Supporting information:

Table S1, Cartesian coordinates, crystal structure and XRD spectrum of the optimized monomers and fragments:

Table S1. Selected experimental and theoretical bond lengths [Å] and angles [º] for **2** and **3**.

2	Experimental	Calculated	3	Experimental	Calculated
Hg(1)−N(1)	2.375(4)	2.675	Hg(1)−N(1)	2.287(12)	2.214
Hg(1)–N(2)	2.419(4)	2.719	Hg(1)–N(2)	2.420(15)	2.375
Hg(1)–Br(2)	2.5193(5)	2.426	Hg(1)–I(2)	2.6907(10)	2.640
Hg(1)–Br(1)	2.5377(5)	2.441	Hg(1)–I(1)	2.6962(10)	2.643
N(1)–C(1)	1.325(6)	1.322	Hg(2)–I(3)	2.6244(10)	2.587
N(1)–C(9)	1.367(6)	1.353	Hg(2)–I(4)	2.6350(10)	2.623
N(2)–C(8)	1.433(6)	1.407	Hg(2)–I(1)^{i}	3.2105(11)	3.376
N(2)–H(1N)	0.9000	0.910	Hg(2)–I(2)	3.2200(11)	3.380
N(2)–H(2N)	0.8999	0.912	Hg(3)–I(5)	2.6057(8)	2.729
N(1)–Hg(1)–N(2)	70.82(13)	63.46	Hg(3)–I(5)^{ii}	2.6057(8)	2.725
N(1)–Hg(1)–Br(2)	101.48(9)	101.03	I(1)–Hg(2)^{iii}	3.2105(11)	3.376
N(2)–Hg(1)–Br(2)	123.46(10)	116.14	N(1)–C(1)	1.29(2)	1.273
N(1)–Hg(1)–Br(1)	110.51(9)	104.06	N(1)–C(9)	1.351(18)	1.357
N(2)–Hg(1)–Br(1)	95.41(10)	88.55	N(2)–C(8)	1.43(2)	1.406
Br(2)–Hg(1)–Br(1)	136.264(16)	150.16	N(2)–H(1N)	0.9000	0.905
C(1)–N(1)–C(9)	118.8(4)	120.31	N(2)–H(2N)	0.8999	0.909
C(1)–N(1)–Hg(1)	123.5(3)	120.65	N(1)–Hg(1)–N(2)	72.5(4)	70.19
C(9)–N(1)–Hg(1)	117.4(3)	114.89	N(1)–Hg(1)–I(2)	112.3(3)	109.40
C(8)–N(2)–Hg(1)	113.6(3)	111.60	N(2)–Hg(1)–I(2)	118.4(3)	123.46
C(8)–N(2)–H(1N)	100.4	109.36	N(1)–Hg(1)–I(1)	118.1(3)	115.79
Hg(1)–N(2)–H(1N)	100.7	105.70	N(2)–Hg(1)–I(1)	107.5(3)	109.32
C(8)–N(2)–H(2N)	136.0	125.38	I(2)–Hg(1)–I(1)	119.30(3)	128.64
Hg(1)–N(2)–H(2N)	99.8	92.54	I(3)–Hg(2)–I(4)	161.95(4)	166.57

H(1N)–N(2)–H(2N)	100.7	104.62	I(3)–Hg(2)–I(1)[ii]	97.06(3)	98.70
			I(4)–Hg(2)–I(1)[ii]	94.34(3)	97.12
			I(3)–Hg(2)–I(2)	103.07(3)	108.69
			I(4)–Hg(2)–I(2)	92.23(3)	95.82
			I(1)#1–Hg(2)–I(2)	82.62(3)	81.30
			I(5)–Hg(3)–I(5)[ii]	180.0	179.68
			Hg(1)–I(1)–Hg(2)[iii]	90.51(3)	92.47
			Hg(1)–I(2)–Hg(2)	108.67(3)	111.52
			C(1)–N(1)–C(9)	117.0(16)	117.23
			C(1)–N(1)–Hg(1)	124.0(13)	126.28
			C(9)–N(1)–Hg(1)	118.4(9)	115.37
			C(8)–N(2)–Hg(1)	111.0(10)	110.94
			C(8)–N(2)–H(1N)	109.3	112.83
			Hg(1)–N(2)–H(1N)	109.5	108.46
			C(8)–N(2)–H(2N)	109.4	109.27
			Hg(1)–N(2)–H(2N)	109.5	107.42
			H(1N)–N(2)–H(2N)	108.1	107.66

Symmetry codes: (i) *x*−1, *y*, *z*; (ii) –*x*+1, −*y*, −*z* +1; (iii) *x*+1, *y*, *z* for **3**

Cartesian coordinates:

Hg-Br

Optimized Monomer Structure

Hg	-1.14680566	0.15045797	-0.06609711
Br	-2.34102649	-1.94111278	0.22724841
Br	-0.94358316	2.57262578	-0.29561211
N	1.22168457	-0.48185841	1.00492388

N	0.61655557	-1.27996941	-1.56276212
H	0.28054577	-1.97179435	-1.07480190
H	0.50097187	-1.45536165	-2.45156840
C	1.49663257	-0.17608241	2.25885488
H	0.65294457	-0.10578441	2.93874788
C	2.80347157	0.07175959	2.72782588
H	2.96281857	0.30705359	3.77260088
C	3.84264357	0.04068859	1.83120188
H	4.85720057	0.25936359	2.14700188
C	3.59389557	-0.27393941	0.47311688
C	4.60912057	-0.29393641	-0.51592612
H	5.62925357	-0.06069241	-0.23324812
C	4.28885657	-0.60293041	-1.81494412
H	5.05763857	-0.61077441	-2.57859212
C	2.96634657	-0.94469641	-2.17217212
H	2.74758357	-1.22425441	-3.19746512
C	1.95311057	-0.95347341	-1.23621112
C	2.24622657	-0.56933241	0.10925288

Hg-I

Optimized Monomer Structure

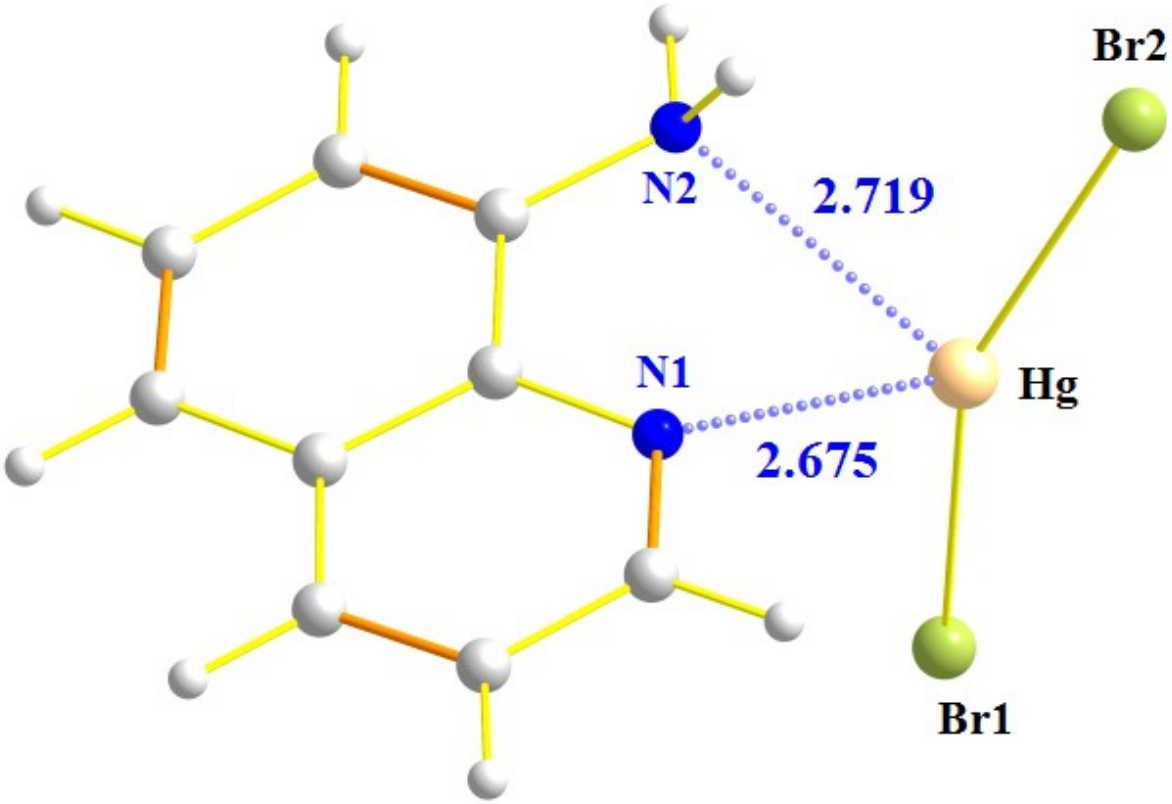
Hg	-4.04190994	1.61434432	4.19898522
N	-5.24222821	1.46240677	2.31293119
N	-4.00651301	-0.66690013	3.53693087
H	-3.15193280	-0.95385654	3.57766997
H	-4.49000883	-1.15628871	4.13181277
C	-5.89991337	2.44344833	1.73781948
H	-5.98011337	3.26134833	2.21181948
C	-6.49261337	2.32974833	0.41251948
H	-6.97741337	3.06744833	0.06131948
C	-6.37891337	1.22604833	-0.31588052
H	-6.73791337	1.17644833	-1.19478052
C	-5.66331337	0.05974833	0.29831948
C	-5.54791337	-1.07895167	-0.39888052
H	-5.91781337	-1.17515167	-1.26748052
C	-4.85001337	-2.11265167	0.22391948
H	-4.70371337	-2.91595167	-0.26048052
C	-4.34791337	-2.01645167	1.55091948
H	-3.91631337	-2.75555167	1.96271948
C	-4.46545403	-0.80871890	2.21029531
C	-5.15997359	0.32008461	1.57046303
I	-9.15569832	1.57629828	12.87412486
Hg	-5.13596111	-1.47414511	13.13334758
Hg	-4.60424823	0.06272280	8.61779157

I	-7.61137111	-2.35391545	13.39993511
I	-3.63205520	-2.25006850	11.05983903
I	-3.79795793	-1.80775058	6.80812629
N	-3.94411869	-1.31202558	14.99167769
N	-5.11455102	0.82824740	13.71742879
H	-5.95452662	1.16236153	13.68357484
H	-4.62135108	1.27980978	13.10162271
C	-3.32025405	-2.29656447	15.58294277
H	-3.27955585	-3.13078035	15.13296506
C	-2.70134231	-2.17084528	16.89813983
H	-2.24087312	-2.91646271	17.26488170
C	-2.76063643	-1.04225280	17.59415271
H	-2.38682551	-0.98079827	18.46610173
C	-3.43875660	0.13278225	16.95471292
C	-3.49883797	1.29546843	17.61850268
H	-3.11245513	1.40272382	18.47859047
C	-4.16472706	2.33711018	16.97425575
H	-4.27198574	3.15969132	17.43598488
C	-4.69006440	2.22144745	15.65784750
H	-5.09833409	2.96454042	15.22976420
C	-4.62943711	0.99113375	15.03529058
C	-3.97091404	-0.14854340	15.69594759
Hg	-0.63804202	0.33918881	6.14873339
I	0.01720145	-1.49004046	4.43942147
I	-1.61790654	1.73995704	8.13812665
I	-5.42214249	1.92648400	10.43533527
Hg	-8.59767162	-0.15541849	11.03561063
I	-1.62337482	2.60433311	3.84698463
I	-5.58475078	2.37994419	6.18741719

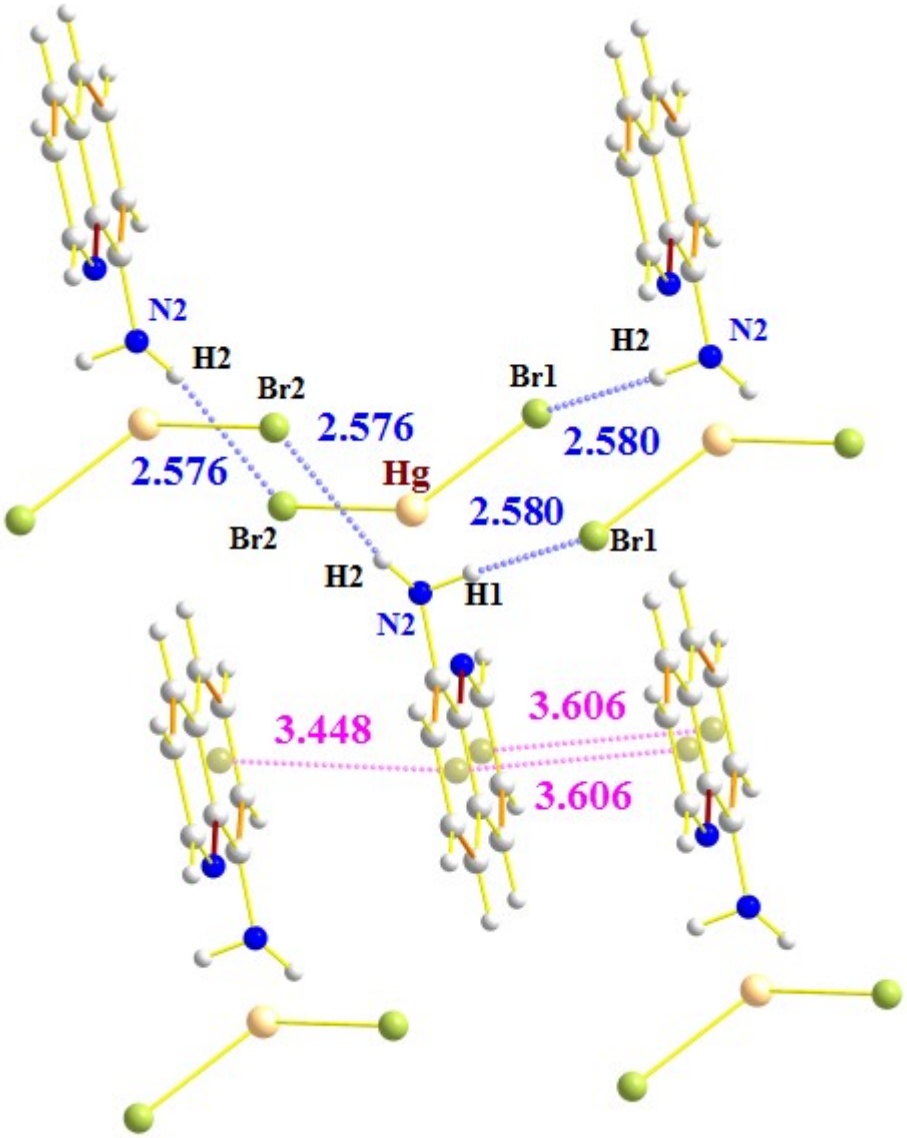
I	-7.59200394	-1.61263057	9.10021231

Crystal Structures

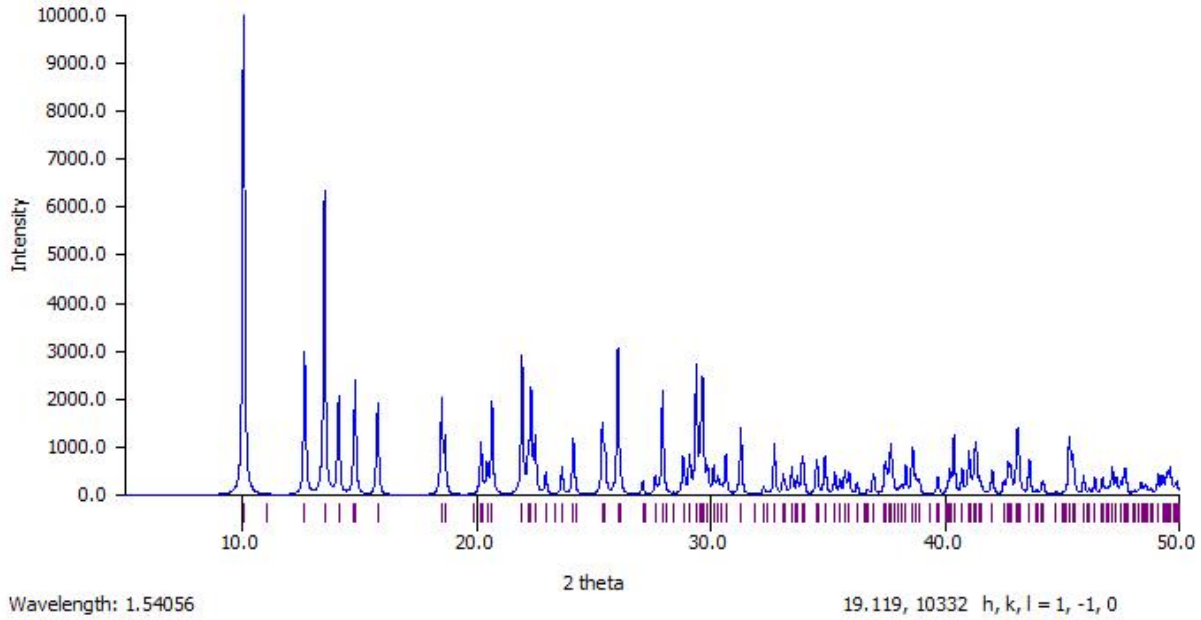
2-monomer



2-Net



2-PXRD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