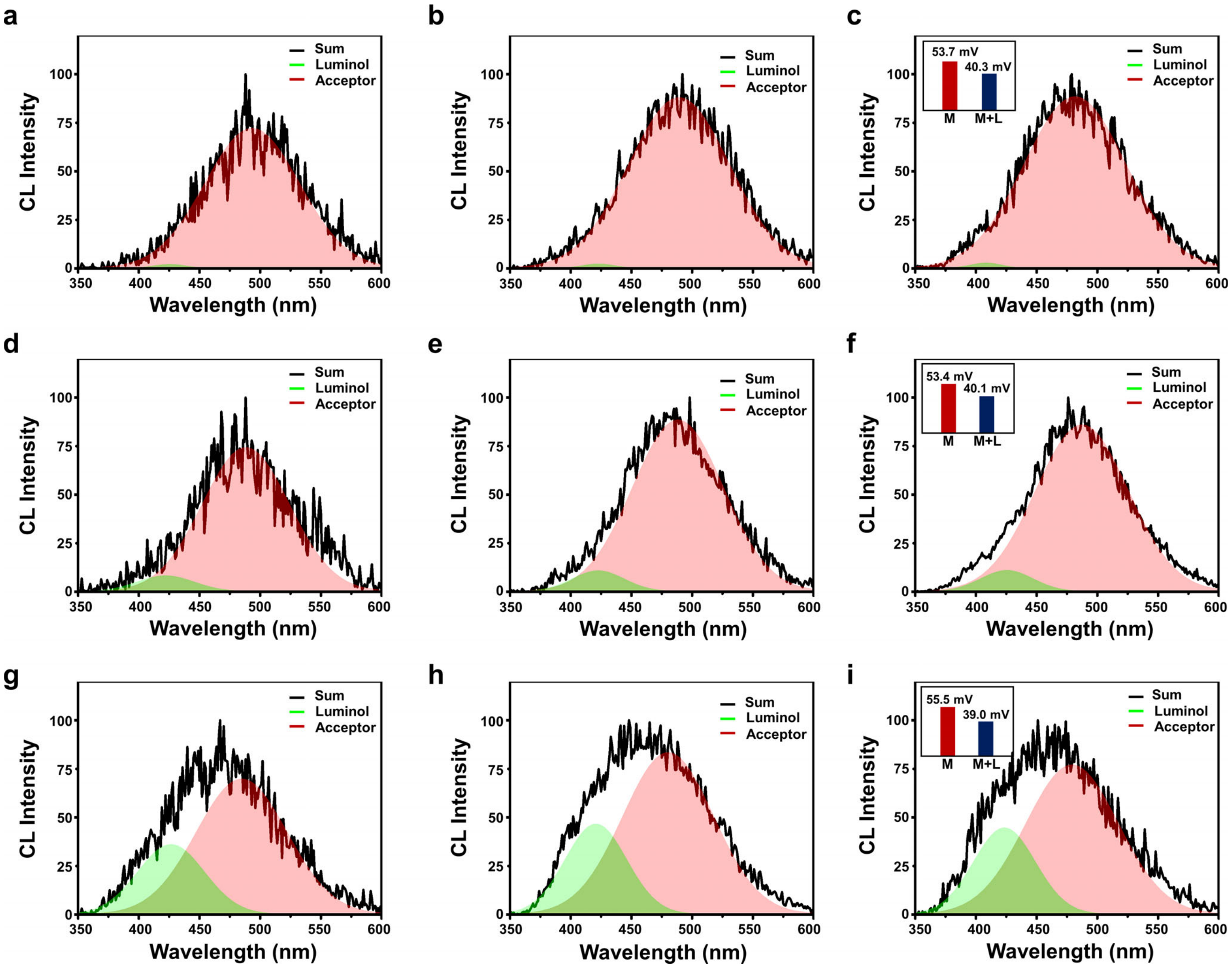

Figure 4. CL spectra of luminol (at 40, 50, and 60 μM) in the presence of a)–c) C_8-TPE-C_4TAB, d)–f) C_4-TPE-C_8TAB, and g)–i) TPE-C_{12}TAB micelles at 80 μM. Insets: corresponding zeta potentials of C_8-TPE-C_4TAB, C_4-TPE-C_8TAB, and TPE-C_{12}TAB micelles in the absence (red, M) and presence (blue, M + L) of 60 μM of luminol. Measurement time: 1.25 s, H_2O_2 concentration: 5 mM, catalyst: 10 μM of Co^{2+}.

Conclusion

We have synthesized the three kinds of AIE-active cationic surfactants. TPE in AIE-active cationic surfactant could act as a CRET acceptor by attaching to the 4th, 8th, and 12th methyl groups adjacent to the cationic headgroup. The three donor–acceptor distances were obtained by electrostatically attracting the luminol anions onto the cationic headgroups of the micelle interface. The CRET efficiency was calculated to be inversely proportional to the sixth power of distance, which agreed well with Förster resonance theory. This work not only establishes a feasible method for verifying the occurrence of the Förster resonance between chemiluminescent donors and acceptors, but also better understands the quantitative relationship between CRET efficiency and CRET donor–acceptor distance. The facile method developed in this study has the potential to greatly inspire the current research of CRET in multiple disciplines.

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Conflict of interest

The authors declare no conflict of interest.

Keywords: aggregation-induced emission · chemiluminescence · energy transfer · Förster resonance · micelles

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Research Articles

Chemiluminescence

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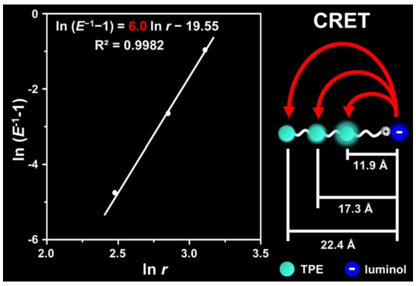
Chemiluminescence Resonance Energy Transfer Efficiency and Donor–Acceptor Distance: from Qualitative to Quantitative



More than 50 years after the birth of chemiluminescence resonance energy transfer (CRET), the first experimental evidence of the quantitative relationship between CRET efficiency and donor–acceptor distance was achieved using aggregation-induced emission (AIE)-active micelles.