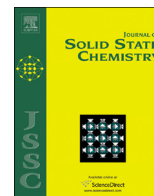




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Anion-controlled assembly of silver-di(aminophenyl)sulfone coordination polymers: Syntheses, crystal structures, and solid state luminescence

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ABSTRACT

Five silver coordination polymers, namely, $\{[\text{Ag}(3,3'\text{-daps})_2] \cdot \text{BF}_4\}_n$ (**1**), $\{[\text{Ag}(3,3'\text{-daps})_2] \cdot \text{NO}_3\}_n$ (**2**), $[\text{Ag}(3,3'\text{-daps})(\text{CF}_3\text{SO}_3)]_n$ (**3**), $\{[\text{Ag}(4,4'\text{-daps})] \cdot \text{CF}_3\text{SO}_3\}_n$ (**4**), and $\{[\text{Ag}(4,4'\text{-daps})] \cdot \text{ClO}_4\}_n$ (**5**) (3,3'-daps = di(3'-aminodiphenyl)sulfone, and 4,4'-daps = di(4'-aminodiphenyl)sulfone) have been synthesized and structural characterized by elemental analyses, IR spectra, powder X-ray diffraction (PXRD) and single-crystal X-ray diffraction analyses. Complex **1** displays a 1D ladder-like chain with four-connected Ag ions and bridged 3,3'-daps. Complex **2** shows other 1D ladder chain modified by tentacles. Complex **3** is a 2D layer structure with both Ag ions and 3,3'-daps are 3-connected nodes. Complex **4** is another 1D ladder chain with three-connected Ag ions and 4,4'-daps. Complex **5** shows a 2D 4^4-sql net with Ag ions and 4,4'-daps as 4-connected nodes. Moreover, their solid state luminescence and thermal stabilities also have been investigated.

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1. Introduction

The assembly of coordination polymers (CPs) with specific network topologies have attracted widespread research interest owing to their tremendous potential applications as functional solid materials [1–5]. The diverse structures of such materials are always dependent on many factors, such as metal ion, template, metal-ligand ratio, pH value, counteranion, and number of coordination sites provided by organic ligands [6–12]. Anions being a permanent part of the final product which can influence the structure directly by coordinating to the metal ions or by acting as templates to induce the self-assembly [13–17]. Therefore, make full use of different counteranions may cause significant structural change of the resultant polymers.

One outstanding aspect of the silver CPs is that they have the unique luminescence features, attracted researchers deeply investigated them. The good affinity of silver ions make the coordination has a dynamic range varied from two to six, even seven and eight, showing strong tendency to display vesatile coordination spheres [18–20]. This tendency partly attributed to the d^{10} electronic configuration of silver [21]. On the other hands, there are so many ligands were rational design and reasonable use to

compose functional coordination polymers with excellent structures and properties [22–24]. Compared with other ligands, the V-shaped ligands are holding particular angles, have diversity coordination modes [25,26]. The ligands with amino as terminal groups are still less reported. The di(aminodiphenyl)sulfone, have never introduced to the coordination chemistry before, proved by a CSD (Cambridge Structure Database) survey with the help of ConQuest version 1.3. The flexible backbones and the coordinated N_{amino} and $\text{O}_{\text{sulfone}}$ can ligate metal centres in different orientations, finally given cavities, interpenetration, helical structures, and other novel motifs with unique topologies. Besides, It can act as hydrogen-bond acceptor as well as donor, depending upon the degree of deprotonation, make the stable supramolecular possible.

Inspired by the above-mentioned points, we synthesized five novel silver coordination polymers by employing two V-shaped daps ligands reacted with AgX ($\text{X} = \text{BF}_4^-$, NO_3^- , ClO_4^- , CF_3SO_3^-). The results indicated that the counteranions are crucial factors for the formation of the different structures. In this paper, we reported the syntheses and characterizations of five novel coordination polymers, $\{[\text{Ag}(3,3'\text{-daps})_2] \cdot \text{BF}_4\}_n$ (**1**), $\{[\text{Ag}(3,3'\text{-daps})_2] \cdot \text{NO}_3\}_n$ (**2**), $[\text{Ag}(3,3'\text{-daps})(\text{CF}_3\text{SO}_3)]_n$ (**3**), $\{[\text{Ag}(4,4'\text{-daps})] \cdot \text{CF}_3\text{SO}_3\}_n$ (**4**) and $\{[\text{Ag}(4,4'\text{-daps})] \cdot \text{ClO}_4\}_n$ (**5**), which exhibit a systematic variation of architectures from 1D wave chain, 1D ladder chain, modified 1D chain, 2D 3-connected nets, to 2D 4^4-sql networks.

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Table 1
Crystal data for 1–5^a.

	1	2	3	4	5
Formula	C ₂₄ H ₂₄ AgBF ₄ N ₄ O ₄ S ₂	C ₂₄ H ₂₄ AgN ₅ O ₇ S ₂	C ₁₃ H ₁₂ AgF ₃ N ₂ O ₅ S ₂	C ₁₃ H ₁₂ AgF ₃ N ₂ O ₅ S ₂	C ₁₂ H ₁₂ AgClN ₂ O ₆ S
M _r	691.27	666.47	505.24	505.24	455.62
Crystal system	Monoclinic	Monoclinic	Monoclinic	Monoclinic	Orthorhombic
Space group	P2 ₁ /n	P2 ₁ /c	P2 ₁ /c	C2/m	Pnma
a (Å)	10.4437(12)	11.9717(11)	13.0649(16)	12.776(5)	19.644(3)
b (Å)	17.795(2)	28.770(3)	11.0845(14)	24.629(13)	13.5727(17)
c (Å)	14.4223(18)	8.0499(8)	12.8884(16)	6.529(3)	5.6780(7)
α (°)	90.00	90.00	90.00	90.00	90.00
β (°)	98.843(3)	109.511(3)	115.418(3)	118.154(5)	90.00
γ (°)	90.00	90.00	90.00	90.00	90.00
V (Å ³)	2648.4(6)	2613.4(4)	1685.8(4)	1811.4(14)	1509.4(3)
Z	4	4	4	4	4
ρ (g cm ⁻³)	1.734	1.694	1.991	1.853	2.005
μ (mm ⁻¹)	0.987	0.986	1.504	1.399	1.683
T (K)	293(2)	293(2)	293(2)	293(2)	293(2)
R _{int}	0.0288	0.0387	0.0423	0.0312	0.0352
R [I > 2σ(I)]	R ₁ = 0.0332 wR ₂ = 0.0850	R ₁ = 0.0586 wR ₂ = 0.1431	R ₁ = 0.0389 wR ₂ = 0.0966	R ₁ = 0.0407 wR ₂ = 0.1076	R ₁ = 0.0506 wR ₂ = 0.1350
R (all data)	R ₁ = 0.0398 wR ₂ = 0.0977	R ₁ = 0.0688 wR ₂ = 0.1471	R ₁ = 0.0418 wR ₂ = 0.0984	R ₁ = 0.0508 wR ₂ = 0.1128	R ₁ = 0.0526 wR ₂ = 0.1362
Gof	1.058	1.089	1.084	1.078	1.095

$$^a R_1 = \sum |F_o| - |F_c| / \sum |F_o|, wR_2 = [\sum w(F_o^2 - F_c^2)^2] / \sum w(F_o^2)^2]^{1/2}.$$

Table 2
Selected bond lengths (Å) and angles (°) for 1–5.

Complex 1							
N3–Ag1	2.398(2)	N4–Ag1 ⁱⁱⁱ	2.451(2)	N1–Ag1	2.432(2)	N2–Ag1	2.333(2)
Ag1–N4 ⁱⁱⁱ	2.451(2)	N2–Ag1–N3	108.11(8)	N2–Ag1–N1	109.62(9)	N3–Ag1–N1	120.28(8)
N2–Ag1–N4 ⁱⁱⁱ	119.14(8)	N3–Ag1–N4 ⁱⁱⁱ	99.99(8)	N1–Ag1–N4 ⁱⁱⁱ	99.96(8)		
Symmetry code: (i) x–1, y, z; (ii) x+1, y, z; (iii) –x+1, –y, –z+2.							
Complex 2							
N1–Ag1	2.330(5)	N3–Ag1	2.408(5)	N4–Ag1 ⁱ	2.446(5)	Ag1–N4 ⁱ	2.446(5)
N1–Ag1–N3	145.06(17)	N1–Ag1–N4 ⁱ	104.75(15)	N3–Ag1–N4 ⁱ	94.98(15)		
Symmetry code: (i) –x+1, –y+2, –z+2.							
Complex 3							
N1–Ag1	2.274(3)	N2–Ag1	2.268(3)	O1–Ag1 ⁱⁱ	2.538(2)	Ag1–O1 ⁱⁱⁱ	2.538(2)
N2–Ag1–N1	136.91(11)	N2–Ag1–O1 ⁱⁱⁱ	106.46(9)	N1–Ag1–O1 ⁱⁱⁱ	106.16(10)		
Symmetry codes: (i) –x, –y, –z+2; (ii) –x, y+1/2, –z+3/2; (iii) –x, y–1/2, –z+3/2.							
Complex 4							
Ag1–N1 ⁱ	2.219(4)	Ag1–N1	2.219(4)	N1 ⁱ –Ag1–N1	156.5(2)		
Symmetry codes: (i) –x+2, y, –z+2; (ii) –x+1, y, –z+2; (iii) x, –y+1, z.							
Complex 5							
O1–Ag1 ⁱ	2.492(5)	O1–Ag1 ⁱ	2.422(6)	O2–Ag1 ⁱⁱ	2.427(6)	O2–Ag1 ⁱⁱⁱ	2.774(11)
Ag1 ⁱ –N1 ⁱⁱⁱ	2.238(5)	Ag1 ⁱ –N1	2.238(5)	Ag1 ⁱ –O1 ⁱⁱ	2.422(6)	Ag1 ⁱ –O2 ⁱ	2.774(11)
Ag1–N1 ⁱⁱⁱ	2.384(5)	Ag1–N1	2.384(5)	Ag1–O2 ⁱ	2.427(6)	Ag1–O1 ⁱⁱ	2.492(5)
N1 ⁱⁱⁱ –Ag1 ⁱ –N1	124.0(4)	N1 ⁱⁱⁱ –Ag1 ⁱ –O1 ⁱⁱ	106.3(2)	N1–Ag1 ⁱ –O1 ⁱⁱ	106.3(2)	N1 ⁱⁱⁱ –Ag1 ⁱ –O2 ⁱ	114.1(3)
N1–Ag1 ⁱ –O2 ⁱ	114.1(3)	O1 ⁱⁱ –Ag1 ⁱ –O2 ⁱ	82.6(2)	N1 ⁱⁱⁱ –Ag1–N1	112.0(2)	N1 ⁱⁱⁱ –Ag1–O2 ⁱ	122.45(14)
N1–Ag1–O2 ⁱ	122.45(14)	N1 ⁱⁱⁱ –Ag1–O1 ⁱ	99.81(16)	N1–Ag1–O1 ⁱⁱ	99.81(16)	O2 ⁱ –Ag1–O1 ⁱⁱ	88.71(18)
Symmetry codes: (i) –x+1/2, –y, z–1/2; (ii) –x+1/2, –y, z+1/2; (iii) x, –y+1/2, z; (iv) x, –y–1/2, z.							

2. Experimental

2.1. Materials and methods

Chemical reagents were purchased commercially and were used as received without further purification. Elemental analyses of C, N, and H were performed on an EA1110 CHNS-0 CE elemental analyzer. IR (KBr pellet) spectra were recorded on a Nicolet Magna 750FT-IR spectrometer. Fluorescent data were collected on an Edinburgh FLS920 TCSPC system. Thermogravimetric measurements were carried out in a nitrogen stream using a Netzsch STA449C apparatus with a heating rate of 10 °C min⁻¹. X-Ray powder diffraction (XRPD) was carried out on a RIGAKU DMAX2500 apparatus. Solid UV–visible spectra were obtained in the 200–800 nm range on a JASCOVIDEC-660

spectrophotometer. Fluorescent data were collected on an Edinburgh FLS920 TCSPC system.

2.2. Synthesis of complexes 1–5

2.2.1. Synthesis of {[Ag(3,3'-daps)₂]-BF₄]_n (1)

AgBF₄ (19.7 mg, 0.1 mmol) in ethanol (20 mL) was added dropwise with stirring to 3,3'-daps (24.8 mg, 0.1 mmol) in ethanol (20 mL) and was continued at room temperature for several days. Slow evaporation of this solution yielded colorless block crystals that proved suitable for X-ray analysis. The precipitate that formed was collected by filtration, and dried at room temperature to give **1** in 65% (33.1 mg) yield based on Ag. Anal. (%) calcd. for C₁₃H₁₂AgF₃N₂O₅S₂: C, 41.70; H, 1.56; N, 8.10. Found: C, 41.29; H, 1.79; N, 8.01. IR (KBr pellet, cm⁻¹): 3459(m), 3138(s), 1609(m),

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