Electronic Supporting Information

Supramolecular Chemistry with Uranyl Tetrahalide ($[UO_2X_4]^{2-}$) Anions

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Synthesis of 2-5

[UO_2Br_4]($C_{12}H_{14}N_2$) (2)

Compound **2** was prepared by dissolving 0.128 g of $UO_2(CH_3COO)_2 \cdot 2H_2O$ in water (4.0 mL) and HBr (0.40 mL, 48% in H_2O) in a 25 mL Erlenmeyer flask. To this yellow solution, 1,2-bis(4-pyridyl)ethane (0.051 g) was added. The resulting mixture was evaporated using gentle heat to an approximate volume of 2 mL and allowed to cool. The flask was then covered with a piece of Parafilm into which several holes were punched. After 45 days X-ray quality crystals were obtained.

[UO_2Br_4]($C_{12}H_{12}N_2$) (3)

Compound **3** was prepared by dissolving 0.256 g of $UO_2(CH_3COO)_2 \cdot 2H_2O$ in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) in a 25 mL Erlenmeyer flask. To this yellow solution, trans-1,2-bis(4-pyridyl)ethylene (0.250 g) in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) was added. The resulting mixture was evaporated using gentle heat to an approximate volume of 4 mL and allowed to cool. The flask was then covered with a piece of Parafilm into which several holes were punched. After 2.5 months, yellow, X-ray quality crystals were obtained as a mixture with white solids. These were easily physically separated under magnification.

[UO_2Br_4]($C_{10}H_{11}N_3$)$_2 \cdot$2Br$\cdot$2H_2O (4)

Compound **4** was prepared by dissolving 0.258 g of $UO_2(CH_3COO)_2 \cdot 2H_2O$ in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) in a 25 mL Erlenmeyer flask. To this yellow solution, 4,4'-dipyridylamine (0.239 g) in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) was added. The resulting mixture was evaporated using gentle heat to an approximate volume of 4 mL and allowed to cool. The flask was then covered with a piece of Parafilm into which several holes were punched. After 2.5 months, large X-ray quality crystals were obtained.

[UO_2Br_4]($C_{13}H_{16}N_2$)$_2 \cdot$2Br (5)

Compound **5** was prepared by dissolving 0.261 g of $UO_2(CH_3COO)_2 \cdot 2H_2O$ in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) in a 25 mL Erlenmeyer flask. To this yellow solution, 4,4'-trimethylene dipyridine (0.236 g) in water (2.5 mL) and HBr (0.75 mL, 48% in H_2O) was added. The resulting mixture was evaporated using gentle heat to an approximate volume of 4 mL and allowed to cool. The flask was then covered with a piece of Parafilm into which several holes were punched. After 2.5 months, yellow-orange, X-ray quality crystals were obtained.

Table S1a. Hydrogen bonds for **1a** [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...Br(1)#3	0.86	2.62	3.319(4)	139.6
N(1)-H(1)...Br(2)#3	0.86	3.08	3.704(4)	131.1

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,-z #2 -x+2,-y+1,-z+1 #3 x,y+1,z

Table S2. Hydrogen bonds for **2** [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...Br(2)#3	0.86	2.77	3.456(3)	137.4
N(1)-H(1)...Br(1)#3	0.86	2.84	3.468(3)	131.1

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,-z #2 -x,-y+1,-z+1 #3 -x+1,-y+1,-z

Table S3. Hydrogen bonds for **3** [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...Br(1)#3	0.86	2.71	3.459(3)	146.4
N(1)-H(1)...Br(2)#3	0.86	2.94	3.487(3)	122.9

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,-z #2 -x-1,-y+1,-z+1 #3 x+1,y,z

Table S4. Hydrogen bonds for **4** [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...Br(1)#2	0.86	2.72	3.455(4)	144.0
N(1)-H(1)...Br(3)#2	0.86	3.06	3.620(5)	124.4
N(3)-H(3)...OW1#3	0.86	1.90	2.724(5)	159.8
N(2)-HN2...Br(3)#4	0.73(4)	2.65(4)	3.374(4)	171(4)

Symmetry transformations used to generate equivalent atoms:
#1 -x+2,-y,-z+2 #2 -x+1,-y,-z+1 #3 x,y,z+1
#4 -x+1,-y+1,-z+1

Table S5. Hydrogen bonds for **5** [A and deg.].

D-H...A	d(D-H)	d(H...A)	d(D...A)	<(DHA)
N(1)-H(1)...Br(3)#2	0.86	2.41	3.221(8)	157.5
N(2)-H(2)...Br(3)	0.86	2.34	3.187(6)	168.1

Symmetry transformations used to generate equivalent atoms:
#1 -x,-y,-z #2 x,y-1,z+1