Supporting Information

Tuning Solid-State Fluorescence to the Near-Infrared: A Combinatorial Approach to Discover Molecular Nanoprobes for Biomedical Imaging

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- Characterization Data for ArV Compounds -

2-(4-bromophenyl)-3-(9-ethyl-9H-carbazol-3-yl)acrylonitrile (CbV1)

Pale-yellow solid. Yield: 71 %. FT-IR cm^{-1}(KBr), 1622 (aromatic C=C), 2208 (CN), 2968 (aliphatic C-H) and 3046 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.46(3H, t, -CH3, J = 7.2), 4.4 (2H, q, -CH2, J = 7.2), 7.30 (1H, t, CbPhH, J = 7.2), 7.42-7.56 (8H, m, CbPhH & COPhH), 7.68 (1H, s, CH), 8.15 (1H, d, CbPhH, J = 7.5), 8.63 (1H, s, CbPhH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 143.92, 141.37, 140.66, 134.39, 132.20, 127.28, 127.13, 126.66, 124.63, 123.49, 122.96, 122.86, 122.58, 120.91, 120.03, 118.89, 109.10, 108.99, 106.22, 37.95, 13.99 ppm. Elem. Anal. Calcd for $C_{23}H_{17}BrN_2$: C, 68.84; H, 4.27; N, 6.98, Found: C, 68.63; H, 4.16; N, 6.96 %. MS (MALDI), calcd for $C_{23}H_{17}BrN_2$, *m/z*= 402.06; found, *m/z*=402.05.

3-(9-ethyl-9H-carbazol-3-yl)-2-(thiophen-2-yl)acrylonitrile (CbV2)

Yellow solid. Yield: 75 %. FT-IR cm^{-1}(KBr), 1628 (aromatic C=C), 2212 (CN), 2977 (aliphatic C-H) and 3096 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.47 (3 H, t, J = 7.2 Hz), 4.40 (2 H, q, J =7.2 Hz), 7.08 (1H, t, J = 7.5 Hz), 7.27-7. 32 (2H, m), 7.37 (1H, d, J = 6.9 Hz), 7.43-7. 54 (3H, m), 7.58 (1H, s), 8.09 (1H, d, J = 8.7 Hz), 8.15 (1H, d, J = 7.8 Hz), 8.61(1H, s). ^{13}C NMR (150 MHz, $CDCl_3$) δ 141.33, 141.21, 140.64, 140.28, 128.13, 126.98, 126.59, 126.10, 125.26, 124.55, 123.48, 122.97, 122.58, 120.92, 119.97, 118.05, 109.06, 109.01, 102.21, 37.93, 13.99 ppm. Elem. Anal. Calcd for $C_{21}H_{16}N_2S$: C, 76.80; H, 4.91; N, 8.53; S, 9.76. Found: C, 76.44; H, 4.89; N, 8.39 ; S, 9.60 %. MS (MALDI), calcd for $C_{21}H_{16}N_2S$, *m/z*=328.10; found, *m/z*=328.11.

2-(biphenyl-4-yl)-3-(9-ethyl-9H-carbazol-3-yl)acrylonitrile (CbV3)

Pale-yellow solid, Yield: 62 %. FT-IR cm^{-1}(KBr), 1612 (aromatic C=C), 2213 (CN), 2908 (aliphatic C-H) and 3038 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.48 (3H, t, -CH3, J = 7.2), 4.41 (2H, q, -CH2, J = 7.2), 7.30 (1H, t, CbPhH, J = 7.5), 7.38 (1H, t, BiPhH, J = 7.2), 7.44-7.55 (6H, m, BiPhH & CbPhH), 7.63-7.68 (3H, m, CbPhH), 7.71 (1H, s, CH), 7.79 (2H, d, CbPhH, J = 8.4), 8.17 (2H, d, BiPhH, J = 8.4), 8.67 (1H, s, CbPhH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 143.29, 141.31, 141.25, 140.65, 140.27, 134.32, 129.04, 127.81, 127.70, 127.31, 127.13, 126.58, 126.20, 124.97, 123.48, 123.02, 122.75, 120.94, 119.97, 119.25, 109.07, 108.96, 107.04, 37.93, 14.0. Elem. Anal. Calcd for $C_{29}H_{22}N_2$: C, 87.41; H, 5.56; N, 7.03. Found: C, 86.27; H, 5.51; N, 6.94 %. MS (MALDI), calcd for $C_{29}H_{22}N_2$, *m/z*= 398.18; found, *m/z*=398.17.

2-Cyano-3-(9-ethylcarbazol-3-yl)acrylic acid (CbV4)

Yellow-green solid, 57 % yield; FT-IR cm^{-1}(KBr), 1624 (aromatic C=C), 1694 (C=O of carboxylic group), 2224 (CN), 2980 (aromatic C-H), 3440 (O-H, carboxylic acid). ^{1}H NMR (300 MHz, DMSO, TMS, ppm): δ 1.35 (3H, t, CH_3, J = 6.9), 4.51 (2H, q, CH_2, J = 6.9), 7.32 (1H, t, J = 7.2), 7.55 (1H, t, J = 7.5), 7.72 (1H, d, J = 8.4), 7.83 (1H, d, J = 8.7), 8.15 (1H, d, J = 7.5) 8.29 (1H, d, J = 8.4), 8.46 (1H, s), 8.86 (1H, s), 13.64 (1H, s, -OH). ^{13}C NMR (150 MHz, DMSO) δ 164.78, 155.79, 142.73, 140.78, 128.33, 127.42, 126.07, 123.13, 122.85, 122.62, 121.09, 120.86, 117.88, 110.51, 98.34, 62.74, 38.03, 14.0. Elem. Anal. Calcd. for $C_{18}H_{14}N_2O_2$: C, 74.47; H, 4.86; N, 9.65; O, 11.02. Found: C, 74.39 %; H, 4.76%; N, 9.52 %. MS (MALDI), calcd for $C_{18}H_{14}N_2O_2$, *m/z*= 290.11; found, *m/z*=290.26.

Ethyl 2-cyano-3-(9-ethyl-9H-carbazol-3-yl)acrylate (CbV5)

Pale-yellow solid, Yield: 58 %. FT-IR cm^{-1}(KBr), 1628 (aromatic C=C), 1722 (C=O, ester), 2218 (CN), 2974 (aliphatic C-H) and 3068 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.39-1.50 (6H, m, -CH3), 4.36-4.44 (4H, m, -CH2), 7.32 (1H, t, Cb-PhH, J = 7.2), 7.46 (1H, t, Cb-PhH, J = 6.9), 7.52-7.57 (2H, m, Cb-PhH), 8.16 (1H, d, Cb-PhH, J = 8.4), 8.22 (1H, d, Cb-PhH, J = 8.7), 8.42 (1H, s, CH), 8.75 (1H, s, Cb-PhH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 163.76, 156.01, 142.89, 140.72, 129.16, 127.05, 125.48, 123.67, 122.72, 121.06, 120.66, 117.08, 109.33, 109.19, 103.3, 97.76, 62.39, 38.05, 14.56, 14.2 ppm. Elem. Anal. Calcd. for $C_{20}H_{18}N_2O_2$: C, 75.45; H, 5.70; N, 8.80. Found: C, 74.94 %; H, 5.61 %; N, 8.66 %. MS (MALDI), calcd for $C_{20}H_{18}N_2O_2$, *m/z*= 318.14; found, *m/z*=318.50.

2-((9-ethyl-9H-carbazol-3-yl)methylene)malononitrile (CbV6)

Yellow solid. Yield 60 %. FT-IR cm^{-1}(KBr), 1626 (aromatic C=C), 2228 (CN), 2980 (aliphatic C-H) and 3056 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.48 (3 H, t, CH_3, J = 7.2 Hz,), 4.41 (2H, q, CH_2, J = 8.0 Hz), 7.36 (1 H, t, PhH, J = 7.2 Hz), 7.47 (2 H, d, PhH, J = 9.3 Hz), 7.57 (1 H, t, PhH, J = 7.2 Hz), 7.86 (1H, s, CH), 8.07–8.15 (2 H, m, PhH), 8.62 (1H, s, PhH) ppm. ^{13}C NMR (150 MHz, $CDCl_3$) δ 160.0, 143.30, 140.65, 128.52, 127.33, 124.98, 123.69, 122.56, 122.26, 120.93, 115.02, 114.08, 109.40, 109.30, 76.36, 38.02, 13.77 ppm. Elem. Anal. Calcd. for $C_{18}H_{13}N_3$: C, 79.68; H, 4.83; N, 15.49 %. Found: C,

79.62 %; H, 4.72 %; N, 15.41 %. MS (MALDI), calcd for $C_{18}H_{13}N_3$, *m/z*= 271.11; found, *m/z*=271.12.

2-benzoyl-3-(9-ethyl-9H-carbazol-3-yl)acrylonitrile (CbV7)

Yellow-green solid, Yield: 68 %. FT-IR cm^{-1}(KBr), 1628 (aromatic C=C), 1665 (C=O), 2210 (CN), 2978 (aliphatic C-H) and 3055 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.48 (3H, t, -CH3, J = 7.2), 4.41 (2H, q, -CH2, J = 7.2), 7.33 (1H, t, CbPhH, J = 7.2), 7.45-7.57 (6H, m, COPhH & CbPhH), 7.63 (1H, t, COPhH, J = 7.2), 7.91 (2H, d, COPhH, J = 7.8), 8.15 (1H, d, CbPhH, J = 7.5), 8.30 (1H, s, CH), 8.78 (1H, s, CbPhH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 189.34, 157.17, 143.14, 140.79, 136.91, 132.93, 129.41, 129.28, 128.66, 127.15, 125.87, 123.86, 123.07, 122.97, 121.12, 120.79, 118.56, 109.41, 109.35, 105.38, 38.15, 14.0 ppm. Elem. Anal. Calcd. for $C_{24}H_{18}N_2O$: C, 82.26; H, 5.18; N, 7.99; O, 4.57 %. Found: C, 81.96 %; H, 5.06 %; N, 7.98 %. MS (MALDI), calcd for $C_{24}H_{18}N_2O$, *m/z*= 350.14; found, *m/z*=350.13.

2-((9-ethyl-9H-carbazol-3-yl)methylene)-1H-indene-1,3 (2H)-dione (CbV8)

Orange solid. Yield: 81 %. FT-IR cm^{-1}(KBr), 1625 (aromatic C=C), 1665 (C=O), 2976 (aliphatic C-H) and 3050 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.47 (3H, t, -CH3, J = 7.2), 4.39 (2H, q, -CH2, J = 7.2), 7.33 (1H, t, CbPhH, J = 7.5), 7.41-7.54 (3H, m, CbPhH), 7.73-7.80 (2H, m, InPhH), 7.95-8.03 (2H, m, InPhH), 8.07 (1H, s, CbPhH), 8.24 (1H, d, CbPhH, J = 8.4), 8.66 (1H, d, CbPhH, J = 8.7), 9.44 (1H, s, CH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 191.29, 143.32, 142.50, 140.71, 140.02, 134.92, 134.66, 133.48, 128.88, 126.79, 125.02, 123.79, 123.41, 122.96, 121.19, 120.69, 109.25, 108.78, 38.06, 14.0 ppm. Elem. Anal. Calcd. for $C_{24}H_{17}NO_2$: C, 82.03; H, 4.88; N, 3.99; %. Found: C, 81.97 %; H, 4.79 %; N, 3.99 %. MS (MALDI), calcd for $C_{24}H_{17}NO_2$, *m/z*= 351.13; found, *m/z*=351.12.

2-(2-((9-ethyl-9H-carbazol-3-yl)methylene)-3-oxo-2,3-dihydro-1H-inden-1-ylidene) malononitrile (CbV9)

Dark red solid. Yield: 67 %. FT-IR cm^{-1}(KBr), 1628 (aromatic C=C), 1704 (C=O), 2220 (CN), 2974 (aliphatic C-H) and 3050 (aromatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.50 (3H, t, -CH3, J = 7.2), 4.42 (2H, q, -CH2, J = 7.2), 7.36 (1H, t, CbPhH, J = 7.8), 7.46-7.57 (3H, m, CbPhH), 7.75-7.81 (2H, m, onmalPhH), 7.95 (1H, d, CbPhH, J= 7.5), 8.23 (1H, d, onmalPhH, J= 7.8), 8.50 (1H, d, onmalPhH, J= 8.7), 8.71 (1H, d, CbPhH, J= 7.8), 8.84 (1H, s, CbPhH), 9.25 (1H, s, CH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 188.32, 163.17, 149.76, 143.83, 140.56, 137.54, 135.09, 134.58, 134.07, 129.6, 127.08, 125.18, 124.53, 123.84, 123.58, 123.4, 123.3, 121.30, 121.11, 119.50, 115.29, 114.82, 109.46, 108.80, 69.88, 38.21, 14.01 ppm. Elem. Anal. Calcd. for $C_{27}H_{17}N_3O$: C, 81.19; H, 4.29; N, 10.52 %. Found: C, 81.08 %; H, 4.37 %; N, 10.68 %. MS (MALDI), calcd for $C_{27}H_{17}N_3O$, *m/z*= 399.14; found, *m/z*=399.13.

2,2'-(2-((9-ethyl-9H-carbazol-3-yl)methylene)-1H-indene-1,3(2H)-diylidene) dimalononitrile (CbV10)

Dark blue solid. Yield: 85 %. FT-IR cm^{-1}(KBr), 1630 (aromatic C=C), 2208 (CN), 2956 (aliphatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 1.51 (3H, t, $-CH_3$, J= 7.2), 4.43 (2H, q, -CH2, J = 7.2), 7.38 (1H, t, CbPhH, J = 7.5), 7.48-7.58 (3H, m, CbPhH), 7.74-7.82 (2H, m), 7.92 (1H, d, J=7.5), 7.96-8.1 (2H, m), 8.72 (1H, d, J= 7.8), 8.85 (1H, s), 9.3 (1H, s, CH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 190.1, 158.74, 148.54, 143.64, 140.54, 138.42, 135.02, 130.69, 129.2, 127.06, 125.2, 124.33, 123.54, 122.14, 118.51, 118.42, 109.26, 108.60, 103.3, 50.95, 39.6, 14.05 ppm. Elem. Anal. Calcd. for $C_{30}H_{17}N_5$: C, 80.52; H, 3.83; N, 15.65 %. Found: C, 80.26; H, 3.57; N, 15.37 %. MS (MALDI), calcd for $C_{30}H_{17}N_5$, *m/z*= 447.15; found, *m/z* =447.28.

2-(4-bromophenyl)-3-(4-(dimethylamino)phenyl)acrylonitrile (AnV1)

Yellow solid, Yield: 86 %. FT-IR cm^{-1}(KBr), 1612 (aromatic C=C), 2206 (CN), 2908 (aliphatic C-H) and 3037 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.1 (6H, s, $(NCH_3)_2$), 6.70 (2H, d, ArH, J = 9 Hz), 7.37 (s, 1H, CH), 7.46-7.53 (m, 4H, PhBr), 7.84 (2H, d, ArH, J = 9 Hz). ^{13}C NMR (150 MHz, $CDCl_3$) δ 151.95, 142.95, 134.74, 132.08, 131.56, 127.02, 121.94, 121.38, 119.24, 111.71, 103.25, 40.14. Elem. Anal. Calcd. for $C_{17}H_{15}BrN_2$: C, 62.40; H, 4.62; Br, 24.42; N, 8.56 %. Found: C, 62.56 %; H, 4. 61 %; N, 8.65 %. MS (MALDI), calcd for $C_{17}H_{15}BrN_2$, *m/z*= 326.04; found, *m/z*=326.04.

2-(4-(dimethylamino)phenyl)-3-(thiophen-2-yl)acrylonitrile (AnV2)

Green solid. Yield: 80 %. FT-IR cm^{-1}(KBr), 1612 (aromatic C=C), 2208 (CN), 2908 (aliphatic C-H) and 3012 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.61 (6H, s, $N(CH_3)_2$), 6.71 (2H, d, ArH, J = 9 Hz), 7.02-7.05 (1H, m, TpH), 7.20 (1H, d, TpH, J = 6 Hz), 7.25 (1H, s, CH), 7.27 (1H, d, TpH, J = 6 Hz), 7.80 (2H, d, ArH, J = 9 Hz). ^{13}C NMR (150 MHz, $CDCl_3$) δ 151.75, 140.64, 131.21, 128.03, 128.01, 125.25, 124.59, 121.30, 118.38, 111.77, 99.48, 40.16. Elem. Anal. Calcd. for $C_{15}H_{14}N_2S$: C, 70.83; H, 5.55; N, 11.01; S, 12.61 %. Found: C, 70.67 %; H, 5.54 %; N, 10.85; S, 12.43 %. MS (MALDI), calcd for $C_{15}H_{14}N_2S$, *m/z*= 254.09; found, *m/z*=254.08.

2-(biphenyl-4-yl)-3-(4-(dimethylamino)phenyl)acrylonitrile (AnV3)

Yellow solid. Yield: 85 %. FT-IR cm^{-1}(KBr), 1612 (aromatic C=C), 2210 (CN), 2908 (aliphatic C-H) and 3038 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.07 (6H, s, $N(CH_3)_2$), 6.74 (2H, d, ArH, J = 9 Hz), 7.46 (1H, s, CH), 7.40-7.51(5H, m, PhH), 7.68-7.61(4H, m, PhH), 7.88 (2H, d, ArH, J = 9 Hz). ^{13}C NMR (150 MHz, $CDCl_3$) δ 151.82, 142.36, 141.24, 140.34, 134.66, 131.47, 128.95, 127.96, 127.20, 125.93, 121.76, 119.56, 111.76, 104.20, 40.18, 23.45. Elem. Anal. Calcd. for $C_{23}H_{20}N_2$: C, 85.15; H, 6.21; N, 8.63 %. Found: C, 85.55 %; H, 6.12 %; N, 8.48 %. MS (MALDI), calcd for $C_{23}H_{20}N_2$, *m/z*= 324.16; found, *m/z*=324.16.

2-cyano-3-(4-(dimethylamino)phenyl)acrylic acid (AnV4)

Yellow solid. Yield 63 %. FT-IR cm^{-1}(KBr), 1615 (aromatic C=C), 1668 (C=O of carboxylic group), 2219 (CN), 2972 (aromatic C-H), 3436 (O-H, carboxylic acid). ^{1}H NMR (300 MHz, DMSO, TMS, ppm): δ 3.07 (6H, s, NCH_3), 6.82 (2H, d, ArH, J= 9), 7.93 (2H, d, ArH, J= 9), 8.07 (1H, s, CH), 13.25 (1H, s, OH). ^{13}C NMR (150 MHz, DMSO) δ 165.40, 154.36, 154.02, 134.06, 118.97, 118.49, 112.17, 94.02, 39.64 ppm. Elem. Anal. Calcd. for $C_{12}H_{12}N_2O_2$: C, 66.65; H, 5.59; N, 12.96 %. Found: C, 66.69; H, 5.66; N, 12.97 %. MS (MALDI), calcd for $C_{12}H_{12}N_2O_2$, *m/z*= 216.09; found, *m/z*=216.10.

Ethyl 2-cyano-3-(4-(dimethylamino)phenyl)acrylate (AnV5)

Orange solid. Yield: 72 %. FT-IR cm^{-1}(KBr), 1612 (aromatic C=C), 1706 (C=O, ester), 2214 (CN), 2935 (aliphatic C-H) and 2990 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 8.06 (1H, s, CH), 7.93 (2H, d, J=9.3 Hz, ArH), 6.69 (2H, d, J=9 Hz, ArH), 4.33 (2H, q, J=7.1 Hz, CH2), 3.10 (s, 6H, NCH3), 1.37 (3H, t, J=7.2 Hz, CH3). ^{13}C NMR (150 MHz, $CDCl_3$) δ 164.39, 154.63, 153.68, 134.15, 119.46, 117.70, 111.59, 86.16, 61.8, 40.26, 14.26 ppm. Elem. Anal. Calcd. for $C_{14}H_{16}N_2O_2$: C, 68.83; H, 6.60; N, 11.47 %. Found: C, 68.61 %; H, 6.66 %; N, 11.40 %. MS (MALDI), calcd for $C_{14}H_{16}N_2O_2$, *m/z*= 244.12; found, *m/z*=244.12.

2-(4-(dimethylamino)benzylidene)malononitrile (AnV6)

Yellow solid. Yield: 67 %. FT-IR cm^{-1}(KBr), 1618 (aromatic C=C), 2216 (CN), 2925 (aliphatic C-H) and 3060 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 7.81 (2H, d, ArH, J = 9.0 Hz), 7.45 (1 H, s, CH), 6.69 (2 H, d, ArH, J = 9.0 Hz), 3.14 (6 H, s, $(NCH_3)_2$). ^{13}C NMR (150 MHz, $CDCl_3$) δ 158.20, 154.37, 133.92, 119.40, 116.13, 115.05, 111.73, 39.76. Elem. Anal. Calcd. for $C_{12}H_{11}N_3$: C, 73.07; H, 5.62; N, 21.30 %. Found: C, 72.89; H, 5.58; N, 20.99 %. MS (MALDI), calcd for $C_{12}H_{11}N_3$, *m/z*= 197.10; found, *m/z*=197.09.

2-benzoyl-3-(4-(dimethylamino)phenyl)acrylonitrile (AnV7)

Orange solid. Yield: 69 %. FT-IR cm^{-1}(KBr), 1618 (aromatic C=C), 1654 (C=O), 2204 (CN), 2948 (aliphatic C-H) and 3054 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.13 (6H, s, N $(CH_3)_2$); 6.72 (2H, d, ArH, J = 9 Hz); 7.46-7.55 (2H, m, COPh, J=7.2); 7.57 (1H, t, COPh, J = 4.8 Hz); 7.84 (2H, d, COPh, J=7.2); 7.89 (1H, s, CH), 8.0 (2H, d, ArH, J = 9 Hz). ^{13}C NMR (150 MHz, $CDCl_3$) δ 188.74, 155.87, 153.98, 137.46, 134.65, 132.46, 129.05, 128.49, 119.84, 119.33, 111.73, 101.75, 40.26 ppm. Elem. Anal. Calcd. for $C_{18}H_{16}N_2O$: C, 78.24; H, 5.84; N, 10.14 %. Found: C, 77.94; H, 5.80; N, 10.03 %. MS (MALDI), calcd for $C_{18}H_{16}N_2O$, *m/z*= 276.13; found, *m/z*=276.12.

2-(4-(dimethylamino)benzylidene)-1H-indene-1,3(2H)-dione (AnV8)

Red solid. Yield: 80%. FT-IR cm^{-1}(KBr): 1608 (aromatic vC=C), 1662 & 1708 (vC=O), 2948 (aliphatic vC-H) and 3000 (aromatic vC-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.15 (6H, s, N $(CH_3)_2$), 6.75 (2H, d, ArH, J = 9 Hz), 7.71-7.75 (2H, m, COPh), 7.83 (1H, s, CH), 7.91-7.95 (2H, m, COPh), 8.55 (2H, d, ArH, J = 9 Hz) ppm. ^{13}C NMR (150 MHz, $CDCl_3$) δ 190.1, 154.06, 147.62, 142.35, 139.99, 138.09, 134.49, 134.21, 122.59, 111.51, 40.18 ppm. Elem. Anal. Calcd. for $C_{18}H_{15}NO_2$: C, 77.96; H, 5.45; N, 5.05 %. Found: C, 77.38; H, 5.45; N, 5.03 %. MS (MALDI), calcd for $C_{18}H_{15}NO_2$, *m/z*= 277.11; found, *m/z*=277.10.

2-(2-(4-(dimethylamino)benzylidene)-3-oxo-2,3-dihydro-1H-inden-1-ylidene) malononitrile (AnV9)

Dark red solid. Yield 60 %. FT-IR cm^{-1}(KBr), 1608 (aromatic C=C), 1674 (C=O), 2214 (CN), 2948 (aliphatic C-H) and 2994 (aromatic C-H). ^{1}H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.07 (6H, s, N(CH3)$_2$), 6.68 (2H, d, J = 8.7 Hz), 7.20-7.36 (2H, m), 7.70 (1H, t, J = 8.1 Hz), 7.85 (1H, t, J = 8.4 Hz), 8.37 (2H, d, ArH, J = 9.3 Hz), 8.50 (1H, s, CH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 198.93, 189.55, 159.17, 149.81, 139.87, 135.86, 134.08, 131.65, 129.51, 127.41, 125.74, 123.54, 120.70, 115.39, 111.64, 61.08, 40.33 ppm. Elem. Anal. Calcd. for $C_{21}H_{15}N_3O$: C, 77.52; H, 4.65; N, 12.91 %. Found: C, 76.98; H, 4.80; N, 13.39 %. MS (MALDI), calcd for $C_{21}H_{15}N_3O$, *m/z*= 325.12; found, *m/z*=325.74.

2,2'-(2-(4-(dimethylamino)benzylidene)-1H-indene-1,3(2H)-diylidene)dimalononitrile (AnV10)

Dark blue solid. Yield 77 %. FT-IR cm^{-1}(KBr), 1632 (aromatic C=C), 2200 (CN), 2950 (aliphatic C-H). ^1H NMR (300 MHz, $CDCl_3$, TMS, ppm): δ 3.08 (6H, s, $N(CH_3)_2$), 5.66 (1H, s, CH), 6.59 (2H, d, ArH, J = 9.3 Hz), 7.21-7.26 (2H, m, PhH), 7.48 (2H, d, ArH, J = 9.3 Hz), 7.92-7.96 (2H, m, PhH). ^{13}C NMR (150 MHz, $CDCl_3$) δ 190.39, 164.78, 158.72, 138.42, 136.49, 132.1, 122.13, 118.51, 118.41, 111.61, 103.32, 59.84, 39.96 ppm. Elem. Anal. Calcd. for $C_{24}H_{15}N_5$: C, 77.20; H, 4.05; N, 18.76 %. Found: C, 77.29; H, 4.37; N, 18.27 %. MS (MALDI), calcd for $C_{24}H_{15}N_5$, *m/z*= 373.13; found, *m/z*=373.56.

- Supplementary Figures and Table -

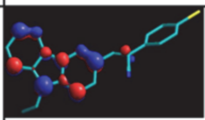
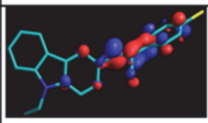
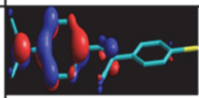
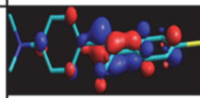
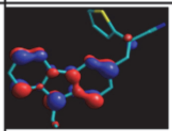
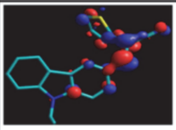
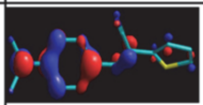
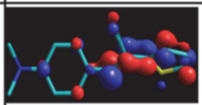
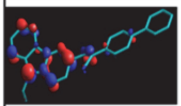
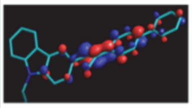
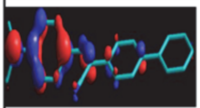
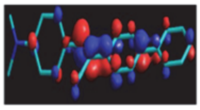
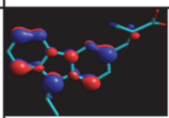
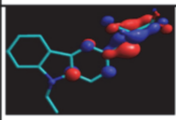
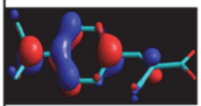
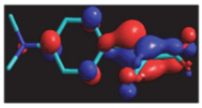
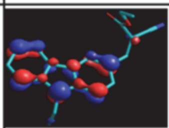
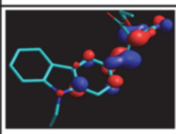
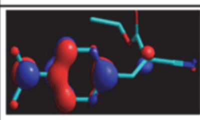
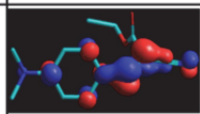
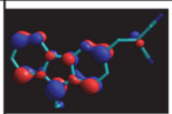
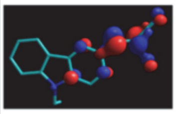
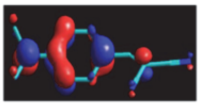
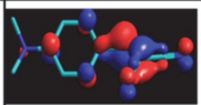
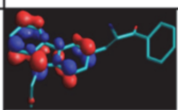
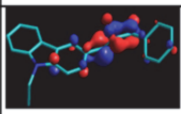
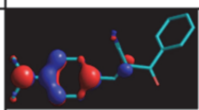
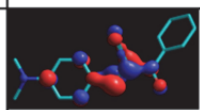
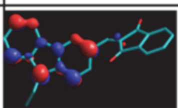
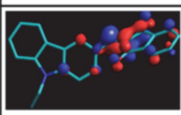
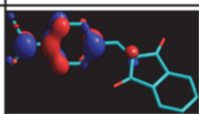
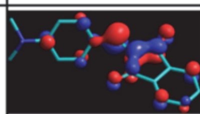
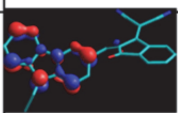
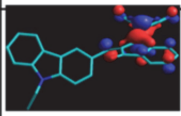
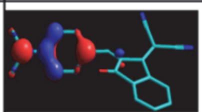
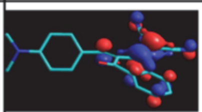
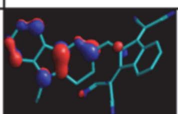
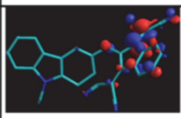
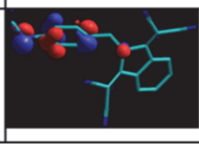
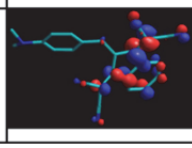
9-ethylcarbazole derivative PL dyes			Dimethyl aniline derivative PL dyes		
PL dyes	HOMO	LUMO	PL dyes	HOMO	LUMO
CbV1			AnV1		
CbV2			AnV2		
CbV3			AnV3		
CbV4			AnV4		
CbV5			AnV5		
CbV6			AnV6		
CbV7			AnV7		
CbV8			AnV8		
CbV9			AnV9		
CbV10			AnV10		

Figure S1. Optimized geometries of ArV, overlaid with HOMO and LUMO contour diagrams (HF/PM3).

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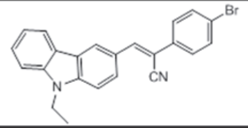
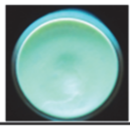
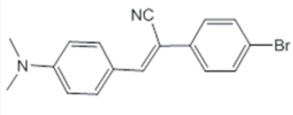
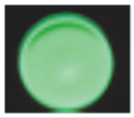
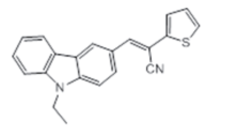
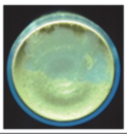
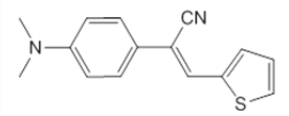
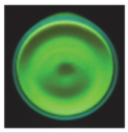
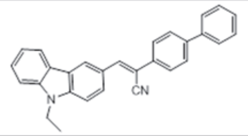
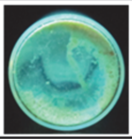
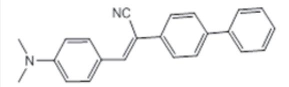
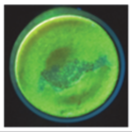
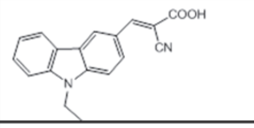
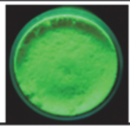
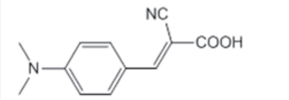
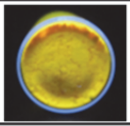
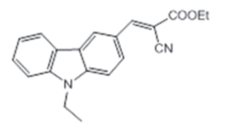
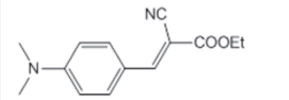
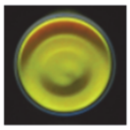
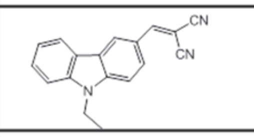
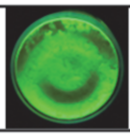
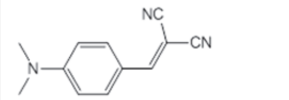
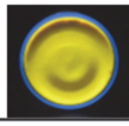
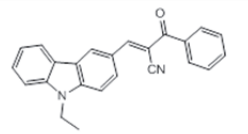
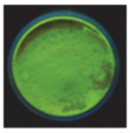
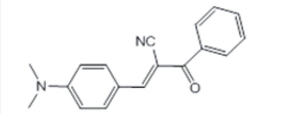
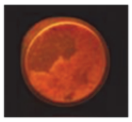
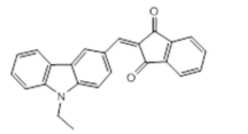
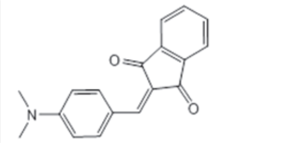
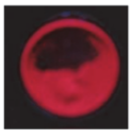
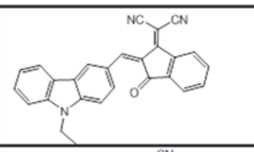
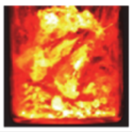
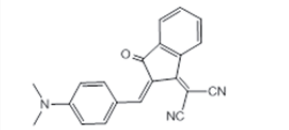
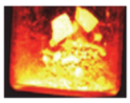
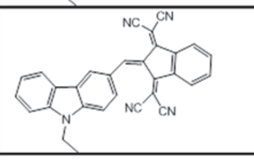
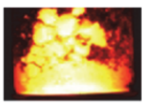
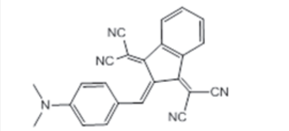
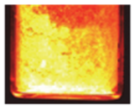
CbV1			AnV1		
CbV2			AnV2		
CbV3			AnV3		
CbV4			AnV4		
CbV5			AnV5		
CbV6			AnV6		
CbV7			AnV7		
CbV8			AnV8		
CbV9			AnV9		
CbV10			AnV10		

Figure S2. Fluorescence of ArV powders under the illumination at 365 nm. The NIR fluorescence images of CbV9-10 and AnV9-10 were taken by a Kodak imaging system with a filter set of excitation/emission for Cy5.5.

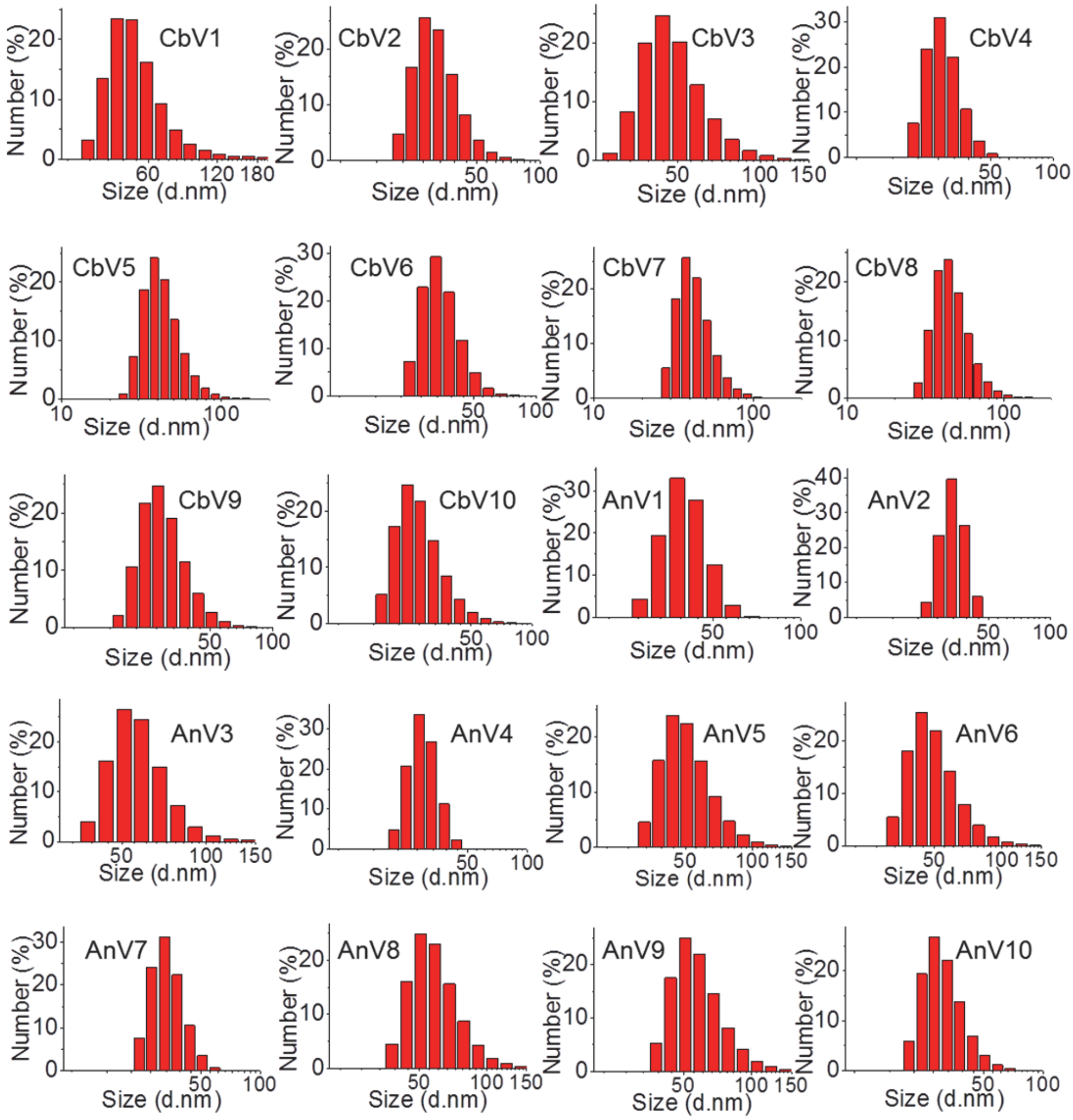


Figure S3. Hydrodynamic sizes of self-aggregated nanoparticles of CbV and AnV in THF/H_2O (= 1/9 by vol.), measured by DLS.

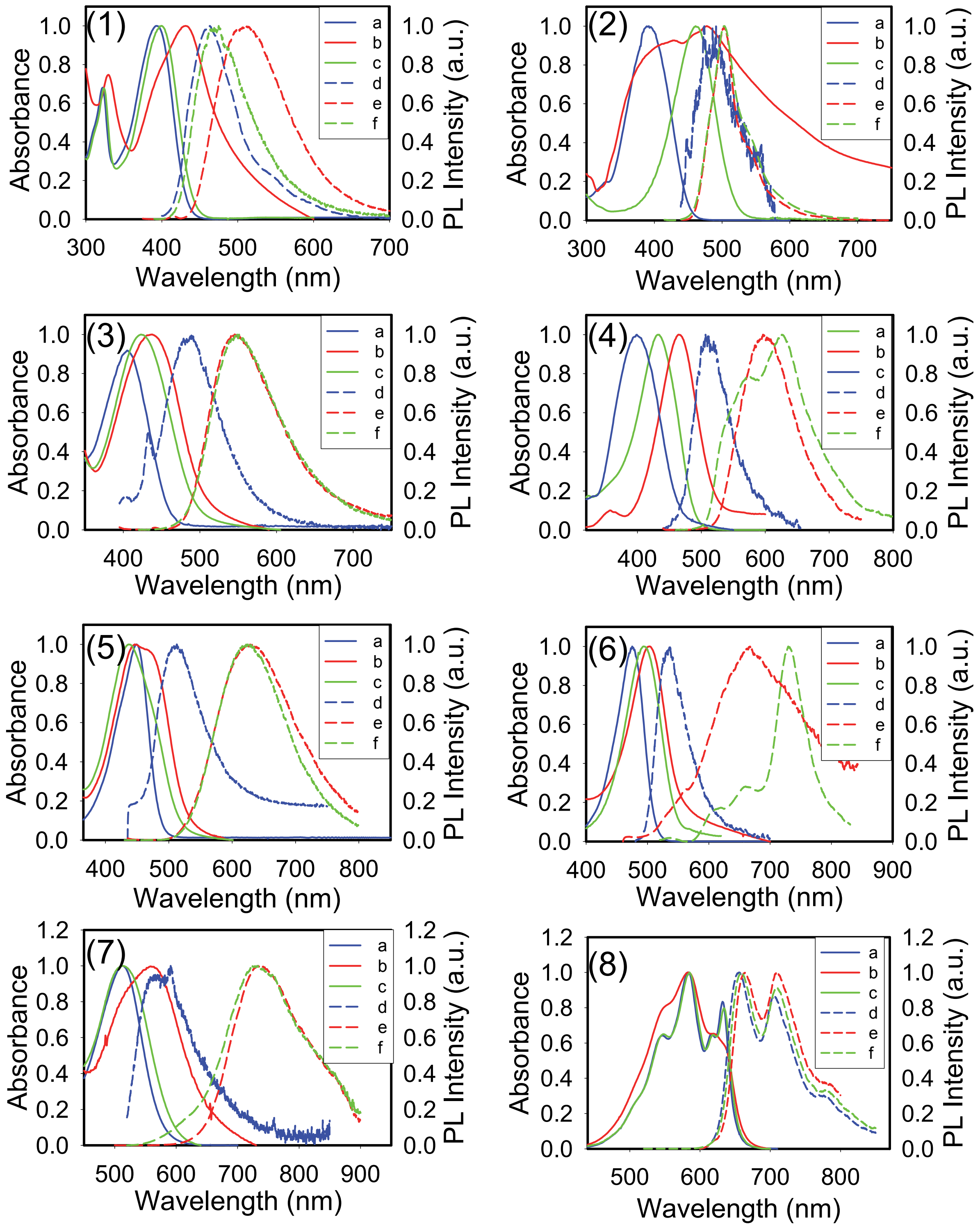


Figure S4. Normalized UV and PL spectra of representative ArV dyes: absorption in (a) THF solution, (b) self-aggregates and (c) FArV NPs; emission in (d) THF solution (e) self-aggregates and (f) FArV NPs. (1) CbV5 (2) AnV1 (3) CbV7 (4) AnV7 (5) CbV8 (6) AnV8 (7) CbV9 (8) CbV10.

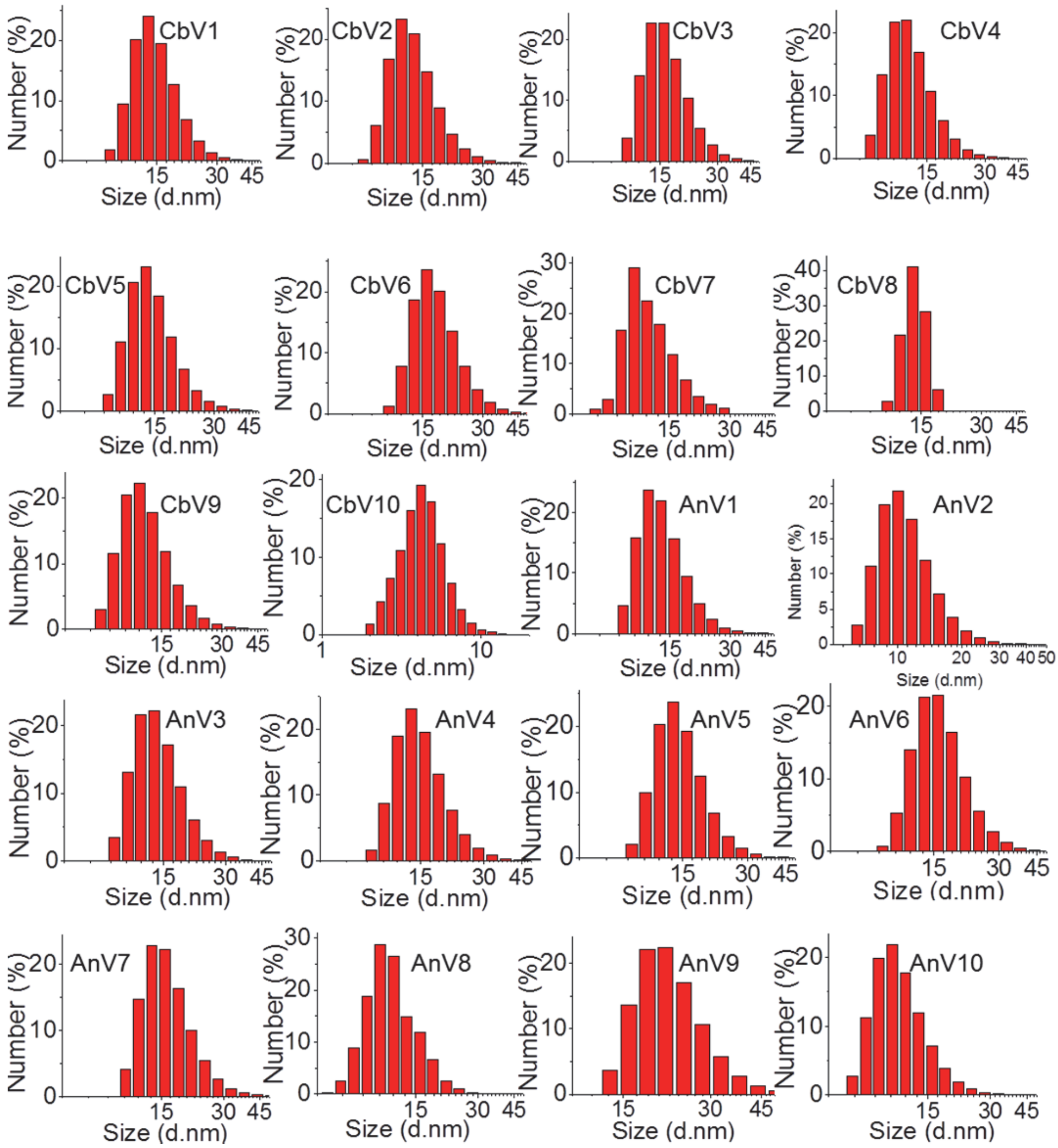


Figure S5. Hydrodynamic sizes of FArV NPs, measured by DLS.

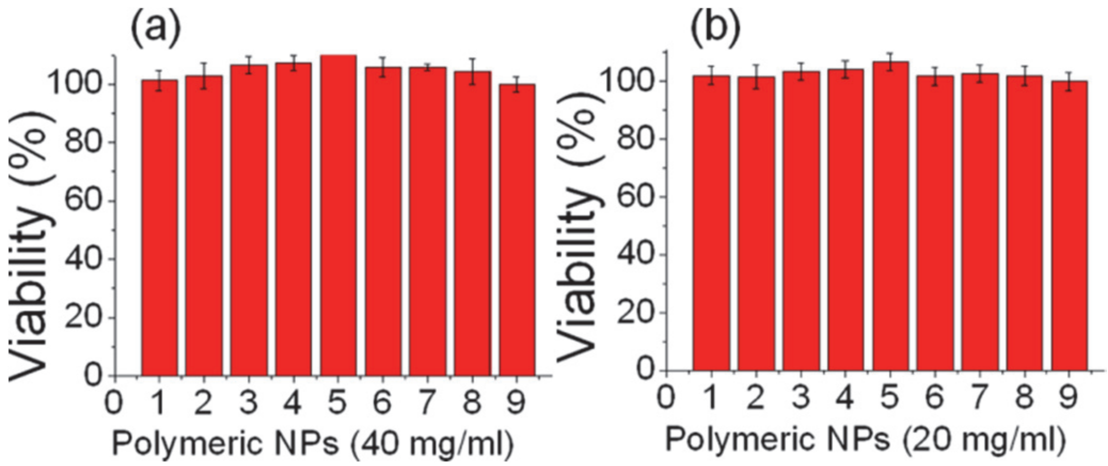


Figure S6. In vitro MTT assay showing cytotoxicity of FArV NPs against HeLa cells at concentrations of (a) 40 mg/mL and (b) 20 mg/mL. (1) CbV5, (2) AnV1, (3) CbV7, (4) AnV7, (5) CbV8, (6) AnV8, (7) CbV10, (8) CbV9, and (9) control (untreated).

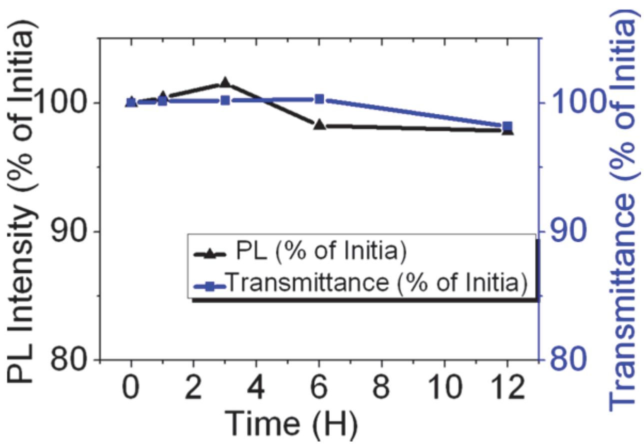


Figure S7. Stability test of FCbV10 NPs in biological condition (90% FBS, 37 ^{o}C), monitored with fluorescence and transmittance (scattering) changes.

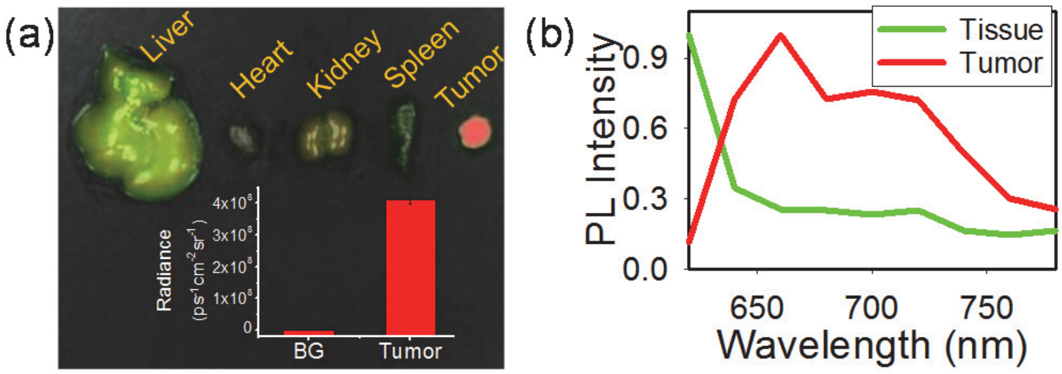


Figure S8. (a) Pseudo-color ex vivo image of tumor and other major organs resected from a mouse inoculated with SCC7 tumor (2 weeks after tumor cell injection). Image was taken at 5 h after tail vein injection of FCbV10 NPs. FCbV10 NPs signal from the tumor (red) and the tissue autofluorescence from other organs (green) were spectrally unmixed according to their spectral profiles shown in the same colors (b). The inset in (a) indicates the ex vivo intensities of FCbV10 NPs in the tumor and the background.

Table S1. Hydrodynamic sizes (nm), determined by DLS

ArV	Self-ggregates	FArV	**ArV**	Self-aggregates	FArV
CbV1	56.37	15.02	AnV1	40.37	14.03
CbV2	32.65	13.83	AnV2	33.29	11.53
CbV3	49.34	16.53	AnV3	62.08	14.62
CbV4	29.56	12.58	AnV4	29.74	15.44
CbV5	43.44	14.93	AnV5	52.18	14.96
CbV6	34.96	17.97	AnV6	50.71	17.34
CbV7	43.76	12.92	AnV7	34.16	16.50
CbV8	47.34	13.95	AnV8	60.01	15.65
CbV9	30.48	12.89	AnV9	59.47	22.42
CbV10	28.62	8.78	AnV10	31.75	11.35