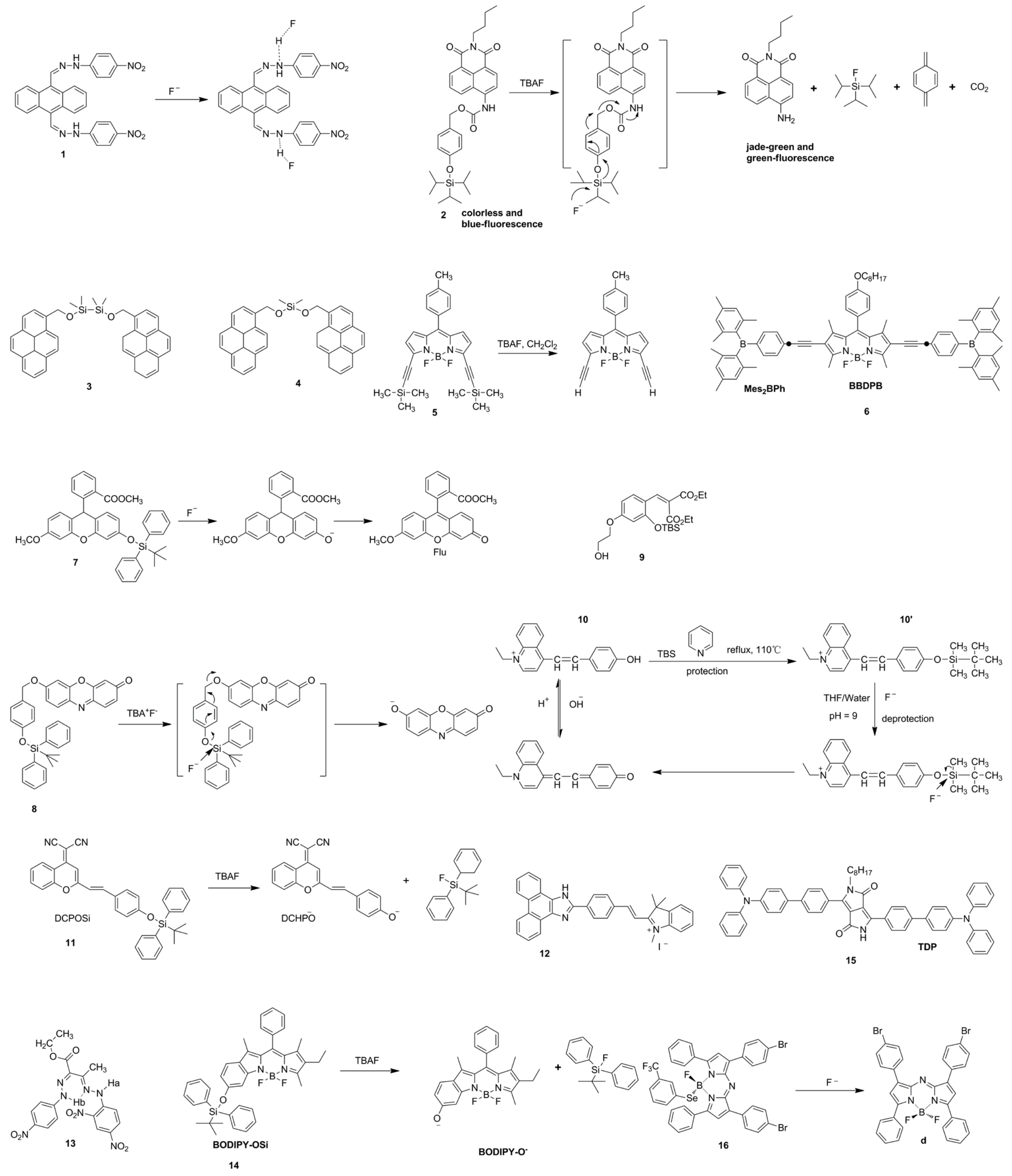
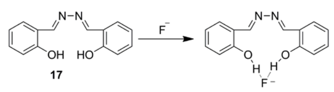
Supplementary materials



S Fig. 1



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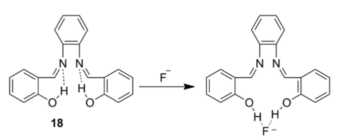

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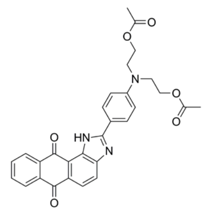

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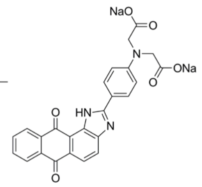

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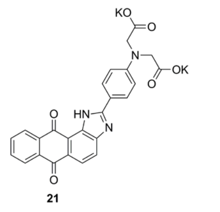

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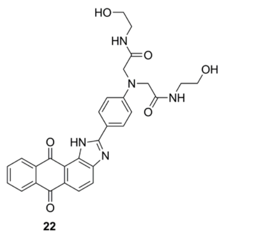

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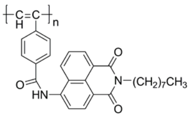
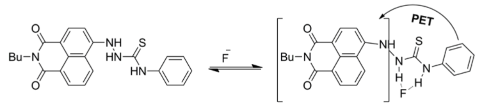
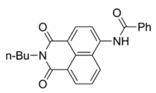




High Fluorescence

24

Weak Fluorescence



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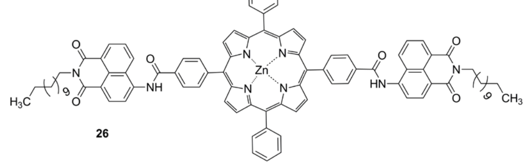

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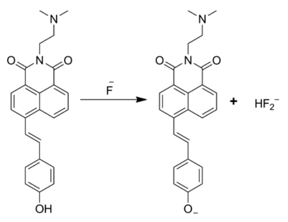

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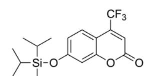

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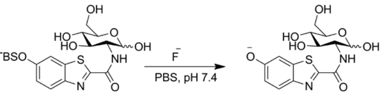

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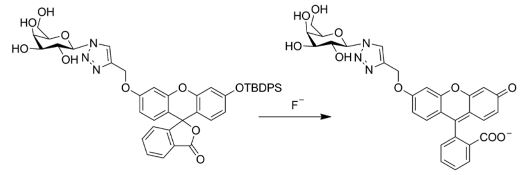
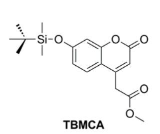


non-fluorescent

30

fluorescent



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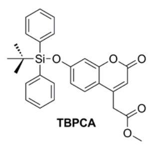

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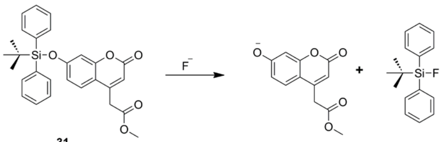

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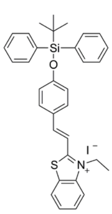

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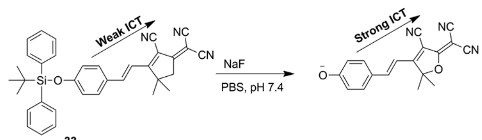

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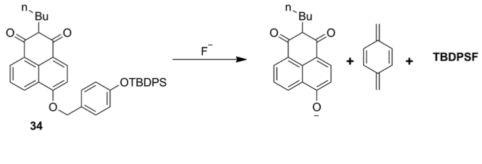

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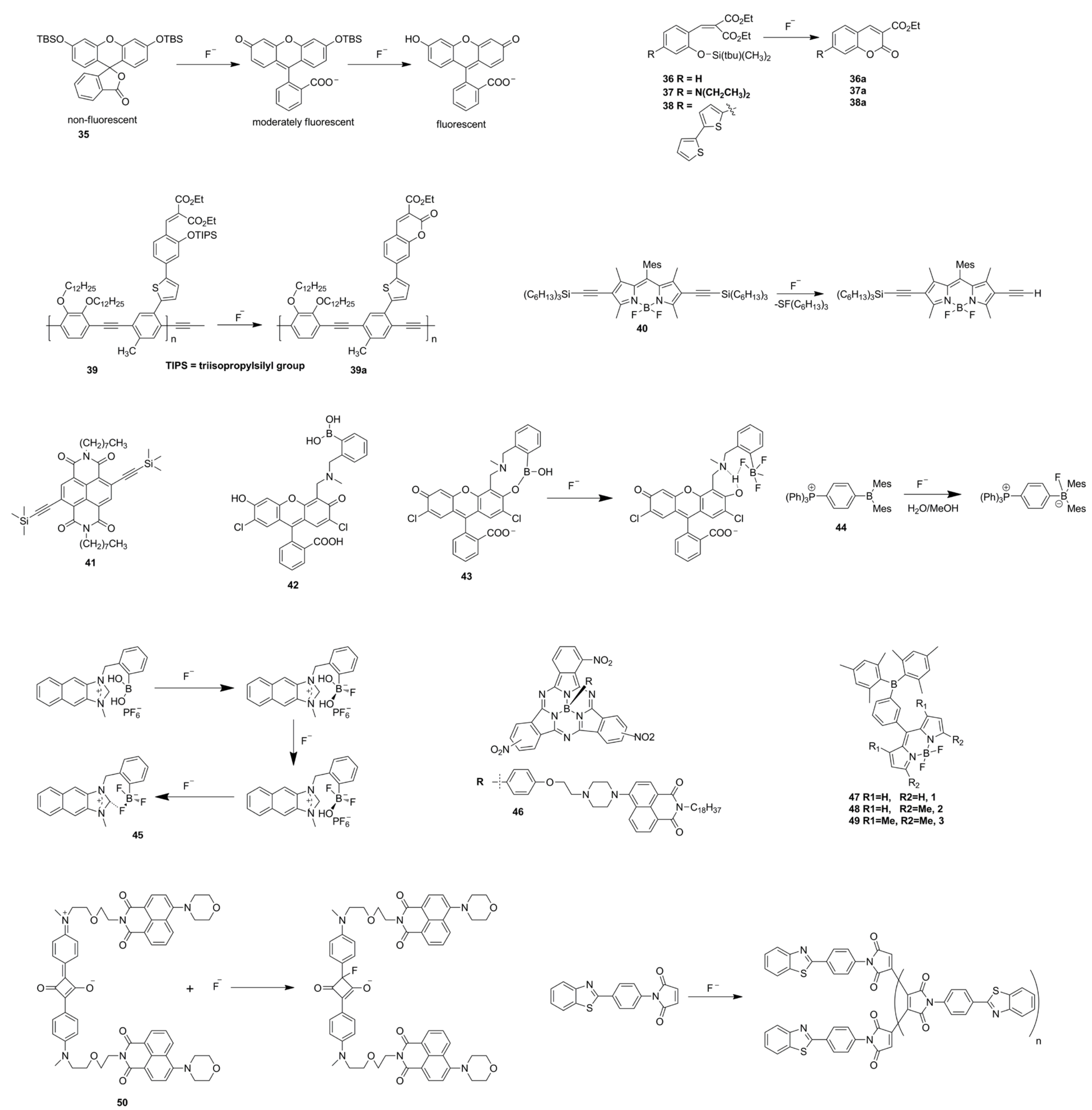
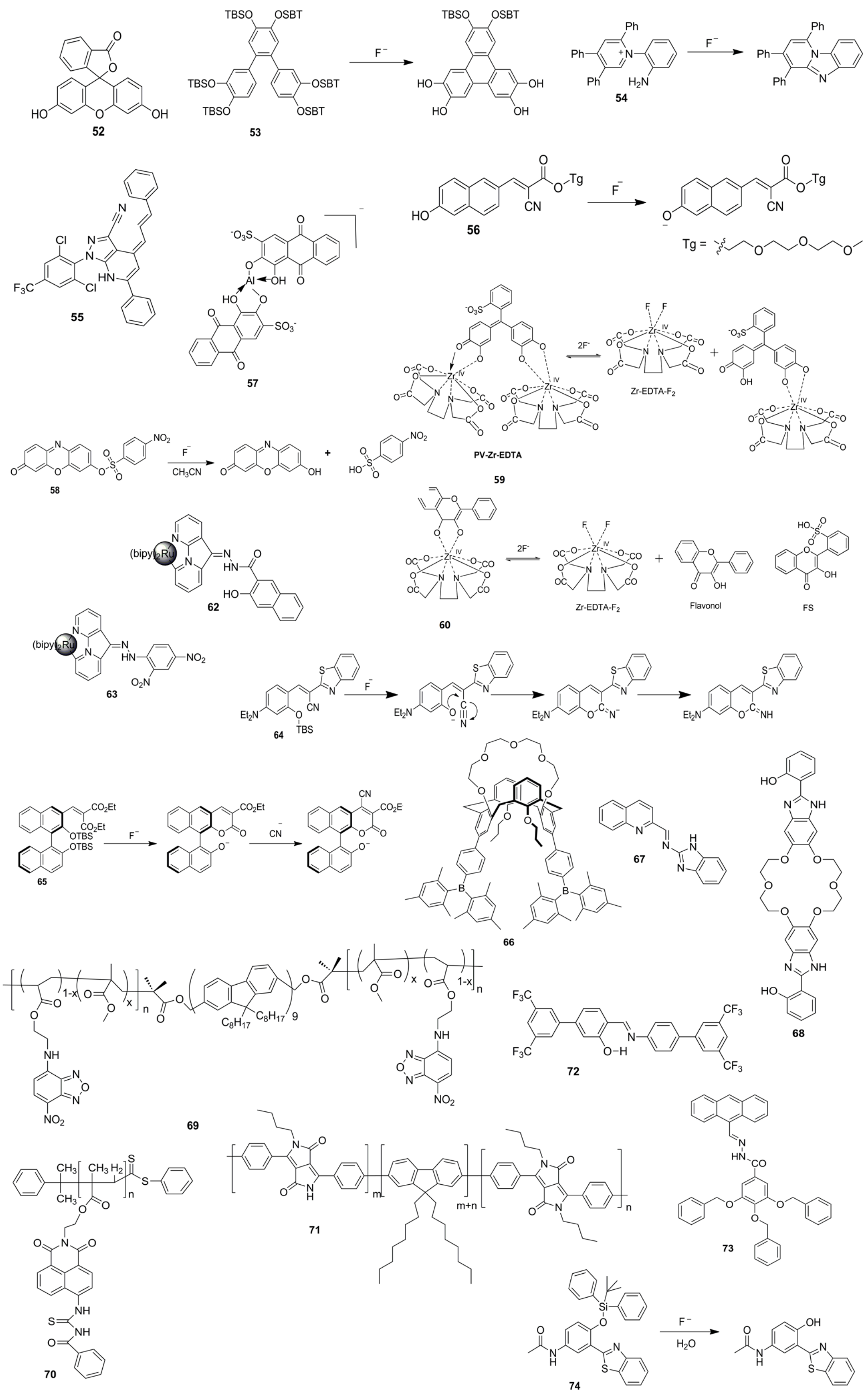
S Fig. 2


non-fluorescent
35
moderately fluorescent
fluorescent
36 R = H
37 R = N(CH2CH3)2
38 R =
36a
37a
38a
39
TIPS = triisopropylsilyl group
39a
40
41
42
43
44
H2O/MeOH
45
46
47 R1=H, R2=H, 1
48 R1=H, R2=Me, 2
49 R1=Me, R2=Me, 3
50

S Fig. 3

S Fig. 4



Supplementary Figure 1~4 Chemical formulas of the fluoride ***Probe1~Probe 74*** (except for ***Probe 61***)

Supplementary Table 1 Comparison of three conventional analytical methods for F^-

	Ion Chromatography	Ion Selective Electrode	Spectrophotometry
Principle	separating ions and polar molecules based on their charge differences	electrochemical reactions of the specific ions	measuring the absorbance or luminescence intensity of the light
Detecting Range of Fluoride (μg/L)	$3.8 \sim 2.0\times10^3$	$0.041 \sim 1.9\times10^6$	$0 \sim 1.0\times10^6$
Advantages	good sensitivity, wide linearity range, fast, simple, accurate, producible	most widely used, cheaper, fast, efficient, simple	stable, simple, fast, amenable to automation
Disadvantages	weak binding between fluoride and ion exchangers	active surface of the electrode requires periodic regeneration, electrode drift and dissolution of the lanthanum fluoride membrane crystal	quantitative spectral overlap caused by the coexistence of spectral interference
Detecting Targets	coffee, tea, spinach, fruits, alcoholic beverages, milk, mulberries, leaves, soil, toothpaste, and plaque biofilm	rain water, vegetables, fruit, tea, mulberries, medicinal materials, toothpaste, human blood, urine, pork, salt, air, cigarette smoke, and coal	mulberry leaves, toothpaste, minerals, ground water, and drinking water
Ref.	Suppl [1-16]	Suppl [4, 16-26]	Suppl [4, 15, 27-32]

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Supplementary Table 2 Spectroscopic and analytical parameters of fluorescent fluoride probes

Fluorescent probe	Quantum yield	Reaction medium	λ_{ex}(nm) / λ_{em} (nm)	Liner detection range / (LOD) (μM)	Selectivity	Mechanism	Analytical applications	Ref.
1	0.76	DMSO	520 / 623	2.0-1.2×10^{2} / 2.0	AcO^-, $H_2PO_4^-$, Cl^-, Br^-, I^-	ICT PET	toothpaste	[20]
2	0.41	CH_3CN	390 / 449	NM	Cl^-, Br^-, I^-, HSO_4^-, NO_3^-, PF_4^-, ClO_4^-, AcO^-, $H_2PO_4^-$	ICT	NM	[25]
3	NM	THF/H_2O (50/50, v/v)	335 / 470-480	NM	Cl^-, Br^-, I^-, CO_3^{2-}, NO_2^-, NO_3^-, SCN^-, ClO_4^-, SO_4^{2-}, HPO_4^{2-}, $H_2PO_4^-$	NM	NM	[27]
4	NM	THF/ H_2O (50/50, v/v)	335 / 470-480	NM	Cl^-, Br^-, I^-, CO_3^{2-}, NO_3^-, NO_2^-, SCN^-, ClO_4^-, SO_4^{2-}, HPO_4^{2-}, $H_2PO_4^-$	NM	HeLa cells	[27]
5	0.29	CH_2Cl_2	425 / 584,564	NM	Br^-, Cl^-, I^-, HPO_4^{2-}, ClO_4^-	NM	NM	[30]
6	0.65	CH_2Cl_2	430 / 602	NM / 2.0×10^{-1}	Br^-, Cl^-, I^-, $H_2PO_4^-$, CH_3COO^-, NO_3^-, ClO_4^-	PET	NM	[31]
7	0.55	THF/H_2O (1/9,v/v)	NM / 520	NM / 18.0	AcO^-, NO_2^-, NO_3^-, SO_3^{2-}, CO_3^{2-}, SO_4^{2-}, PO_4^{3-}, Cl^-, IO_3^-, ClO_3^-, Br^-, HPO_4^{2-}, HSO_3^-, S^{2-}, $S_2O_3^{2-}$, I^-, SCN^-, CN^-,	NM	HeLa cells	[34]
8	NM	CH_3CN/H_2O (50/50, v/v)	550 / 585	NM	Cl^-, Br^-, I^-, HSO_4^-, NO^-, N^-	NM	NM	[37]
9	0.86	THF	365 / 402	NM / 0-15.0 ppm (unimers), 0-8.0 ppm(micelles)	Cl^-, Br^-, I^-, CO_3^{2-}, NO_3^-, HSO_4^-, HPO_4^{2-}, $H_2PO_4^-$, AcO^-,	NM	aqueous media	[38]
10	0.98	THF/water (7/3, v/v)	NM	NM / 1.0×10^{-1}	Cl^-, Br^-, I^-, SO^{2-}, SCN^-, PO_3^-	NM	water samples	[40]
11	0.43	DMSO/H_2O (95/5, v/v)	645 / 718	NM / 8.5×10^{-2}	Cl^-, Br^-, I^-, SO^{2-}, NO^-, N^-, AcO^-, HPO^{2-}	NM	NM	[42]
12	0.12	DCM	555 / 738	NM	Cl^-, Br^-, I^-	NM	NM	[43]
13	NM	DMSO	NM / 675	2.0-4.0 / 2.59×10^{-1}	CH_3COO^-, $C_6H_5COO^-$, $H_2PO_4^-$, Cl^-, Br^-, I^-, HSO_4^-, ClO_4^-, BF_4^-, PF_6^-	NM	NM	[44]
14	NM	CH_2Cl_2	644 / 676	NM / 1.18×10^{-1}	Cl^-, Br^-, I^-, HSO_4^{2-}, HPO_4^{2-}, $H_2PO_4^-$, N_3^-, NO_3^{2-}	NM	NM	[45]
15	0.27	THF	800 / 534, 594,645	NM	Cl^-, Br^-, I^-	NM	NM	[46]
16	NM	PBS buffer solution	660 / 690	0.10-1.0 / 7.4×10^{-2}	$C_2O_4^{2-}$, NO_2^-, NO_3^-, I^-, HCO_3^-, Br^-, CO_3^{2-}, Cl^-, CN^-, SO_4^{2-}	PET	HepG2 cells	[47]
17	0.85	DMSO	396 / 520	NM	Cl^-, Br^-, I^-, AcO^-, ClO_4^-, HSO_4^-, $H_2PO_4^-$	NM	NM	[49]
18	0.82	DMSO	390 /	NM	NM		NM	[49]

			420					
19	0.8	CH_3CN/DMSO (5/95, v/v)	360 / 430	NM	Cl^-, Br^-, I^-, AcO^-, $H_2PO_4^-$, NO_3^-, ClO_4^-, HSO_4^-	ICT	aqueous medium	[51]
20	0.9	CH_3CN/DMSO (5/95, v/v)	360 / 430	NM	Cl^-, Br^-, I^-, AcO^-, $H_2PO_4^-$, NO_3^-, CN^-, ClO_4^-, HSO_4^-	ICT	aqueous medium	[50]
21	0.8	CH_3CN/DMSO (5/95, v/v)	360 / 430	NM	Cl^-, Br^-, I^-, AcO^-, $H_2PO_4^-$, NO_3^-, CN^-, ClO_4^-, HSO_4^-	ICT	aqueous medium	[50]
22	0.8	CH_3CN/DMSO (5/95, v/v)	360 / 430	NM	Cl^-, Br^-, I^-, AcO^-, $H_2PO_4^-$, NO_3^-, ClO_4^-, HSO_4^-	ICT	aqueous medium	[50]
23	0.89	CH_3CN	360 / 460	NM / 10.0	Cl^-, Br^-, I^-	ICT	NM	[51]
24		CH_3CN	390 / 450,650	NM	Cl^-, Br^-, I^-	ICT, PET	NM	[52]
25	0.75	CH_3CN	340 / 468,583	2.0×10^4-1.0×10^5 / NM	Cl^-, Br^-, I^-	ICT	NM	[53]
26	0.037→0.026	THF	365 / 608 (655)	NM	Cl^-, Br^-, I^-	ICT, PET, FRET	NM	[54]
27	NM	DMSO	490 / 582	NM	Cl^-, Br^-, I^-, NO_3^-, $H_2PO_4^-$, AcO^-, SO_4^{2-}, BF_4^-, CN^-, OH^-	ICT	RAW 264.7 cells, 4T1 cells, HeLa cells	[56]
28	0.67	CH_3CN	410 / 500	NM	Cl^-, Br^-, I^-, CN^-, AcO^-, NO_3^-, $PhCO_2^-$, SCN^-, $H_2PO_4^-$, HSO_4^-	NM	aqueous medium	[57]
29	NM	DMSO/PBS (1/199, v/v)	380 / 508	1.0×10^2-1.0×10^3 / NM	Cl^-, Br^-, I^-, $H_2PO_4^-$, N_3^-, NO_2^-, NO_3^-, AcO^-, SO_4^{2-}	NM	KB cells	[58]
30	0.86	CTAB/PBS (1/10, v/v)	470 / 520	NM / 10.5	Cl^-, $H_2PO_4^-$, I^-, SO_4^{2-}, NO_3^-, CO_3^{2-}, N_3^-, HPO_4^{2-} , CN^-, HCO_3^-, AcO^-, Br^-	NM	water samples, HepG2 cells	[59]
31	NM	HEPES	365 / 461	NM / 1.86×10^{-5}	Cl^-, Br^-, I^-, AcO^-, NO_3^-, N_3^-, $H_2PO_4^-$	ICT	A549 cells	[60]
32	NM	C_2H_5OH/H_2O (7/3, v/v)	540-580 / 470-510	5.0×10^2-2.8×10^4 / 80.0	CO_3^{2-}, SO_4^{2-}, SCN^-, NO_3^-, N_3^-, Cl^-, Br^-, I^-, Cys, GSH, BSA, HSA	ICT	RAW 264.7 cells	[61]
33	0.58	C_2H_5OH/H_2O (7/3, v/v)	560 / 612	0-6.0×10^3 / 70.0	Cl^-, Br^-, I^-, SCN^-, N_3^-, NO_3^-, HCO_3^-	ICT	HeLa cells	[62]
34	NM	CH_3CN/H_2O (50/50, v/v)	475 / 560	0-7.2×10^2 / 0.35 mg L^{-1}	Cl^-, Br^-, I^-, CN^-, AcO^-, $H_2PO_4^-$, NO_3^-, SO_4^{2-}	NM	A549 cells, toothpaste	[63]
35	0.676	DMF/H_2O (7/3, v/v)	480 / 532	0.1-2.0 / 4.1×10^{-2}	Cl^-, Br^-, I^-	NM	toothpaste	[64]
36	0.0001	NM	322 / NM	NM	NM	FRET	NM	[65]
37	0.002	THF	386 / 450	NM	NM	FRET	NM	[65]
38	0.008	NM	384 / 489	NM	NM	FRET	NM	[65]
39	0.12	THF	378 / 482	NM	NM	FRET	NM	[65]
40	0.71	C_3H_6O	400 / 555	NM / 6.74×10^{-2}	Cl^-, Br^-, I^-, ClO_4^-, $H_2PO_4^-$, CH_3COO^-, SCN^-, CO_3^{2-}, NO_3^-,	NM	NM	[66]

					SO_4^{2-}			
41	0.98	CH_2Cl_2	289 / 455	NM	Cl^-, Br^-, HSO_4^-, AcO^-, $H_2PO_4^-$	NM	NM	[67]
42	0.63	C_2H_3N/CH_3OH (9/1, v/v)	483 / 522	NM	Cl^-, Br^-, I^-	PET	NM	[33]
43	0.6	THF	324 / 531	NM	NM	NM	NM	[68]
44	0.63	H_2O/CH_3OH (9/1, v/v)	NM	NM	Cl^-, Br^-, I^-, NO^-, H PO^-, HSO_4^-	NM	NM	[69]
45	NM	CH_3CN	323 / 440	NM	Cl^-, Br^-, CH_3COO^-, HSO_4^-, $H_2PO_4^-$	NM	aqueous medium	[70]
46	NM	THF	390 / 495, 500 / 578/	NM	Cl^-, Br^-, I^-, ClO_4^-	NM	NM	[71]
47	0.48	DCM	350 / 408, 519	NM / 1.2 ppm	Cl^-, Br^-, I^-, ClO_4^-, $H_2PO_4^-$, HSO_4^-, NO_3^-, PF_6^-, CN^-	FRET	NM	[72]
48	0.55	DCM	350 / 405, 450, 491, 526 408, 515	NM / 2.3 ppm	Cl^-, Br^-, I^-, ClO_4^-, $H_2PO_4^-$, HSO_4^-, NO_3^-, PF_6^-, CN^-	FRET	NM	[72]
49	0.72	DCM	350 / 408, 519	NM / 0.2 ppm	Cl^-, Br^-, I^-, ClO_4^-, $H_2PO_4^-$, HSO_4^-, NO_3^-, PF_6^-, CN^-	FRET	NM	[72]
50	0.059	CH_2Cl_2	402 / 659	NM / 2.0×10^2	Br^-, I^-, OH^-, PO_4^{3-}, SCN^-, CH_3COO^-, ClO_4^-, NO_3^-, $C_2O_4^{2-}$, SO_4^{2-}, S^{2-}, CN^-	FRET， PET	NM	[73]
51	0.89	DMSO	365 / 412	NM / 5.0×10^4	CN^-， Cl^-， OH^-, Br^-, I^-, PF_6^-, BF_4^-, HSO_4^-, OAc^-	NM	NM	[74]
52	0.61	CH_3CN	480 / 532	NM	Cl^-, Br^-, I^-, HSO_4^-	NM	NM	[75]
53	0.86	THF	294 / 393	NM / 2.0	Cl^-, Br^-, I^-, CN^-, HSO^-, $H_2PO_4^-$, NO^-, OAc^-	NM	NM	[76]
54	0.46	CH_3CN	366 / 469	NM / 2.72	Cl^-, Br^-, I^-, NO_3^-, $H_2PO_4^-$, AcO^-	NM	NM	[77]
55	0.15	CH_2Cl_2	324 / 400	NM / 8.54×10^{-3}	Cl^-, Br^-, I^-, NO_3^-, HSO_4^-	NM	NM	[78]
56	0.94	PBS buffer	388 / 490	NM	$NO3^-$, Br^-, Cl^-, HSO_4^-, CO_3^{2-}, S^{2-}, I^-, N_3^-, PO_4^-, SCN^-, CH_3COO^-	NM	PC3 cells	[79]
57	NM	C_2H_5OH/H_2O (1/5, v/v)	247 / 532	6.0×10^5-8.0×10^7 / 6.0×10^5	Cl^-, SO_4^{2-}, NO_3^-, PO_4^{3-}, I^-, Br^-	NM	NM	[80]
58	0.58	CH_3CN	485 / 550, 591	NM / 1.9	Cl^-, Br^-, I^-, AcO^-, NO_3^-, N_3^-, ClO_4^-, HSO_4^-	NM	NM	[81]
59	NM	aqueous solution		15.0-1.5×10^2 / 4.5234×10^2	SO_4^{2-}, SO_3^{2-}, CO_3^{2-}, Cl^-, NO_3^-, NO_2^-, PO_4^{3-}	NM	NM	[83]
60	NM	aqueous	377 /	0.5-1.0×10^3 /	Cl^- , Br^-, NO_3^-,	NM	toothpaste,	[84]

		solution	460	NM	CH_3COO^-, SO_4^{2-}, $H_2PO_4^-$		water samples	
61	NM	HAc/NaAc buffer solution	NM / 562.4, 519.9	0.10-10.0 / 0.031	Cl^-, I^-, Br^-, NO_3^-, $H_2PO_4^-$, HCO_3^-, HPO_4^{2-}	NM	toothpaste, tap water samples	[85]
62	NM	CH_3CN	355 / 530	NM	Cl^-, I^-, Br^-, HSO_4^-, NO_3^-, $H_2PO_4^-$	PET, ESIPT	NM	[86]
63	0.6	CH_3CN	465 / 610	NM	HSO_4^-, NO_3^-, Cl^-, Br^-, I^-	PET	aqueous medium	[87]
64	0.0008 →0.0709	C_2H_5OH/H_2O (1/1, v/v)	460 / 523	NM / 1.177×10^{-1}	Cl^-, Br^-, I^-, AcO^-, ClO_4^-, $CF_3SO_3^-$, NO_3^-, HSO_4^-, $H_2PO_4^-$, BF_4^-, N_3^-, SCN^-, OH^-	ACRR	NM	[88]
65	0.009	THF	330 / 480	5.0-45.0 / 1.86	Cl^-, Br^-, I^-, CN^-, AcO^-, ClO_4^-, $CF_3SO_3^-$, NO_3^-, HSO_4^-, $H_2PO_4^-$, BF_4^-, N_3^-, SCN^-, $HSCH_2COOH$	ARR	NM	[89]
66	0.70	CH_2Cl_2	342 / 430	NM	Cl^-, Br^-, I^-, $H_2PO_4^-$, AcO^-	NM	NM	[90]
67	0.43	CH_3CN/H_2O (1/4, v/v)	405 / 472	NM	Cl^-, Br^-, I^-, CN^-, $H_2PO_4^-$, NO_3^-, NO_2^-, SO_4^{2-}, HSO_3^-, S^{2-}	NM	HeLa cells	[92]
68	0.62→ 0.57	CH_3CN	330 / 455	NM / 4.0×10^{-2}	$H_2PO_4^-$, AcO^-, NO_3^-, Cl^-, Br^-, I^-	ESIPT	HeLa cells	[93]
69	0.83	C_3H_6O	390 / 515	NM / 4.78	Cl^-, Br^-, I^-, CH_3COO^-, ClO_4^-, HSO_4^-, $H_2PO_4^-$	FRET	NM	[95]
70	0.93	CH_2Cl_2/DMSO (9/1, v/v)	340 / 468/, 490 / 580	NM	Cl^-, Br^-, I^-	ICT	NM	[99]
71	0.98	THF	532 / 570	1.5 μg/mL-15.0 μg/mL / NM	AcO^-, $H_2PO_4^-$	FRET, ICT	KB cells	[102]
72	0.82	THF	405 / 588	NM	Cl^-, Br^-, I^-, NO_3^-, $H_2PO_4^-$, AcO^-	ESIPT	NM	[107]
73	NM	THF	470 / 490→ 505	NM	Cl^-, Br^-, I^-, ClO_4^-, CH_3COO^-, $H_2PO_4^-$, HSO_4^-	NM	NM	[108]
74	0.52	aqueous solution	360 / 468	NM	anions (sodium salts) and cations (nitrate salts)	ESIPT	water samples	[109]

ACRR=anion to cation relay recognition
ARR=anion relay recognition
A549 cells=A549 human lung carcinoma cell lines
BSA=bovine serum albumin
CTAB=cetyl trimethyl ammonium bromide
Cys=cysteine
DCM=Dichloromethane
DMF=N,N-Dimethylformamide
DMSO=(dimethyl sulfoxide
FRET=fluorescence resonance energy transfer

GSH=glutathione
HeLa cells =human cervical cancer cell line cells
HepG2 cells=Human hepatocellular liver carcinoma cells
HAS=human serum albumin
ICT=internal charge transfer
LOD=limit of detection
NM=not mentioned
PBS=phosphate buffered saline
PET=photoinduced electron transfer
RAW 264.7 cells =mouse monocyte macrophage cell line cells
TBA=tetrabutylammonium
THF=Tetrahydrofuran
4T1 cells =mouse breast cancer cell line cells