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Survey, fluorescence spectra, and solubility of liquid cyanine dyes†

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We synthesized four liquid cyanine dyes, namely *N*,*N*′-didodecyl **44,** *N*,*N*′-dioctadecyl indolenium **45,** *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl)benzoxazolium **50** and benzothiazolium **51**, which have a bis(perfluorobutylsulfonyl)imide anion. In the neat form, these liquid dyes exhibit more intense fluorescence at −196 °C than at 25 °C. The liquid *N*,*N*′-dialkylated indolenium derivatives **44** and **45** exhibited larger solubility in hexane than the corresponding solid derivatives. In addition, the liquid *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl)benzoxazolium and benzothiazolium derivatives **50** and **51** were more soluble in propanol than the corresponding solid derivatives.

Introduction

Cyanine dyes are very interesting and important compounds owing to their excellent photophysical properties such as their high absorption coefficient, high fluorescence quantum yield, and long absorption and emission wavelengths. Therefore, they are used as near-infrared absorbing dyes,[1,2] probes,[3–5] sensors,[6–8] information recording materials,[9] emitters,[10] and sensitizers.[11,12] Organic dyes are usually solids because of π–π interactions, hydrogen bonding, and their strong polarity. Recently, we serendipitously discovered several neutral-type liquid dyes, coumarins,[13] (dialkylamino)perfluorophenazines,[14] and azo compounds.[15] Recently, much attention has been paid to ionic liquids, whose chemical structures consist of heteroaromatic cations and anions. They are used as reaction solvents,[16–26] electrolytes for secondary batteries[27–30] and in dye-sensitised solar cells.[31,32] Interestingly, amino acid ionic liquids,[33] magnetic ionic liquids,[34] and herbicidal ionic liquids[35] have also been reported. On this basis, we wondered whether cationic-type cyanine liquid dyes could be produced since the structure of cyanine dyes also consists of a cationic heteroaromatic moiety and an anion. To the best of our knowledge, no liquid cyanine dyes have been reported so far. As "liquid dye" is a new concept, its practical applications have not been reported yet. Herein, we report the survey of a series of liquid cyanine dyes and their properties.

Results and discussion

Scheme 1 shows the synthesis of cyanine dyes. Methyl-substituted nitrogen-containing heteroaromatic compounds **1–5** were *N*-alkylated with alkyl iodides **6–9** and/or *N*-oxaalkylated with an oxaalkyl tosylate **10** to yield *N*-alkyl **11–18** and/or *N*-oxaalkyl derivatives **19–22**, which were allowed to react with triethoxymethane to yield cyanine iodides **23–30** and/or tosylates **31–34**, respectively. These compounds were treated with tetrafluoroboric acid (**35**) and sodium or lithium reagents **36–38** to produce **39–53**.

Scheme 2 demonstrates the preparation of quinaldinium cyanine dye **57**. 6-Hydroxy-2-methylquinoline (**54**) was allowed to react with 2,5,8-trioxa-10-octadecanyl tosylate (**10**) to give a *N*,*O*-bis(2,5,8-trioxa-10-octadecanyl) derivative **55**, followed by reaction with triethoxymethane to provide **56**, which was treated with **38** to yield **57**. The purity of cyanine dyes was determined *via* ^{1}H NMR spectroscopy (Fig. S1–S17, ESI†) and elemental analyses.

Four liquid cyanine dyes, namely **44**, **45**, **50**, and **51**, were produced in this study. They were viscous liquids at room temperature. Even when they were cooled using liquid nitrogen, no crystals and/or powders were formed. Fig. 1a shows the differential scanning calorimetry (DSC) results of **44**. When compound **44** was heated from −50 to 50 °C, a small endothermic peak with a ΔH of 1.05 kJ mol^{-1} was observed at −18 °C and no peak was observed up to 50 °C. When this compound was cooled from 50 to −50 °C, a small exothermic peak was observed at −24 °C. The second heating–cooling cycle showed similar results to the first one. Thus, compound **44** was an oil and presented a glass transition temperature (T_g) of −18 °C. In contrast, compound **45** exhibited a large endothermic peak at −9 °C as shown in Fig. 1b. The shape of the peak in the second cycle was slightly smaller compared with that in the first cycle. This result suggests that although the majority of the liquid phase was crystallised by cooling, a small amount of the amorphous phase still remained. The heat of fusion was calculated to be 18.3 kJ mol^{-1}, which is significantly larger than that of **44** (1.05 kJ mol^{-1}). A freezing point was clearly observed at −19 °C. Therefore, the endothermic peak

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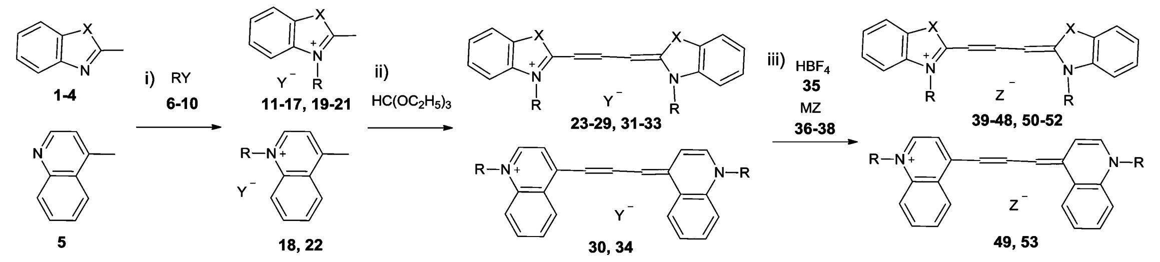
† Electronic supplementary information (ESI) available: ^{1}H NMR spectra of **23**, **39**, **40**, **41**, **42**, **43**, **44**, **45**, **46**, **47**, **48**, **49**, **50**, **51**, **52**, **53**, and **57** and thermal analyses data. See DOI: 10.1039/c6nj02160c



Compd	X
1	$C(CH_3)_2$
2	O
3	S
4	-(CH=CH)-

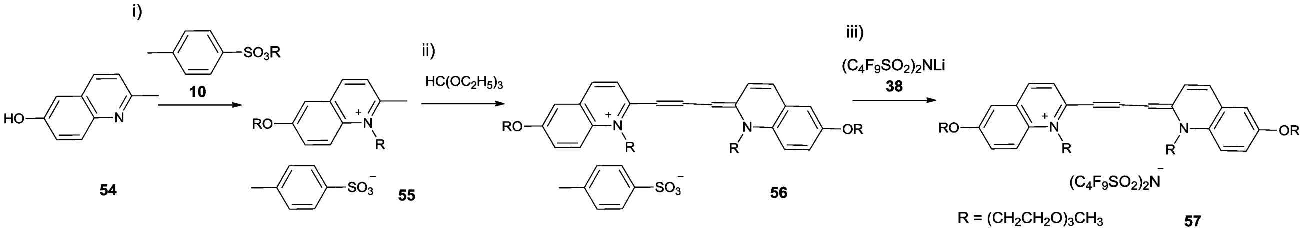
Compd	X	R	Y
11, 23	$C(CH_3)_2$	C_2H_5	I
12, 24	$C(CH_3)_2$	C_8H_{17}	I
13, 25	$C(CH_3)_2$	$C_{12}H_{25}$	I
14, 26	$C(CH_3)_2$	$C_{18}H_{37}$	I
15, 27	O	$C_{12}H_{25}$	I
16, 28	S	$C_{12}H_{25}$	I
17, 29	-(CH=CH)-	$C_{12}H_{25}$	I
18, 30	-	$C_{12}H_{25}$	I
19, 31	O	$(CH_2CH_2O)_3CH_3$	Tosylate
20, 32	S	$(CH_2CH_2O)_3CH_3$	Tosylate
21, 33	-(CH=CH)-	$(CH_2CH_2O)_3CH_3$	Tosylate
22, 34	-	$(CH_2CH_2O)_3CH_3$	Tosylate

Compd	M	Z
36	Na	$B(C_6H_5)_4$
37	Li	$(CF_3SO_2)_2N$
38	Li	$(C_4F_9SO_2)_2N$

Compd	X	R	Z
39	$C(CH_3)_2$	C_2H_5	BF_4
40	$C(CH_3)_2$	C_2H_5	$B(C_6H_5)_4$
41	$C(CH_3)_2$	C_2H_5	$(CF_3SO_2)_2N$
42	$C(CH_3)_2$	C_2H_5	$(C_4F_9SO_2)_2N$
43	$C(CH_3)_2$	C_8H_{17}	$(C_4F_9SO_2)_2N$
44	$C(CH_3)_2$	$C_{12}H_{25}$	$(C_4F_9SO_2)_2N$
45	$C(CH_3)_2$	$C_{18}H_{37}$	$(C_4F_9SO_2)_2N$
46	O	$C_{12}H_{25}$	$(C_4F_9SO_2)_2N$
47	S	$C_{12}H_{25}$	$(C_4F_9SO_2)_2N$
48	-(CH=CH)-	$C_{12}H_{25}$	$(C_4F_9SO_2)_2N$
49	-	$C_{12}H_{25}$	$(C_4F_9SO_2)_2N$
50	O	$(CH_2CH_2O)_3CH_3$	$(C_4F_9SO_2)_2N$
51	S	$(CH_2CH_2O)_3CH_3$	$(C_4F_9SO_2)_2N$
52	-(CH=CH)-	$(CH_2CH_2O)_3CH_3$	$(C_4F_9SO_2)_2N$
53	-	$(CH_2CH_2O)_3CH_3$	$(C_4F_9SO_2)_2N$

Compd	R	Y
6	C_2H_5	I
7	C_8H_{17}	I
8	$C_{12}H_{25}$	I
9	$C_{18}H_{37}$	I
10	$(CH_2CH_2O)_3CH_3$	Tosylate

Scheme 1 *Conditions and reagents*: (i) **1–5** (1.0 equiv.), **RY** (3.0 equiv.), $CHCl_3$ or toluene, rt, 2 days, (ii) **11–22** (1.0 equiv.), $CH(OC_2H_5)_3$ (0.6 equiv.), pyridine, reflux, overnight, (iii) **23–34** (1.0 equiv.), **35–38** (1.5 equiv.), $(CH_3)_2CO–H_2O$ (1 : 1), rt, overnight.



Scheme 2 *Conditions and reagents*: (i) **54** (1.0 equiv.), NaH (1.1 equiv.), **10** (3.0 equiv.), THF, reflux, 3 h, then **10** (3.0 equiv.), toluene, reflux, overnight, (ii) **55** (1.0 equiv.), $CH(OC_2H_5)_3$ (0.6 equiv.), toluene, reflux, overnight, (iii) **56** (1.0 equiv.), **38** (1.5 equiv.), $(CH_3)_2CO–H_2O$ (1 : 1), rt, overnight.

at −9 °C was assigned to the melting point of **45**. Fig. 1c indicates that compound **50** showed a similar DSC behaviour to **44** with **50** having a T_g at −22 °C (ΔH = 1.08 kJ mol^{-1}). Compound **51** also showed a similar behaviour to **50** as shown in Fig. 1d. Its T_g was observed at −23 °C with a ΔH of 0.85 kJ mol^{-1}. Thus, the cyanine dyes **44**, **45**, **50**, and **51** are liquids at room temperature. Among them, only the liquid **45** displayed melting and freezing points owing to its long octadecyl groups, which have strong alkyl–alkyl interactions.[14,15]

The effect of the counter anions (I^-, BF_4^-, BPh_4^-, $(CF_3SO_2)_2N^-$, and $(C_4F_9SO_2)_2N^-$) in 1,1′-diethylindolenium cyanine dyes **23**, **39**, **40**, **41**, and **42** on the melting point is shown in Fig. 2a. Thermal analyses of these dyes are presented in Fig. S18–S22 (ESI†). The dependence of the melting point on the anion is in the order: $(C_4F_9SO_2)_2N^-$ (**42**, 105 °C) < $(CF_3SO_2)_2N^-$ (**41**, 167 °C) < BPh_4^- (**40**, 219 °C) < BF_4^-, (**39**, 247 °C) < I^- (**23**, 267 °C). Usually, the melting points of fluorine-containing compounds is lower than those of non-fluorine-containing derivatives.[36] In addition, bulky fluorine-containing anions can reduce the π–π interactions between the cations to lower the melting point. Therefore, we prepared bis(perfluorobutylsulfonyl)imide derivatives to produce liquid cyanine dyes.

Fig. 2b shows the relationship between the melting point or T_g and the alkyl substituents on the cyanine dyes that have a bis(perfluorobutylsulfonyl)imide anion. Liquid dyes were obtained for the dodecyl **44** and octadecyl **45** derivatives. Thier DSC charts are shown in Fig. 1a and b. The ethyl **42** and octyl **43** derivatives were solids (Fig. S22 and S23, ESI†). When the alkyl group is short, π–π interactions are dominant. Conversely, the dodecyl and octadecyl groups can weaken and/or inhibit the intermolecular interactions to lower the melting point.

On the basis of these results, the *N*,*N*′-didodecyl heteroaromatic cyanine dyes with bis(perfluorobutylsulfonyl)imide anions were

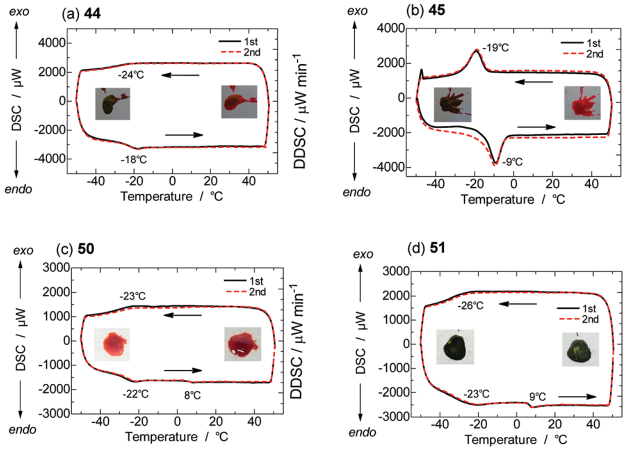


Fig. 1 DSC curves of (a) **44**, (b) **45**, (c) **50**, and (d) **51**. A sample (5–7 mg) was heated from −50 to 50 °C (10 °C min^{-1}), kept at 50 °C for 2 min, cooled from 50 to −50 °C (10 °C min^{-1}), and kept at −50 °C for 2 min under a nitrogen atmosphere. This process was repeated again.

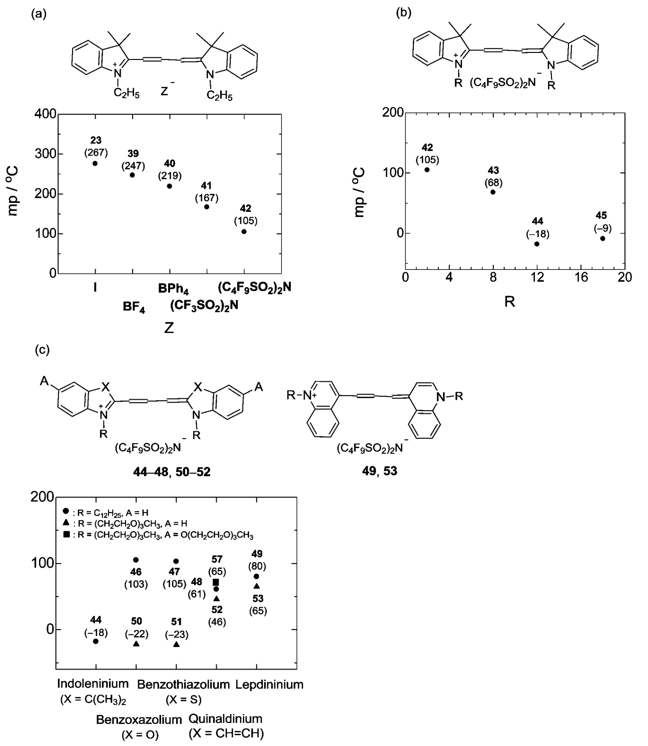


Fig. 2 Relationship between the melting point and (a) counter anions, (b) the number of carbon atoms in R, and (c) heteroaromatic moieties in cyanine dyes.

synthesised to produce a series of liquid cyanine dyes. The melting points of **46**, **47**, **48**, and **49** were observed at 103, 105, 61, and 80 °C (Fig. S24–S27, ESI†), respectively, as shown in Fig. 2c. Thus, the *N*,*N*′-didodecyl benzoxazolium, benzothiazolium, quinaldinium, and lepidinium cyanine dyes were solids. This result can be explained *via* their planar heteroaromatic moieties, which enhance π–π interactions compared with indolenium derivatives wherein two methyl groups attached to the sp^3-carbon provide steric hindrance.

To obtain liquid derivatives, oxyethylene units were introduced on the nitrogen atoms. The unshared electron pairs on the oxygen atoms of the oxyethylene unit produce an electronic repulsion, resulting in less intermolecular interactions, whereas alkyl groups have van der Waals interactions. The melting points of *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl) cyanine dyes **50**, **51**, **52** and **53** are also indicated in Fig. 2c. As expected, the benzoxazolium and benzothiazolium derivatives **50** and **51** were liquids with T_gs of −22 and −23 °C, respectively. The DSC curves are shown in Fig. 1c and d. However, the melting points of quinaldinium **52** and lepidinium dyes **53** were observed at 46 and 65 °C (Fig. S28 and S29, ESI†), respectively. The melting point of quinaldinium cyanine dye **57**, in which four oxyethylene units are introduced in a molecule, was observed at 65 °C (Fig. S30, ESI†), owing to the wide quinaldinium π-plane.

Fig. 3a and b show the effects of the counter anion and the alkyl group in indolenium cyanine dyes **23**, **39–45** on their UV-vis absorption and fluorescence spectra in toluene, respectively (Table 1). No significant differences in the spectra were observed by changing the counter anions and the alkyl substituents on the nitrogen atoms. The Stokes shift was small at *ca.* 20 nm. The observed fluorescence quantum yield (Φ_f) ranged from 0.11 to 0.14. The absorption maxima (λ_{max}) of benzoxazolium dyes **46** and **50** were observed at around 498 nm; they were the most hypsochromic among the indolenium, benzoxazolium, benzothiazolium, quinaldinium, and lepidinium cyanine dyes as shown in Fig. 3c. The fluorescence maxima (F_{max}) of **46** and **50** were observed at 512 and 511 nm, respectively. The Φ_f of **46**

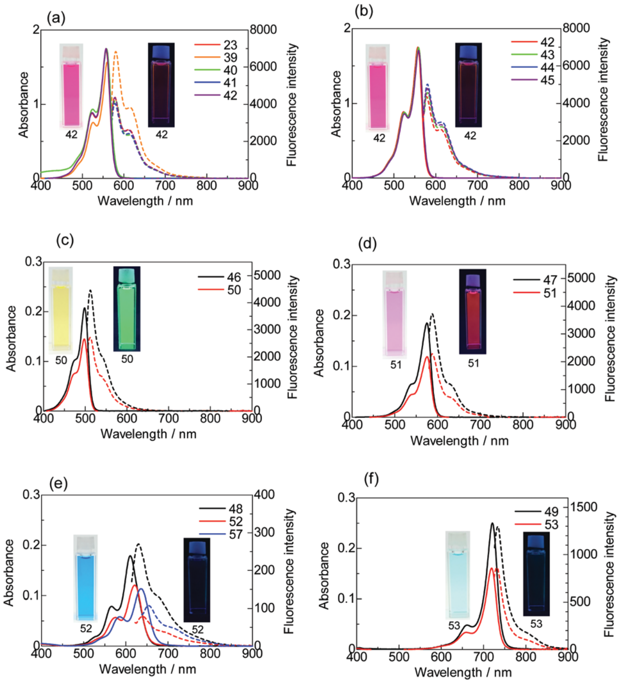


Fig. 3 UV-vis absorption and fluorescence spectra in toluene: (a) **23**, **39**, **40**, **41**, and **42** (1.0 × 10^{-5} mol dm^{-3}), (b) **42**, **43**, **44**, and **45** (1.0 × 10^{-5} mol dm^{-3}), (c) **46** and **50** (1.0 × 10^{-6} mol dm^{-3}), (d) **47** and **51** (1.0 × 10^{-6} mol dm^{-3}), (e) **48**, **52**, and **57** (1.0 × 10^{-6} mol dm^{-3}), and (f) **49** and **53** (1.0 × 10^{-6} mol dm^{-3}).

Table 1 Physical properties of cyanine dyes

Compd	mp/°C	λ_{max} (ε)^a/nm	F_{max}^{a}/nm	Φ_f^{a}
23	267	523 (88 000), 557 (175 000)	578	0.12
39	247	524 (75 000), 560 (157 000)	581	0.14
40	219	524 (93 000), 559 (174 800)	579	0.12
41	167	524 (89 000), 557 (174 000)	579	0.12
42	105	523 (89 000), 557 (175 000)	578	0.12
43	68	523 (89 000), 557 (174 000)	580	0.11
44	−18^b	524 (89 000), 559 (170 000)	580	0.12
45	−9	525 (86 000), 559 (171 000)	580	0.12
46	103	476 (sh), 498 (208 000)	512	0.28
47	105	537 (sh), 575 (185 000)	587	0.19
48	61	565 (79 000), 610 (179 000)	630	< 0.01
49	80	660 (48 000), 721 (250 000)	734	0.04
50	−22^b	472 (sh), 498 (145 000)	511	0.18
51	−23^b	537 (sh), 576 (119 000)	583	0.16
52	46	574 (57 000), 621 (121 000)	640	< 0.01
53	65	659 (34 000), 719 (167 000)	732	0.03
57	65	584 (58 000), 636 (114 000)	654	< 0.01

[a] Measured on 1.0×10^{-5} mol dm^{-3} of a substrate in toluene. [b] Glass transition temperature (T_g).

and **50** were observed to be 0.28 and 0.18, respectively. Benzothiazolium dyes **47** and **51** had their λ_{max} and F_{max} at around 575 and 585 nm in toluene, respectively, as indicated in Fig. 3d. The Φ_f values of **47** and **51** were observed to be 0.19 and 0.17, respectively. Fig. 3e depicts that the λ_{max} of quinaldinium dyes depended on the substituent in the order: *N*,*N*′-dodecyl **48** (610 nm) < *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl) **52** (621 nm) < *N*,*N*′,*O*,*O*′-tetrakis(2,5,8-trioxa-10-octadecanyl) **57** (636 nm). This result originates from the electron-donating nature of the oxyethylene unit. The F_{max} of **48**, **52**, and **57** were observed at 630, 640, and 654 nm, respectively. The Φ_f of **48**, **52**, and **57** was very small at less than 0.01. The λ_{max} of lepidinium dyes **49** and **53** were observed at 721 and 719 nm, respectively; they were amongst the most bathochromic of the cyanine dyes, as shown in Fig. 3f. Their Φ_f was small at *ca.* 0.04. The UV-vis absorption bands of the indolenium, benzoxazolium, benzothiazolium, quinaldinium, and lepidinium cyanine dyes showed a large first absorption band with a shoulder peak at a short wavelength. The fluorescence spectra were mirror images of the UV-vis absorption spectra. This result indicates that not only the $S_{0,0} \rightarrow S_{1,0}$ and $S_{1,0} \rightarrow S_{0,0}$ transitions were observed for the cyanine dyes in toluene but also the vibrational $S_{0,0} \rightarrow S_{1,1}$ and $S_{1,0} \rightarrow S_{0,1}$ transitions were seen.

Interestingly, the liquid dyes **44**, **45**, and **50** were fluorescent in their neat form. Therefore, their fluorescence spectra were compared with those of the corresponding solid derivatives **42** and **46** at 25 and −196 °C (in liquid nitrogen). The solid derivatives **42** and **46** were recrystallized from ethanol. Fig. 4a–c depict the fluorescence spectra of the solid indolenium dye **42** (mp = 105 °C) and its liquid derivatives **44** (T_g = −18 °C) and **45** (mp = −9 °C), respectively. The F_{max} of **42** was observed at 643 nm with a Φ_f of 0.12 at 25 °C. When the fluorescence spectrum was measured at −196 °C, the F_{max} was observed at 643 nm, which is at the same position as observed at 25 °C. The fluorescence intensity increased *ca.* 1.7-times at −196 °C. The F_{max} of dye **44** was at 636 nm with a Φ_f of 0.01 at 25 °C. At −196 °C, F_{max} was observed at 648 nm, which constitutes a red-shift of 12 nm.

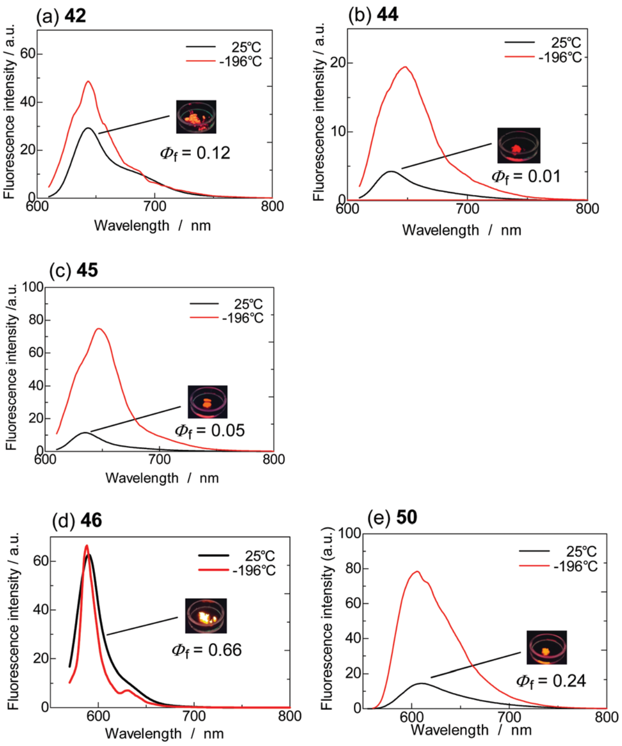


Fig. 4 Fluorescence spectra of (a) **42**, (b) **44**, (c) **45**, (d) **46**, and (e) **50** in the crystalline or neat form at 25 and −196 °C.

The fluorescence at −196 °C was *ca.* 4.5-times more intense than that at 25 °C. For **45**, F_{max} was observed at 635 with a Φ_f of 0.05 at 25 °C. At −196 °C, F_{max} was observed at 647 nm. The fluorescence was 6.6-times more intense than that at 25 °C.

Fig. 4d indicates the fluorescence spectra of the solid benzoxazolium dye **46**(mp = 103 °C). F_{max} was observed at 590 nm with a Φ_f of 0.66. This compound showed a F_{max} at 588 nm with a similar fluorescence intensity at −196 °C. Fig. 4e demonstrates that the liquid derivative **50** showed its F_{max} at 610 nm with a Φ_f of 0.25 at 25 °C. At low temperatures, its F_{max} was observed at 605 nm and its fluorescence was 5.4-times more intense.

As the intermolecular interactions of the solid derivatives **42** and **46** were scarcely affected even at −196 °C, no remarkable changes in the F_{max} and fluorescence intensity were observed. We propose that the shift of F_{max} of the liquid derivatives **44**, **45**, and **50** at −196 °C depends on the packing motif formed owing to the liquid nitrogen.[14] The increase in the fluorescence intensity of the liquid derivatives **44**, **45**, and **50** at −196 °C arises from the inhibition of rotational and vibrational processes. The increase in the fluorescence intensity of fluorophores in solution at low temperatures has been reported previously.[37] However, this paper is the first report of the fluorescence of liquid dyes in their neat form drastically increasing at low temperatures.

The difference in the solubility between the solid and liquid indolenium dyes in hexane at room temperature is shown in Fig. 5a. Usually, cationic dyes are insoluble in non-polar hexane. However, the liquid derivative **44** (T_g = −17 °C) was much more

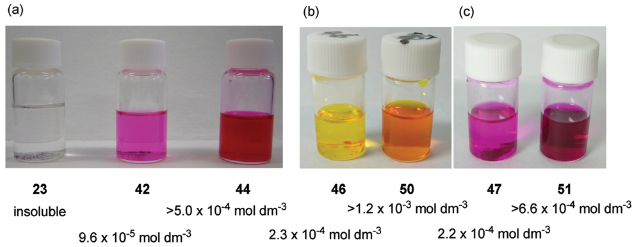


Fig. 5 Solubility of (a) **23**, **42**, and **44** in hexane, (b) **46** and **50** in propanol, and (c) **47** and **51** in propanol at 25 °C.

soluble in hexane compared to solid derivatives **23** (mp = 267 °C) and **42** (mp = 105 °C). This result may originate from the weak intermolecular interactions and affinities of the long alkyl groups of **42** and **44** with hexane. The oxyethylene derivatives were insoluble in hexane. However, they were easily soluble in ether, acetone, and methanol. The difference in the solubility between the liquid and solid derivatives was also observed in propanol; Fig. 5b shows that the liquid oxyethylene benzoxazolium dye **50** ($T_g = -22$ °C) was more soluble than the solid derivative **46** (mp = 103 °C). The liquid oxyethylene benzothiazolium dye **51** ($T_g = -23$ °C) was also more soluble than the solid derivative **47** (mp = 105 °C) as shown in Fig. 5c.

Conclusions

Liquid indolenium, benzoxazolium, and benzothiazolium cyanine dyes were synthesised. For the indolenium cyanine dyes, the *N*,*N*′-didodecyl and *N*,*N*′-dioctadecyl derivatives that had a bis(perfluorobutylsulfonyl)imide anion were liquids at room temperature. Thus, the medium alkyl-substituted derivatives are liquids, probably owing to less π–π-interactions. For the benzoxazolium and benzothiazolium cyanine dyes, the *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl) derivatives that had a bis(perfluorobutylsulfonyl)imide anion were liquids due to the electronic repulsion that comes from lone-electron pairs on the oxygen atoms. The fluorescence of the liquid cyanine dyes in their neat form was more intense (4.5–6.6 times) at −196 °C than at 25 °C, whereas that of fluorescence of the solid derivatives showed only a slight intensity increase. This result can come from the inhibition of rotational and vibrational processes at −196 °C. The *N*,*N*′-dialkylated liquid indolenium derivatives were more soluble in hexane than the solid derivatives due to the weak intermolecular interactions and affinities of the long alkyl groups with hexane. In addition, the *N*,*N*′-bis(2,5,8-trioxa-10-octadecanyl) liquid benzoxazolium and benzothiazolium derivatives were more soluble in propanol than the solid derivatives. The concept "liquid dyes" is new, and there are no practical applications yet. However, for example, liquid dyes have potential applications as inkjet dyes which need high solubility in solvents.

Experimental

Instruments

DSC and TG/DTA curves were measured with a SII Technology EXSTAR 6000 thermal analysis system. NMR spectra were obtained using JEOL 400 and 600 spectrometers. Mass spectra were obtained with a JEOL MStation JMS-700 photometer. Elemental analysis was performed with a J. Science Lab. CHN coder JM10. UV-vis absorption and fluorescence spectra were recorded on Hitachi U-4100 and JASCO FP8600 spectrophotometers, respectively.

Materials

2,3,3-Trimethylindolenine (**1**), 2-methylbenzoxazole (**2**), 2-methylbenzothiazole (**3**), 2-methylquinoline (**4**), 4-methylquinoline (**5**), iodoethane (**6**), 1-iodooctane (**7**), 1-iodododecane (**8**), 1-iodooctadecane (**9**), sodium tetraphenylborate (**36**), and triethyl orthoformate were purchased from TCI Co., Ltd. Tetrafluoroboric acid (**35**) was obtained from Sigma-Aldrich. Lithium bis(trifluoromethylsulfonyl)imide (**37**) and lithium bis(perfluorobutylsulfonyl)imide (**38**) were purchased from Wako Chemical Industries, Ltd. 2,5,8-Trioxa-10-octadecanyl tosylate (**10**),[38] 1-ethyl-2,3,3-trimethyl-3*H*-indolenium iodide (**11**),[39] 2,3,3-trimethyl-1-octyl-3*H*-indolenium iodide (**12**),[40] 2,3,3-trimethyl-1-octadecyl-3*H*-indolenium iodide (**14**),[41] 1,1′-diethyl-3,3,3′,3′-tetramethylindolenium cyanine iodide (**23**),[42] and 2-methylquinoline-6-ol (**54**)[43] were prepared as described in the literature.

Synthesis of *N*-dodecyl iodides 13, 15, 16, 17, and 18

To a chloroform solution (10 mL) of nitrogen-containing heteroaromatic compounds **1–5** (10 mmol) was added dodecyl iodide (8.9 g, 30 mmol) and refluxed for two days. After the reaction was completed, the mixture was poured into ether. The resulting precipitate was filtered and washed with ether. The crude product was dissolved in chloroform and poured into ether. The resulting precipitate was washed with ether and dried.

1-Dodecyl-2,3,3-trimethyl-3*H*-indolenium iodide (13). Yield 32%; mp 90–92 °C; ^{1}H NMR (CDCl$_3$) δ = 0.88 (t, J = 7.5 Hz, 3H), 1.24–1.29 (m, 14H), 1.36 (quin, J = 7.5 Hz, 2H), 1.45 (quin, J = 7.5 Hz, 2H), 1.67 (s, 6H), 1.94 (quin, J = 7.5 Hz, 2H), 3.13 (s, 3H), 4.67 (t, J = 7.5 Hz, 2H), 7.58–7.60 (m, 3H), 7.66–7.67 (m, 1H); FABMS (NBA) m/z 328 (M$^+$ − I).

1-Dodecyl-2-methylbenzoxazolium iodide (15). Yield 22%; mp 45–47 °C; ^{1}H NMR (CDCl$_3$) δ = 0.88 (t, J = 7.6 Hz, 3H), 1.20–1.33 (m, 14H), 1.36 (quin, J = 7.6 Hz, 2H), 1.46 (quin, J = 7.6 Hz, 2H), 2.02 (quin, J = 7.6 Hz, 2H), 3.42 (s, 3H), 4.75 (t, J = 7.6 Hz, 2H), 7.75–7.79 (m, 2H), 7.84–7.86 (m, 1H), 7.88–7.90 (m, 1H); FABMS (NBA) m/z 302 (M$^+$ − I).

1-Dodecyl-2-methylbenzothiazolium iodide (16). Yield 60%; mp 122–124 °C; ^{1}H NMR (CDCl$_3$) δ = 0.88 (t, J = 7.6 Hz, 3H), 1.24–1.30 (m, 14H), 1.36 (quin, J = 7.6 Hz, 2H), 1.48 (quin, J = 7.6 Hz, 2H), 1.95 (quin, J = 7.6 Hz, 2H), 3.44 (s, 3H), 4.85 (t, J = 7.6 Hz, 2H), 7.71 (t, J = 8.2 Hz, 1H), 7.82 (t, J = 8.2 Hz, 1H), 8.03 (d, J = 8.2 Hz, 1H), 8.42 (d, J = 8.2 Hz, 1H); FABMS (NBA) m/z 318 (M$^+$ − I).

1-Dodecyl-2-methylquinaldinium iodide (17). Yield 12%; mp 93–95 °C; ^{1}H NMR (CDCl$_3$) δ = 0.88 (t, J = 7.6 Hz, 3H), 1.22–1.32 (m, 14H), 1.42 (quin, J = 7.6 Hz, 2H) 1.63 (quin, J = 7.6 Hz, 2H), 2.00 (quin, J = 7.6 Hz, 2H), 3.34 (s, 3H), 5.06 (t, J = 7.6 Hz, 2H), 7.90 (t, J = 8.3 Hz, 1H), 8.12 (d, J = 8.3 Hz, 1H), 8.18 (t, J = 8.3 Hz, 1H), 8.30 (d, J = 8.3 Hz, 1H), 8.33 (d, J = 8.3 Hz, 1H), 8.98 (d, J = 8.3 Hz, 1H); FABMS (NBA) m/z 312 ($M^+ - I$).

1-Dodecyl-2-methyllepidinium iodide (18). Yield 90%; oil; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.6 Hz, 3H), 1.22–1.24 (m, 14H), 1.35 (quin, J = 7.6 Hz 2H), 1.50 (quin, J = 7.6 Hz, 2H), 2.10 (quin, J = 7.6 Hz, 2H), 3.05 (s, 3H), 5.27 (t, J = 7.6 Hz, 2H), 8.03 (t, J = 8.2 Hz, 1H), 8.04 (d, J = 5.5 Hz, 1H), 8.26 (t, J = 8.2 Hz, 1H), 8.43 (d, J = 8.2 Hz, 1H), 8.45 (d, J = 8.2 Hz, 1H), 10.09 (d, J = 5.5 Hz, 1H); FABMS (NBA) m/z 312 ($M^+ - I$).

Synthesis of *N*-(2,5,8-trioxa-10-octadecanyl) heteroaromatic tosylates 19–22

To a toluene solution (10 mL) of nitrogen-containing heteroaromatic compounds **1–5** (10 mmol) was added 2,5,8-trioxa-10-octadecanyl tosylate (9.54 g, 30 mmol) and refluxed for two days. After the reaction was completed, the mixture was poured into ether. The resulting precipitate was filtered. The crude product was purified by column chromatography (SiO_2, $CHCl_3$: MeOH = 10 : 1).

2-Methyl-1-(2,5,8-trioxa-10-octadecanyl)benzoxazolium tosylate (19). Yield 5%; oil; ^{1}H NMR ($CDCl_3$) δ = 2.27 (s, 3H), 3.27 (s, 3H), 3.32 (s, 3H), 3.42–3.45 (m, 8H), 3.93 (t, J = 4.9 Hz, 2H), 5.12 (t, J = 4.9 Hz, 2H), 7.02 (d, J = 7.9 Hz, 2H), 7.62 (t, J = 7.6 Hz, 1H), 7.65 (d, J = 7.9 Hz, 2H), 7.70 (t, J = 7.6 Hz, 1H), 8.09 (d, J = 7.6 Hz, 1H), 8.18 (d, J = 7.6 Hz, 1H); FABMS (NBA) m/z 280 ($M^+ - TsO$).

2-Methyl-1-(2,5,8-trioxa-10-octadecanyl)benzothiazolium tosylate (20). Yield 43%; mp 61–63 °C; ^{1}H NMR ($CDCl_3$) δ = 2.27 (s, 3H), 3.26 (s, 3H), 3.32 (s, 3H), 3.41–3.46 (m, 8H), 3.93 (t, J = 4.9 Hz, 2H), 5.12 (t, J = 4.9 Hz, 2H), 7.02 (d, J = 7.9 Hz, 2H), 7.62 (t, J = 7.6 Hz, 1H), 7.65 (d, J = 7.9 Hz, 2H), 7.70 (t, J = 7.6 Hz, 1H) 8.09 (d, J = 7.6 Hz, 1H), 8.18 (d, J = 7.6 Hz, 1H); FABMS (NBA) m/z 296 ($M^+ - TsO$).

2-Methyl-1-(2,5,8-trioxa-10-octadecanyl)quinaldinium tosylate (21). Yield 12%; oil; ^{1}H NMR ($CDCl_3$) δ = 2.31 (s, 3H), 3.25 (s, 3H), 3.32 (s, 3H), 3.42–3.70 (m, 8H), 4.13–4.15 (m, 2H), 5.48–5.49 (m, 2H), 7.09 (d, J = 7.9 Hz, 2H), 7.73 (d, J = 7.9 Hz, 2H), 7.81 (t, J = 7.6 Hz, 1H), 7.84 (d, J = 7.6 Hz, 1H), 8.10 (t, J = 7.6 Hz, 1H), 8.13 (d, J = 7.6 Hz, 1H), 8.58 (d, J = 8.2 Hz, 1H), 8.75 (d, J = 8.2 Hz, 1H); FABMS (NBA) m/z 290 ($M^+ - TsO$).

2-Methyl-1-(2,5,8-trioxa-10-octadecanyl)lepidinium tosylate (22). Yield 69%; oil; ^{1}H NMR ($CDCl_3$) δ = 2.27 (s, 3H), 2.91 (s, 3H), 3.31 (s, 3H), 3.43–3.46 (m, 6H), 3.55 (t, J = 4.8 Hz, 2H), 4.03 (t, J = 4.8 Hz, 2H), 5.35 (t, J = 4.8 Hz, 2H), 7.06 (d, J = 7.9 Hz, 2H), 7.74 (d, J = 7.9 Hz, 2H), 7.84 (d, J = 8.2 Hz, 1H), 7.88 (t, J = 8.2 Hz, 1H), 8.09 (t, J = 8.2 Hz, 1H), 8.25 (d, J = 8.2 Hz, 1H), 8.56 (d, J = 8.2 Hz, 1H), 9.44 (d, J = 8.2 Hz, 1H); FABMS (NBA) m/z 290 ($M^+ - TsO$).

Synthesis of cyanine dyes 23–34. To pyridine (5 mL) were added compounds **11–22** (2 mmol) and triethyl orthoformate (178 mg, 1.2 mmol). The mixture was refluxed overnight. After cooling, pyridine was removed *in vacuo*. The product was extracted with dichloromethane (50 mL × 3). The extract was dried over anhydrous sodium sulfate. After removing the solvent *in vacuo*, the product was purified by chromatography (SiO_2, $CHCl_3$: MeOH = 10 : 1) and recrystallized from ethanol. No molecular ion peak was observed for **31**, **32**, **33**, and **34** by FABMS.

1,1′-Diethyl-3,3,3′,3′-tetramethylindolenium carbocyanine iodide (23). Yield 95%; ^{1}H NMR ($CDCl_3$) δ = 1.51 (t, J = 7.2 Hz, 6H), 1.71 (s, 12H), 4.34 (q, J = 7.2 Hz, 4H), 7.14 (d, J = 8.5 Hz, 2H), 7.25 (t, J = 8.5 Hz, 2H), 7.37 (d, J = 8.5 Hz, 2H), 7.38 (t, J = 8.5 Hz, 2H), 7.44 (d, J = 13.3 Hz, 2H), 8.44 (t, J = 13.3 Hz, 1H); FABMS (NBA) 385 ($M^+ - I$).

3,3,3′,3′-Tetramethyl-1,1′-dioctylindolenium carbocyanine iodide (24). Yield 67%; mp 86–88 °C; ^{1}H NMR ($CDCl_3$) δ = 0.86 (t, J = 7.4 Hz, 6H), 1.22–1.40 (m, 16H), 1.56 (quin, J = 7.4 Hz, 4H), 1.71 (s, 12H), 1.88 (quin, J = 7.4 Hz, 4H), 4.28 (t, J = 7.4 Hz, 4H), 7.11 (d, J = 7.8 Hz, 2H), 7.24 (t, J = 7.8 Hz, 2H), 7.36 (d, J = 7.8 Hz, 2H), 7.37 (t, J = 7.8 Hz, 2H), 7.46 (d, J = 13.6 Hz, 2H), 8.44 (t, J = 13.6 Hz, 1H); FABMS (NBA) m/z 554 ($M^+ - I$); anal. found: C, 68.50; H, 8.52; N, 4.10%. Calcd for $C_{39}H_{57}IN_2$: C, 68.81; H, 8.44; N, 4.11%.

1,1′-Didodecyl-3,3,3′,3′-tetramethylindolenium carbocyanine iodide (25). Yield 70%; mp 36 °C; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.5 Hz, 6H), 1.23–1.28 (m, 28H), 1.36 (quin, J = 7.5 Hz, 4H), 1.54 (quin, J = 7.5 Hz, 4H), 1.72 (s, 12H) 1.89 (quin, J = 7.5 Hz, 4H), 4.27 (t, J = 7.5 Hz, 4H), 7.12 (d, J = 7.6 Hz, 2H), 7.25 (t, J = 7.6 Hz, 2H), 7.33 (d, J = 13.4 Hz, 2H), 7.37 (d, J = 7.6 Hz, 2H), 7.39 (t, J = 7.6 Hz, 2H), 8.43 (t, J = 13.4 Hz, 1H); FABMS (NBA) m/z 666 ($M^+ - I$).

1,1′-Dioctadecyl-3,3,3′,3′-tetramethylindolenium carbocyanine iodide (26). Yield 72%; mp 11 °C; ^{1}H NMR ($CDCl_3$) δ = 0.88 (t, J = 7.5 Hz, 6H), 1.24–1.29 (m, 52H), 1.37 (quin, J = 7.5 Hz, 4H), 1.54 (quin, J = 7.5 Hz, 4H) 1.74 (s, 12H), 1.88 (quin, J = 7.5 Hz, 4H), 4.23 (t, J = 7.5 Hz, 4H), 7.02 (d, J = 13.7 Hz, 2H), 7.14 (d, J = 7.6 Hz, 2H), 7.27 (t, J = 7.6 Hz, 2H), 7.40 (t, J = 7.6 Hz, 2H), 7.41 (d, J = 7.6 Hz, 2H), 8.44 (t, J = 13.7 Hz, 1H); FABMS (NBA) m/z 834 ($M^+ - I$).

1,1′-Didodecylbenzoxazolium carbocyanine iodide (27). Yield 67%; mp 150 °C; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.6 Hz, 6H), 1.23–1.29 (m, 28H), 1.38 (quin, J = 7.6 Hz, 4H), 1.48 (quin, J = 7.6 Hz, 4H), 1.92 (quin, J = 7.6 Hz, 4H), 4.27 (t, J = 7.6 Hz, 4H), 6.80 (d, J = 13.0 Hz, 2H), 7.28 (d, J = 8.2 Hz, 2H), 7.33 (t, J = 8.2 Hz, 2H), 7.39 (t, J = 8.2 Hz, 2H), 7.47 (d, J = 8.2 Hz, 2H), 8.48 (t, J = 13.0 Hz, 1H); FABMS (NBA) m/z 614 ($M^+ - I$).

1,1′-Didodecylbenzothiazolium carbocyanine iodide (28). Yield 33%; mp 155 °C; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.6 Hz, 6H), 1.23–1.29 (m, 28H), 1.35 (quin, J = 7.6 Hz, 4H), 1.54 (quin, J = 7.6 Hz, 4H), 1.87 (quin, J = 7.6 Hz, 4H), 4.35 (t, J = 7.6 Hz, 4H), 7.28 (d, J = 7.6 Hz, 2H), 7.31 (t, J = 7.6 Hz, 2H), 7.37 (d, J = 12.4 Hz, 2H), 7.46 (t, J = 7.6 Hz, 2H), 7.69 (d, J = 7.6 Hz, 2H), 7.73 (t, J = 12.4 Hz, 1H); FABMS (NBA) m/z 646 ($M^+ - I$).

1,1′-Didodecylquinaldinium carbocyanine iodide (29). Yield 21%; 199 °C; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.3 Hz, 6H), 1.26–1.43 (m, 36H), 1.62–1.79 (m, 4H), 4.27 (br s, 4H), 6.41 (d, J = 12.8 Hz, 2H), 7.18 (t, J = 8.2 Hz, 2H), 7.26 (d, J = 7.6 Hz, 2H), 7.40 (d, J = 8.2 Hz, 2H), 7.45 (t, J = 8.2 Hz, 2H), 7.73 (d, J = 9.4 Hz, 2H), 8.66 (d, J = 9.4 Hz, 2H), 8.70 (t, J = 12.8 Hz, 1H); FABMS (NBA) m/z 634 ($M^+ - I$).

1,1′-Didodecyllepidinium carbocyanine iodide (30). Yield 26%; mp 133 °C; ^{1}H NMR ($CDCl_3$) δ = 0.87 (t, J = 7.6 Hz, 6H), 1.24–1.32 (m, 32H), 1.40 (quin, J = 7.6 Hz, 4H), 1.86 (quin, J = 7.6 Hz, 4H), 4.23 (t, J = 7.6 Hz, 4H), 6.87 (d, J = 13.0 Hz, 2H), 7.47 (t, *J* = 7.6 Hz, 2H), 7.51 (d, *J* = 7.6 Hz, 2H), 7.69 (t, *J* = 7.6 Hz, 2H), 7.91 (d, *J* = 7.6 Hz, 2H), 7.94 (d, *J* = 7.6 Hz, 2H), 8.24 (d, *J* = 7.6 Hz, 2H), 8.60 (t, *J* = 13.0 Hz, 1H); FABMS (NBA) *m*/*z* 634 ($M^+ - I$).

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)benzoxazolium carbocyanine tosylate (31). Yield 51%; ^{1}H NMR ($CDCl_3$) δ = 2.31 (s, 3H), 3.33 (s, 6H), 3.43–3.45 (m, 4H), 3.48–3.52 (m, 8H), 3.59–3.61 (m, 4H), 3.95 (t, *J* = 4.8 Hz, 4H), 4.42 (t, *J* = 4.8 Hz, 4H), 6.66 (d, *J* = 13.2 Hz, 2H), 7.13 (d, *J* = 7.8 Hz, 2H), 7.28 (t, *J* = 7.6 Hz, 2H), 7.33 (t, *J* = 7.6 Hz, 2H), 7.43 (d, *J* = 7.6 Hz, 2H), 7.46 (d, *J* = 7.6 Hz, 2H), 7.85 (d, *J* = 7.8 Hz, 2H), 8.43 (t, *J* = 13.2 Hz, 1H).

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)benzothiazolium carbocyanine tosylate (32). Yield 67%; ^{1}H NMR ($CDCl_3$) δ = 2.35 (s, 3H), 3.24 (s, 6H), 3.40–3.42 (m, 4H), 3.44–3.45 (m, 4H), 3.46–3.48 (m, 4H), 3.54–3.56 (m, 4H), 3.97 (t, *J* = 4.1 Hz, 4H), 4.60 (t, *J* = 4.1 Hz, 4H), 7.14 (d, *J* = 12.4 Hz, 2H), 7.29 (t, *J* = 7.8 Hz, 2H), 7.42 (t, *J* = 7.8 Hz, 2H), 7.43 (d, *J* = 7.6 Hz, 2H), 7.56 (d, *J* = 7.8 Hz, 2H), 7.57 (d, *J* = 7.8 Hz, 2H), 7.72 (t, *J* = 12.4 Hz, 1H), 7.86 (d, *J* = 7.6 Hz, 2H).

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)quinaldinium carbocyanine tosylate (33). Yield 14%; ^{1}H NMR ($CDCl_3$) δ = 2.35 (s, 3H), 3.33 (s, 6H), 3.44–3.45 (m, 4H), 3.50–3.52 (m, 4H), 3.54–3.56 (m, 4H), 3.62–3.64 (m, 4H), 4.00 (br s, 4H), 4.60 (br s, 4H), 6.56 (d, *J* = 13.1 Hz, 2H), 7.17 (d, *J* = 8.2 Hz, 2H), 7.20 (t, *J* = 8.2 Hz, 2H), 7.42 (d, *J* = 7.6 Hz, 2H), 7.47 (t, *J* = 8.2 Hz, 2H), 7.65 (d, *J* = 8.2 Hz, 2H), 7.67 (d, *J* = 8.2 Hz, 2H), 7.94 (d, *J* = 7.6 Hz, 2H), 8.40 (d, *J* = 8.2 Hz, 2H), 8.70 (t, *J* = 13.1 Hz, 1H).

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)lepidinium carbocyanine tosylate (34). Yield 19%; ^{1}H NMR ($CDCl_3$) δ = 2.32 (s, 3H), 3.31 (s, 6H), 3.43–3.44 (m, 4H), 3.50–3.52 (m, 8H), 3.55–3.56 (m, 4H), 3.82–3.84 (m, 4H), 4.39–4.42 (m, 4H), 6.73 (d, *J* = 13.1 Hz, 2H), 7.16 (d, *J* = 8.2 Hz, 2H), 7.36–7.38 (m, 4H), 7.60–7.63 (m, 4H), 7.71 (d, *J* = 8.2 Hz, 2H), 7.94 (d, *J* = 7.6 Hz, 2H), 8.09 (d, *J* = 8.2 Hz, 2H), 8.28 (t, *J* = 13.1 Hz, 1H).

Syntheses of 39–53. To an acetone–chloroform 1 : 1 mixed solution (10 mL) of iodides **23–30** or tosylates **31–34** (0.30 mmol) were added tetrafluoroboric acid (**35**), sodium tetraphenylborate (**36**), lithium bis(trifluoromethylsulfonyl)imide (**37**), and lithium bis(perfluorobutylsulfonyl)imide (**38**) (0.45 mmol). The mixture was stirred overnight. After the reaction was completed, the solvent was removed *in vacuo*. The residue was dissolved in dichloromethane, purified by column chromatography (SiO_2, $CHCl_3 : MeOH = 10 : 1$) and recrystallized from ethanol. Compounds **44** and **45** were purified by column chromatography (SiO_2, $C_6H_{14} : CHCl_3 = 1 : 1$, $CHCl_3$, and then $CHCl_3 : MeOH = 10 : 1$). In the case of **42–54**, no carbon peaks assigned to the perfluorobutyl group of the counter anion were shown.

1,1′-Diethyl-3,3,3′,3′-tetramethylindolenium carbocyanine tetrafluoroborate (39). Yield 59%; ^{1}H NMR ($CDCl_3$) δ = 1.48 (t, *J* = 7.6 Hz, 6H), 1.72 (s, 12H), 4.22 (q, *J* = 7.6 Hz, 4H), 6.81 (d, *J* = 13.0 Hz, 2H), 7.14 (d, *J* = 7.7 Hz, 2H), 7.25 (t, *J* = 7.7 Hz, 2H), 7.37 (d, *J* = 7.7 Hz, 2H), 7.40 (t, *J* = 7.7 Hz, 2H), 8.40 (t, *J* = 13.0 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ = 12.5 (2C), 28.0 (4C), 39.7 (2C), 49.0 (2C), 104.0 (2C), 110.8 (2C), 122.1 (2C), 125.2 (2C), 128.9 (2C), 140.7 (2C), 141.8 (2C), 150.9, 173.3 (2C); ^{19}F NMR ($CDCl_3$) δ = −152.7 (4F); FABMS (NBA) *m*/*z* 385 ($M^+ - BF_4$); anal. found: C, 68.32; H, 7.43; N, 5.89%; calcd for $C_{27}H_{33}BF_4N_2$: C, 68.65; H, 7.04; N, 5.93%.

1,1′-Diethyl-3,3,3′,3′-tetramethylindolenium carbocyanine tetraphenylborate (40). Yield 77%; ^{1}H NMR ($CDCl_3$) δ = 1.18 (t, *J* = 7.3 Hz, 6H), 1.68 (s, 12H), 3.60 (q, *J* = 7.3 Hz, 4H), 5.87 (d, *J* = 13.3 Hz, 2H), 6.86 (t, *J* = 7.3 Hz, 4H), 6.99–7.03 (m, 10H), 7.26 (t, *J* = 7.3 Hz, 2H), 7.35 (d, *J* = 7.3 Hz, 2H), 7.39 (t, *J* = 7.3 Hz, 2H), 7.43–7.47 (m, 8H), 8.28 (t, *J* = 13.3 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ = 12.4 (2C), 27.9 (4C), 39.4 (2C), 49.1 (2C), 103.2 (2C), 110.9 (2C), 121.7 (4C), 122.1 (2C), 125.46 (2C), 125.52 (8C), 129.0 (2C), 136.3 (8C), 140.6 (2C), 141.5 (2C), 150.3, 173.2 (2C); in this compound, no four-carbon peaks assigned to the carbon atoms attached to boron were observed. FABMS (NBA) *m*/*z* 385 ($M^+ - BPh_4$); anal. found: C, 86.64; H, 7.65; N, 3.66%; calcd for $C_{51}H_{53}BN_2$: C, 86.91; H, 7.58; N, 3.97%.

1,1′-Diethyl-3,3,3′,3′-tetramethylindolenium carbocyanine bis(trifluoromethylsulfonyl)imide (41). Yield 87%; ^{1}H NMR ($CDCl_3$) δ = 1.45 (t, *J* = 7.2 Hz, 6H), 1.73 (s, 12H), 4.16 (q, *J* = 7.2 Hz, 4H), 6.45 (d, *J* = 13.4 Hz, 2H), 7.14 (d, *J* = 7.8 Hz, 2H), 7.28 (t, *J* = 7.8 Hz, 2H), 7.40 (d, *J* = 7.8 Hz, 2H), 7.41 (t, *J* = 7.8 Hz, 2H), 8.43 (t, *J* = 13.4 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ = 12.4 (2C), 28.1 (4C), 39.6 (2C), 49.3 (2C), 103.0 (2C), 110.9 (2C), 120.1 (q, *J* = 320 Hz, 2C), 122.4 (2C), 125.7 (2C), 129.1 (2C), 140.8 (2C), 141.6 (2C), 150.8, 173.8 (2C); ^{19}F NMR ($CDCl_3$, ext. $CFCl_3$) δ = −78.5 (3F); FABMS (NBA) *m*/*z* 385 ($M^+ - (CF_3SO_2)_2N$); anal. found: C, 52.14; H, 5.01; N, 6.21%. Calcd for $C_{29}H_{33}F_6N_3O_4S_2$: C, 52.32; H, 5.00; N, 6.31%.

1,1′-Diethyl-3,3,3′,3′-tetramethylindolenium carbocyanine bis(perfluorobutylsulfonyl)imide (42). Yield 66%; ^{1}H NMR ($CDCl_3$) δ = 1.43 (t, *J* = 7.2 Hz, 6H), 1.73 (s, 12H), 4.16 (q, *J* = 7.2 Hz, 4H), 6.47 (d, *J* = 13.7 Hz, 2H), 7.14 (d, *J* = 8.2 Hz, 2H), 7.27 (t, *J* = 8.2 Hz, 2H) 7.40 (d, *J* = 8.2 Hz, 2H), 7.41 (t, *J* = 8.2 Hz, 2H), 8.42 (t, *J* = 13.7 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ = 12.2 (2C), 27.9 (4C), 39.5 (2C), 49.2 (2C), 102.9 (2C), 110.8 (2C), 122.3 (2C), 125.6 (2C), 129.0 (2C), 140.7 (2C), 141.6 (2C), 150.7, 173.7 (2C); ^{19}F NMR ($CDCl_3$, ext. $CFCl_3$) δ = −125.8 (4F), −120.9 (4F), −112.7 (4F), −80.6 (6F); FABMS (NBA) *m*/*z* 385 ($M^+ - (C_4F_9SO_2)_2N$); anal. found: C, 43.45; H, 3.50; N, 4.39%. Calcd for $C_{35}H_{33}F_{18}N_3O_4S_2$: C, 43.53; H, 3.44; N, 4.35%.

3,3,3′,3′-Tetramethyl-1,1′-dioctylindolenium carbocyanine bis(perfluorobutylsulfonyl)imide (43). Yield 56%; ^{1}H NMR ($CDCl_3$) δ = 0.86 (t, *J* = 8.3 Hz, 6H), 1.22–1.30 (m, 12H), 1.34 (quin, *J* = 8.3 Hz, 4H), 1.44 (quin, *J* = 8.3 Hz, 4H), 1.70 (s, 12H), 1.82 (quin, *J* = 8.3 Hz, 4H), 4.09 (t, *J* = 8.3 Hz, 4H), 6.42 (d, *J* = 13.7 Hz, 2H), 7.12 (d, *J* = 7.6 Hz, 2H), 7.27 (t, *J* = 7.6 Hz, 2H), 7.39 (d, *J* = 7.6 Hz, 2H), 7.50 (t, *J* = 7.6 Hz, 2H), 8.42 (t, *J* = 13.7 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ = 14.1 (2C), 22.7 (2C), 26.8 (2C), 27.6 (2C), 28.1 (4C), 29.1 (2C), 29.4 (2C), 31.8 (2C), 44.6 (2C), 49.3 (2C), 103.2 (2C), 111.2 (2C), 122.3 (2C), 125.6 (2C), 129.0 (2C), 140.7 (2C), 142.1 (2C), 150.6, 174.1 (2C); ^{19}F NMR ($CDCl_3$, ext. $CFCl_3$) δ = −125.8 (4F), −120.9 (4F), −112.6 (4F), −80.7 (6F); FABMS (NBA) *m*/*z* 554 ($M^+ - (C_4F_9SO_2)_2N$); anal. Found: C, 50.01; H, 5.17; N, 3.67%. Calcd for $C_{47}H_{57}F_{18}N_3O_4S_2$: C, 49.78; H, 5.07; N, 3.71%.

1,1′-Didodecyl-3,3,3′,3′-tetramethylindolenium carbocyanine bis(perfluorobutylsulfonyl)imide (44). Yield 86%; ^{1}H NMR ($CDCl_3$) δ = 0.86 (t, *J* = 7.6 Hz, 6H), 1.20–1.31 (m, 28H), 1.34

(quin, J = 7.6 Hz, 4H), 1.44 (quin, J = 7.6 Hz, 4H), 1.72 (s, 12H), 1.81 (quin, J = 7.6 Hz, 4H), 4.10 (t, J = 7.6 Hz, 4H), 6.43 (d, J = 13.1 Hz, 2H), 7.12 (d, J = 7.5 Hz, 2H), 7.26 (t, J = 7.5 Hz, 2H), 7.39 (d, J = 7.5 Hz, 2H), 7.40 (t, J = 7.5 Hz, 2H), 8.41 (t, J = 13.1 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 14.1 (2C), 22.7 (2C), 26.8 (2C), 27.5 (2C), 28.0 (6C), 29.4 (4C), 29.6 (6C), 31.9 (2C), 44.5 (2C), 49.2 (2C), 103.2 (2C), 111.1 (2C), 122.2 (2C), 125.5 (2C), 128.9 (2C), 140.6 (2C), 142.1 (2C), 150.5, 174.0 (2C); ^{19}F NMR (CDCl$_3$, ext. CFCl$_3$) δ = −125.8 (4F), −120.9 (4F), −112.6 (4F), −80.7 (6F); FABMS (NBA) m/z 666 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. Found: C, 52.96; H, 6.05; N, 3.31%; calcd for $C_{55}H_{73}N_3O_4S_2$: C, 53.00; H, 5.90; N, 3.37%.

1,1′-Dioctadecyl-3,3,3′,3′-tetramethylindolenium carbocyanine bis(perfluorobutylsulfonyl)imide (45). Yield 94%; ^{1}H NMR (CDCl$_3$) δ = 0.88 (t, J = 7.6 Hz, 6H), 1.24–1.38 (m, 56H), 1.44 (quin, J = 7.6 Hz, 4H), 1.73 (s, 12H), 1.82 (quin, J = 7.6 Hz, 4H), 4.09 (t, J = 7.6 Hz, 4H), 6.39 (d, J = 13.7 Hz, 2H), 7.12 (d, J = 6.9 Hz, 2H), 7.27 (t, J = 6.9 Hz, 2H), 7.39 (d, J = 6.9 Hz, 2H), 7.40 (t, J = 6.9 Hz, 2H), 8.42 (t, J = 13.7 Hz,1H); ^{13}C NMR (CDCl$_3$) δ=14.1 (2C), 22.7 (2C), 26.8 (2C), 27.5 (2C), 28.0 (6C), 29.4 (4C), 29.7 (18C), 31.9 (2C), 44.5 (2C), 49.3 (2C), 103.0 (2C), 111.1 (2C), 122.2 (2C), 125.6 (2C), 128.9 (2C), 140.6 (2C), 142.1 (2C), 150.5, 174.1 (2C); ^{19}F NMR (CDCl$_3$, ext. CFCl$_3$) δ = −125.8 (4F), −120.9 (4F), −112.6 (4F), −80.7 (6F); FABMS (NBA) m/z 834 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 56.66; H, 6.67; N, 2.88%; calcd for $C_{67}H_{97}N_3O_4S_2$: C, 56.89; H, 6.91; N, 2.97%.

1,1′-Didodecylbenzoxazolium carbocyanine bis(perfluorobutylsulfonyl)imide (46). Yield 73%; ^{1}H NMR (CDCl$_3$) δ = 0.87 (t, J = 7.4 Hz, 6H), 1.23–1.45 (m, 36H), 1.85 (quin, J = 7.4 Hz, 4H), 4.11 (t, J = 7.4 Hz, 4H), 6.10 (d, J = 13.3 Hz, 2H), 7.26 (d, J = 8.2 Hz, 2H), 7.32–7.41 (m, 4H), 7.50 (d, J = 8.2 Hz, 2H), 8.47 (t, J = 13.3 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 14.0 (2C), 22.6 (2C), 26.6 (2C), 27.9 (2C), 29.2 (2C), 29.3 (2C), 29.3 (2C), 29.5 (2C), 29.5 (4C), 31.8 (2C), 44.5 (2C), 85.9 (2C), 110.3 (2C), 110.8 (2C), 125.1 (2C), 126.0 (2C), 121.3 (2C), 146.8 (2C), 148.0, 162.1 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.8 (4F), −120.9 (4F), −112.7 (4F), −80.7 (6F); FABMS (NBA) m/z 614 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 49.65; H, 5.38; N, 3.43%; calcd for $C_{49}H_{61}N_3O_6S_2$: C, 49.28; H, 5.15; N, 3.52%.

1,1′-Didodecylbenzothiazolium carbocyanine bis(perfluorobutylsulfonyl)imide (47). Yield 61%; ^{1}H NMR (CDCl$_3$) δ = 0.86 (t, J = 6.9 Hz, 6H), 1.23–1.41 (m, 36H), 1.78 (quin, J = 6.9 Hz, 4H), 4.18 (t, J = 6.9 Hz, 4H), 6.58 (d, J = 12.8 Hz, 2H), 7.21 (d, J = 7.8 Hz, 2H), 7.31 (t, J = 7.8 Hz, 2H), 7.44 (t, J = 7.8 Hz, 2H), 7.64 (d, J = 7.8 Hz, 2H), 7.73 (t, J = 12.8 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 14.1 (2C), 22.7 (2C), 26.6 (2C), 27.8 (2C), 29.3 (2C), 29.4 (2C), 29.4 (2C), 29.6 (2C), 29.6 (4C), 31.9 (2C), 46.9 (2C), 99.5 (2C), 112.8 (2C), 122.4 (2C), 125.2 (2C), 125.4 (2C), 128.0 (2C), 141.4 (2C), 147.4, 165.0 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.8 (4F), −120.9 (4F), −112.7 (4F), −80.7 (6F); FABMS (NBA) m/z 646 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 47.81; H, 5.42; N, 3.36%. Calcd for $C_{49}H_{61}F_{18}N_3O_4S_4$: C, 47.99; H, 5.01; N, 3.73%.

1,1′-Didodecylquinaldinium carbocyanine bis(perfluorobutylsulfonyl)imide (48). Yield 43%; ^{1}H NMR (CDCl$_3$) δ = 0.87 (t, J = 6.6 Hz, 6H), 1.26–1.42 (m, 32H), 1.53–1.60 (m, 4H), 1.80 (br s, 4H), 4.23 (br s, 4H), 6.19 (d, J = 12.8 Hz, 2H), 7.20 (t, J = 7.9 Hz, 2H), 7.30 (d, J = 7.9 Hz, 2H), 7.43–7.51 (m, 4H), 7.68 (d, J = 9.6 Hz, 2H), 8.00 (d, J = 9.6 Hz, 2H), 8.41 (t, J = 12.8 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 14.1 (2C), 22.7 (2C), 26.6 (2C), 27.2 (2C), 29.4 (2C), 29.4 (2C), 29.5 (2C), 29.6 (4C), 29.7 (2C), 31.9 (2C), 47.8 (2C), 104.7 (2C), 115.1 (2C), 120.1 (2C), 124.7 (2C), 125.0 (2C), 129.2 (2C), 131.8 (2C), 135.9 (2C), 138.7 (2C), 147.0, 152.5 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.8 (4F), −120.8 (4F), −112.7 (4F), −80.6 (6F); FABMS (NBA) m/z 634 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 52.76; H, 5.76; N, 3.43%. Calcd for $C_{53}H_{65}N_3O_4S_2$: C, 52.43; H, 5.40; N, 3.46%.

1,1′-Didodecyllepidinium carbocyanine bis(perfluorobutylsulfonyl)imide (49). Yield 63%; ^{1}H NMR (CDCl$_3$) δ = 0.87 (t, J = 7.1 Hz, 6H), 1.23–1.36 (m, 36H), 1.81 (quin, J = 7.1 Hz, 4H), 4.09 (t, J = 7.1 Hz, 4H), 6.79 (d, J = 13.3 Hz, 2H), 7.43–7.48 (m, 6H), 7.59 (d, J = 7.9 Hz, 2H), 7.67 (t, J = 7.9 Hz, 2H), 8.19 (d, J = 7.9 Hz, 2H), 8.40 (t, J = 13.3 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 14.1 (2C), 22.7 (2C), 26.6 (2C), 29.0 (2C), 29.2 (2C), 29.3 (2C), 29.4 (2C), 29.5 (2C), 29.6 (4C), 31.9 (2C), 54.4 (2C), 109.2 (2C), 110.3 (2C), 116.1 (2C), 124.8 (2C), 125.3 (2C), 125.5 (2C), 132.3 (2C), 138.1 (2C), 140.1 (2C), 142.5, 159.2 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.7 (4F), −120.8 (4F), −112.6 (4F), −80.6 (6F); FABMS (NBA) m/z 634 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 52.73; H, 5.50; N, 3.38%. Calcd for $C_{53}H_{65}F_{18}N_3O_4S_2$: C, 52.43; H, 5.40; N, 3.46%.

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)benzoxazolium carbocyanine bis(perfluorobutylsulfonyl)imide (50). Yield 83%; ^{1}H NMR (CDCl$_3$) δ = 3.33 (s, 6H), 3.43–3.45 (m, 4H), 3.49–3.50 (m, 4H), 3.51–3.53 (m, 4H), 3.59–3.60 (m, 4H), 3.91 (t, J = 4.8 Hz, 4H), 4.32 (t, J = 4.8 Hz, 4H), 6.19 (d, J = 13.1 Hz, 2H), 7.31 (t, J = 7.6 Hz, 2H), 7.36 (t, J = 7.6 Hz, 2H), 7.45 (d, J = 7.6 Hz, 2H), 7.45 (d, J = 7.6 Hz, 2H), 8.47 (t, J = 13.1 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 45.0 (2C), 58.9 (2C), 68.3 (2C), 70.3 (2C), 70.4 (2C), 70.6 (2C), 71.8 (2C), 86.2 (2C), 110.3 (2C), 112.0 (2C), 124.9 (2C), 125.8 (2C), 132.3 (2C), 146.7 (2C), 148.1, 162.2 (2C); ^{19}F NMR (CD$_3$OD) δ = −125.8 (4F), −120.9 (4F), −112.7 (4F), −80.7 (6F); FABMS (NBA) m/z 559 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 40.78; H, 3.37; N, 3.42%. Calcd for $C_{39}H_{41}F_{18}N_3O_{12}S_2$: C, 40.74; H, 3.59; N, 3.65%.

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)benzothiazolium carbocyanine bis(perfluorobutylsulfonyl)imide (51). Yield 86%; ^{1}H NMR (CDCl$_3$) δ = 3.33 (s, 6H), 3.41–3.44 (m, 4H), 3.46–3.48 (m, 4H), 3.50–3.52 (m, 4H), 3.56–3.58 (m, 4H), 3.95 (t, J = 4.9 Hz, 4H), 4.48 (t, J = 4.9 Hz, 4H), 6.66 (d, J = 12.4 Hz, 2H), 7.32 (t, J = 7.6 Hz, 2H), 7.45 (t, J = 7.6 Hz, 2H), 7.57 (d, J = 7.6 Hz, 2H), 7.60 (d, J = 7.6 Hz, 2H), 7.73 (t, J = 12.4 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 47.6 (2C), 59.0 (2C), 68.5 (2C), 70.3 (2C), 70.5 (2C), 70.8 (2C), 71.9 (2C), 100.0 (2C), 114.6 (2C), 121.8 (2C), 124.8 (2C), 125.3 (2C), 127.8 (2C), 142.6 (2C), 147.3, 162.5 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.8 (4F), −120.9 (4F), −112.8 (4F), −80.6 (6F); FABMS (NBA) m/z 601 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 40.01; H, 3.83; N, 3.56%. Calcd for $C_{39}H_{41}F_{18}N_3O_{10}S_4$: C, 39.63; H, 3.50; N, 3.56%.

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)quinaldinium carbocyanine bis(perfluorobutylsulfonyl)imide (52). Yield 54%; ^{1}H NMR (CDCl$_3$) δ = 3.31 (s, 6H), 3.42–3.67 (m, 16H), 3.97 (t, J = 4.9 Hz, 4H), 4.50 (t, J = 4.9 Hz, 4H), 6.28 (d, J = 12.8 Hz, 2H), 7.16 (t, J = 7.3 Hz, 2H) 7.39 (d, J = 7.3 Hz, 2H), 7.43 (t, J = 7.3 Hz, 2H), 7.61 (d, J = 7.3 Hz, 2H), 7.66 (d, J = 9.6 Hz, 2H), 7.91 (d, J = 9.6 Hz, 2H), 8.31 (t, J = 12.8 Hz, 1H);

^{13}C NMR (CDCl$_3$) δ = 47.9 (2C), 58.9 (2C), 68.1 (2C), 70.4 (2C), 70.5 (2C), 71.0 (4C), 71.8 (2C), 105.2 (2C), 116.6 (2C), 119.7 (2C), 124.7 (2C), 124.8 (2C), 128.7 (2C), 131.6 (2C), 136.2 (2C), 139.7 (2C), 146.5, 152.9 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.7 (4F), −120.8 (4F), −112.7 (4F), −80.6 (6F); FABMS (NBA) *m*/*z* 589 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 44.34; H, 4.18; N, 3.45%. Calcd for C$_{43}$H$_{45}$F$_{18}$N$_3$O$_{10}$S$_2$: C, 44.14; H, 3.88; N, 3.59%.

1,1′-Bis(2,5,8-trioxa-10-octadecanyl)lepidinium carbocyanine bis(perfluorobutylsulfonyl)imide (53). Yield 74%; ^{1}H NMR (CDCl$_3$) δ = 3.32 (s, 6H), 3.44–3.58 (m, 16H), 3.84 (t, *J* = 4.8 Hz, 4H), 4.28 (t, *J* = 4.8 Hz, 4H), 6.79 (d, *J* = 12.8 Hz, 2H), 7.28 (d, *J* = 7.8 Hz, 2H) 7.43 (t, *J* = 7.8 Hz, 2H), 7.53 (d, *J* = 7.8 Hz, 2H), 7.55 (d, *J* = 7.8 Hz, 2H), 7.63 (t, *J* = 7.8 Hz, 2H), 8.15 (d, *J* = 7.8 Hz, 2H), 8.26 (t, *J* = 12.8 Hz, 1H); ^{13}C NMR (CDCl$_3$) δ = 53.6 (2C), 58.9 (2C), 68.3 (2C), 70.5 (4C), 70.8 (2C), 71.8 (2C), 108.4 (2C), 110.6 (2C), 116.3 (2C), 124.7 (2C), 125.2 (2C), 125.6 (2C), 132.4 (2C), 138.3 (2C), 140.6 (2C), 142.0, 149.1 (2C); ^{19}F NMR (CDCl$_3$) δ = −125.7 (4F), −120.8 (4F), −112.6 (4F), −80.6 (6F); FABMS (NBA) *m*/*z* 589 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 44.07; H, 4.17; N, 3.55%. Calcd for C$_{43}$H$_{45}$F$_{18}$N$_3$O$_{10}$S$_2$: C, 44.14; H, 3.88; N, 3.59%.

Synthesis of 2-methyl-1-(2,5,8-trioxa-10-octadecanyl)-6-(2,5,8-trioxa-10-octadecanyloxy)quinolinium tosylate (55). To a THF solution (10 mL) of **54** (581 mg, 3.6 mmol) were added 60% sodium hydride (146 mg, 3.7 mmol) and **10** (3.5 g, 11.0 mmol). The mixture was refluxed for 3 h. After the reaction was completed, the solvent was removed *in vacuo*. To the resulting residue were added water (50 mL) and dichloromethane (50 mL). The organic layer was dried over anhydrous sodium sulfate. Dichloromethane was removed *in vacuo*. This crude 2-methyl-6-(2,5-8-trioxa-10-octadecanyloxy)quinolone was dissolved in toluene (10 mL). To this solution was added **10** (3.5 g, 11.0 mmol). The mixture was refluxed overnight. After the reaction was completed, the mixture was poured into ether. The resulting precipitate was filtered and purified by column chromatography (SiO$_2$, CH$_3$Cl : MeOH = 5 : 1). Yield 21%; ^{1}H NMR (CDCl$_3$) δ = 2.32 (s, 3H), 3.12 (s, 3H), 3.31 (s, 3H), 3.37 (s, 3H), 3.40–3.42 (m, 8H), 3.55–3.58 (m, 2H), 3.66–3.68 (m, 2H), 3.69–3.72 (m, 2H), 3.75–3.77 (m, 2H), 3.90 (t, *J* = 4.6 Hz, 2H), 4.04 (t, *J* = 4.6 Hz, 2H), 4.27 (t, *J* = 4.6 Hz, 2H), 5.34 (s, 2H), 7.11 (d, *J* = 8.0 Hz, 2H), 7.51–7.55 (m, 2H), 7.71 (d, *J* = 8.2 Hz, 1H), 7.78 (d, *J* = 8.0 Hz, 2H), 8.36 (d, *J* = 8.2 Hz, 1H), 8.86 (d, *J* = 8.2 Hz, 1H); FABMS (NBA) *m*/*z* 452 (M$^+$ − Tos).

Synthesis of 1,1′-bis(2,5,8-trioxa-10-octadecanyl)-6,6′-bis(2,5,8-trioxa-10-octadecanyloxy)quinaldinium carbocyanine tosylate (56). To toluene (5 mL) were added **55** (500 mg, 0.8 mmol) and triethyl orthoformate (70 mg, 0.5 mmol). The mixture was refluxed overnight. After cooling, toluene was removed *in vacuo*. The product was extracted with dichloromethane (50 mL × 3). The extract was dried over anhydrous sodium sulfate. After removing the solvent *in vacuo*, the product was purified by chromatography (SiO$_2$, CHCl$_3$: MeOH = 5 : 1). No molecular ion peak of this compound was observed by the FABMS method. Yield 35%; mp 66 °C; ^{1}H NMR (CDCl$_3$) δ = 2.34 (s, 3H), 3.32 (s, 6H), 3.39 (s, 6H), 3.44–3.45 (m, 4H), 3.50–3.58 (m, 16H), 3.66–3.68 (m, 4H), 3.71–3.72 (m, 4H), 3.75–3.77 (m, 4H), 3.86 (t, *J* = 4.6 Hz, 4H), 3.95 (br s, 4H), 4.06 (t, *J* = 4.6 Hz, 4H), 4.49 (br s, 4H), 6.38 (d, *J* = 12.8 Hz, 2H), 6.72 (d, *J* = 3.2 Hz, 2H), 7.00 (dd, *J* = 9.6 and 3.2 Hz, 2H), 7.22 (d, *J* = 8.2 Hz, 2H), 7.49 (d, *J* = 10.1 Hz, 2H), 7.53 (d, *J* = 9.6 Hz, 2H), 7.97 (d, *J* = 8.2 Hz, 2H), 8.30 (d, *J* = 10.1 Hz, 2H), 8.47 (t, *J* = 12.8 Hz, 1H); FABMS (NBA) *m*/*z* 914 (M$^+$ − Tos).

Synthesis of 1,1′-bis(2,5,8-trioxa-10-octadecanyl)-6,6′-bis(2,5,8-trioxa-10-octadecanyloxy)quinaldinium carbocyanine bis(perfluorobutylsulfonyl)imide (57). To a 1 : 1 acetone–chloroform mixed solution (10 mL) of **56** (150 mg, 0.14 mmol) was added **38** (120 mg, 0.21 mmol). The mixture was stirred overnight. After the reaction was completed, to the residue were added water (30 mL) and dichloromethane (30 mL). The organic layer was dried over anhydrous sodium sulfate. After removing the organic layer *in vacuo*, the product was dissolved in dichloromethane, and purified by column chromatography (SiO$_2$, CHCl$_3$: MeOH = 10 : 1). Yield 45%; ^{1}H NMR (CD$_3$OD) δ = 3.28 (s, 6H), 3.35 (s, 6H), 3.44 (t, *J* = 4.5 Hz, 4H), 3.52–3.54 (m, 8H), 3.59–3.60 (m, 4H), 3.63–3.68 (m, 12H), 3.72–3.73 (m, 4H), 3.88 (t, *J* = 4.5 Hz, 4H), 4.00 (t, *J* = 4.5 Hz, 4H), 4.20 (t, *J* = 4.5 Hz, 4H), 4.63 (br s, 4H), 6.43 (d, *J* = 12.5 Hz, 2H), 7.17 (d, *J* = 2.8 Hz, 2H), 7.28 (dd, *J* = 9.6 and 2.8 Hz, 2H), 7.74 (d, *J* = 9.6 Hz, 2H), 7.81 (d, *J* = 9.6 Hz, 2H), 7.98 (d, *J* = 9.6 Hz, 2H), 8.42 (t, *J* = 12.5 Hz, 1H); ^{19}F NMR (CD$_3$OD) δ = −125.1 (4F), −122.0 (4F), −114.1 (4F), −82.3 (6F); FABMS (NBA) *m*/*z* 914 (M$^+$ − (C$_4$F$_9$SO$_2$)$_2$N); anal. found: C, 45.93; H, 4.74; N, 3.1%. Calcd for C$_{57}$H$_{73}$F$_{18}$N$_3$O$_{18}$S$_2$: C, 45.81; H, 4.92; N, 2.81%.

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