Supplementary Information

$K_2SO_4(NH_3SO_3)_2$ and $Cs_2SO_4(NH_3SO_3)_3$: Two Deep-UV Transparent Sulfamate-Sulfates with Moderate Birefringence

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Table S1. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $K_2SO_4(NH_3SO_3)_2$. U(eq) is defined as one-third of the trace of the orthogonalized U_{ij} tensor

Atom	x	y	z	U(eq)
K(1)	7404(4)	3630(1)	4196(2)	65(1)
K(2)	2459(3)	4687(1)	1991(1)	46(1)
S(1)	7446(2)	2361(1)	7181(1)	28(1)
S(2)	7443(2)	4032(1)	339(1)	28(1)
S(3)	12506(3)	4220(1)	6229(1)	38(1)
O(1)	8429(13)	1678(4)	7552(6)	90(2)
O(2)	8539(14)	2889(4)	7950(6)	114(3)
O(3)	7861(8)	2456(3)	5854(4)	49(11)
O(4)	4948(8)	2350(3)	7431(5)	60(1)
O(5)	10514(10)	4662(3)	6372(5)	67(2)
O(6)	14712(10)	4565(3)	6338(5)	70(2)
O(7)	12336(10)	3696(3)	5256(4)	61(2)
O(8)	7434(8)	4229(2)	1634(4)	44(1)
O(9)	5351(8)	4225(2)	-352(4)	46(1)
O(10)	9618(8)	4181(2)	-269(4)	43(1)
N(1)	12467(9)	3687(3)	7600(5)	36(1)
N(2)	7324(8)	3100(2)	348(4)	26(1)

Table S2. Atomic coordinates ($\times 10^4$) and equivalent isotropic displacement parameters ($Å^2 \times 10^3$) for $Cs_2SO_4(NH_3SO_3)_3$. U(eq) is defined as one-third of the trace of the orthogonalized U_{ij} tensor

Atom	x	y	z	U(eq)
Cs(1)	6667	3333	9635(1)	42(1)
S(1)	10113(4)	6256(4)	7500	26(1)
S(2)	10000	10000	10000	94(3)
O(1)	8371(11)	6000(12)	7500	35(2)
O(2)	10607(10)	5697(10)	8480(6)	56(2)
O(3)	10000	10000	8944(16)	59(6)
O(4)	10530(40)	12080(30)	9644(18)	113(10)
N(1)	11490(20)	8545(19)	7500	28(2)

Table S3. Selected bond lengths (Å) and angles (degrees) for $K_2SO_4(NH_3SO_3)_2$

K(2)-O(10)#5	3.022(5)	K(2)-O(6)#7	2.752(5)
K(2)-O(9)#5	2.966(5)	K(2)-O(8)	2.976(5)
K(2)-O(9)	3.135(5)	K(2)-O(8)#2	2.989(5)
K(1)-O(1)#3	2.885(7)	S(1)-O(3)	1.449(5)
K(1)-O(3)	2.823(5)	S(1)-O(4)	1.445(5)
K(1)-O(4)#4	3.011(6)	S(2)-O(8)	1.429(4)
K(1)-O(6)#2	3.274(6)	S(2)-O(9)	1.429(5)
K(1)-O(7)#2	3.109(6)	S(2)-O(10)	1.430(4)
K(1)-O(7)	2.995(6)	S(3)-N(1)	1.767(6)
K(1)-O(8)	2.951(5)	S(3)-O(5)	1.409(5)
S(3)-O(6)	1.409(5)	S(1)-O(2)	1.416(6)
S(3)-O(7)	1.426(5)	S(2)-N(2)	1.740(5)
K(2)-O(1)#3	2.671(8)	S(1)-O(1)	1.442(6)
K(2)-O(5)#6	2.733(5)		
O(9)#5-K(2)-O(8)#2	121.92(13)	O(8)#2-K(2)-O(10)#2	47.02(12)
O(8)-K(1)-O(7)	108.16(15)	O(8)-K(2)-O(10)#5	118.77(13)

O(8)-K(1)-O(6)#2	117.03(15)	O(5)-S(3)-O(7)	115.9(4)
O(8)-K(1)-O(4)#4	68.93(14)	O(5)-S(3)-O(6)	115.7(4)
O(9)#5-K(2)-O(8)	73.32(13)	O(6)#7-K(2)-O(9)	110.38(15)
O(9)#5-K(2)-O(9)	59.17(14)	O(6)#7-K(2)-O(9)#5	77.84(15)
O(1)#3-K(1)-O(4)#4	82.92(18)	O(6)#7-K(2)-O(10)#2	165.56(15)
O(1)#3-K(1)-O(6)#2	99.27(18)	O(6)#7-K(2)-O(10)#5	104.72(16)
O(1)#3-K(1)-O(7)#2	60.86(16)	O(8)-K(2)-O(10)#2	107.40(12)
O(1)#3-K(1)-O(7)	161.58(18)	O(8)#2-K(2)-O(9)	109.33(12)
O(1)#3-K(1)-O(8)	62.07(16)	O(8)#2-K(2)-O(10)#5	76.33(13)
O(9)#5-K(2)-O(10)#2	88.07(12)	O(5)#6-K(2)-O(10)#5	80.79(14)
O(9)#5-K(2)-O(10)#5	47.64(12)	O(10)#5-K(2)-O(9)	84.90(12)
O(8)-K(2)-O(8)#2	143.44(16)	O(5)#6-K(2)-O(9)	165.69(15)
O(3)-K(1)-O(1)#3	106.66(18)	O(5)#6-K(2)-O(9)#5	110.00(16)
O(3)-K(1)-O(4)#4	83.16(15)	O(5)#6-K(2)-O(10)#2	108.82(16)
O(3)-K(1)-O(6)#2	90.87(15)	O(10)#2-K(2)-O(9)	63.72(12)
O(3)-K(1)-O(7)#2	82.94(15)	O(10)#5-K(2)-O(10)#2	62.53(14)
O(3)-K(1)-O(7)	73.88(16)	O(5)#6-K(2)-O(8)#2	67.13(16)
O(3)-K(1)-O(8)	150.59(15)	O(5)#6-K(2)-O(8)	143.70(16)
O(8)-K(2)-O(9)	46.23(12)	O(5)#6-K(2)-O(6)#7	73.74(19)
O(7)-K(1)-O(4)#4	78.85(14)	O(7)-S(3)-N(1)	102.4(3)
O(6)#7-K(2)-O(8)#2	140.22(15)	O(1)-S(1)-O(3)	107.9(3)
O(4)#4-K(1)-O(6)#2	174.01(16)	O(1)-S(1)-O(4)	108.1(4)
O(4)#4-K(1)-O(7)#2	135.06(15)	O(2)-S(1)-O(1)	107.1(5)
O(7)#2-K(1)-O(6)#2	99.13(14)	O(2)-S(1)-O(3)	113.7(4)
O(7)-K(1)-O(7)#2	136.25(17)	O(2)-S(1)-O(4)	108.7(4)
O(6)#7-K(2)-O(8)	71.78(16)	O(4)-S(1)-O(3)	111.1(3)
O(10)-S(2)-N(2)	103.3(2)	O(9)-S(2)-O(10)	115.5(3)
O(1)#3-K(2)-O(5)#6	114.05(19)	O(9)-S(2)-N(2)	102.9(2)
O(1)#3-K(2)-O(6)#7	102.9(2)	O(6)-S(3)-O(7)	114.9(3)
O(1)#3-K(2)-O(8)#2	86.97(18)	O(6)-S(3)-N(1)	102.4(3)
O(1)#3-K(2)-O(8)	64.14(18)	O(5)-S(3)-N(1)	102.6(3)
O(1)#3-K(2)-O(9)#5	134.25(18)	O(8)-S(2)-N(2)	104.5(3)
O(1)#3-K(2)-O(9)	78.92(17)	O(8)-S(2)-O(9)	114.5(3)
O(1)#3-K(2)-O(10)#5	151.44(17)	O(8)-S(2)-O(10)	114.5(3)
O(1)#3-K(2)-O(10)#2	89.12(16)		

Symmetry transformations used to generate equivalent atoms:
#1x+1,y,z#2x-1,y,z#3 x-1/2,-y+1/2,z-1/2,#4x+1/2,-y+1/2,z-1/2#5-x+1,-y+1,-z#6-x+1,-y+1,-z+1#7 -x+2,-y+1,-z+1,#8x+1/2,-y+1/2,z+1/2#9 x-1/2,-y+1/2,z+1/2

Table S4. Selected bond lengths (Å) and angles (degrees) for $Cs_2SO_4(NH_3SO_3)_3$

Cs(1)-O(1)#1	3.309(6)	S(1)-O(2)#9	1.436(7)
Cs(1)-O(1)	3.309(6)	S(1)-N(1)	1.743(16)
Cs(1)-O(1)#2	3.309(6)	S(2)-O(4)	1.69(2)
Cs(1)-O(2)#3	3.107(7)	O(4)-O(4)#3	1.18(4)
Cs(1)-O(2)	3.314(8)	S(1)-O(2)	1.436(7)
Cs(1)-O(2)#4	3.107(7)	S(2)-O(3)#10	1.287(19)
Cs(1)-O(2)#2	3.314(8)	S(2)-O(3)	1.287(19))
Cs(1)-O(2)#1	3.314(8)	S(2)-O(4)#11	1.69(2)
Cs(1)-O(2)#5	3.107(7)	S(2)-O(4)#12	1.69(2)
Cs(1)-O(4)#6	3.61(3)	S(2)-O(4)#3	1.69(2)
Cs(1)-O(4)#7	3.61(3)	S(2)-O(4)#10	1.69(2)
Cs(1)-O(4)#8	3.61(3)	S(2)-O(4)#8	1.69(2)
S(1)-O(1)	1.422(8)		
O(1)#2-Cs(1)-O(1)#1	64.65(19)	O(4)#7-Cs(1)-O(4)#8	114.3(3)
O(1)#2-Cs(1)-O(1)	64.65(19)	O(4)#6-Cs(1)-O(4)#7	114.3(3)
O(1)#1-Cs(1)-O(1)	64.6(2)	Cs(1)-O(1)-Cs(1)#9	103.7(2)

O(1)#2-Cs(1)-O(2)	61.8(2)	Cs(1)#13-O(2)-Cs(1)	105.7(2)
O(1)-Cs(1)-O(2)#1	61.8(2)	O(2)#1-Cs(1)-O(4)#8	69.8(4)
O(1)-Cs(1)-O(2)	42.74(18)	O(1)-S(1)-O(2)	115.2(4)
O(1)#1-Cs(1)-O(2)	101.75(17)	O(1)-S(1)-O(2)#9	115.2(4)
O(1)#2-Cs(1)-O(2)#2	42.74(18)	O(1)-S(1)-N(1)	104.4(6)
O(1)#2-Cs(1)-O(2)#1	101.75(17)	O(2)#3-Cs(1)-O(4)#8	58.2(4)
O(1)#1-Cs(1)-O(2)#2	61.8(2)	O(4)#6-Cs(1)-O(4)#8	114.3(3)
O(1)-Cs(1)-O(2)#2	101.75(17)	O(4)#3-O(4)-S(2)	69.6(8)
O(1)#1-Cs(1)-O(2)#1	42.74(17)	O(4)#3-O(4)-Cs(1)#14	102.6(17)
O(1)#1-Cs(1)-O(4)#8	109.4(4)	O(2)-S(1)-O(2)#9	112.6(7)
O(1)-Cs(1)-O(4)#6	109.4(4)	O(2)#9-S(1)-N(1)	103.8(4)
O(1)-Cs(1)-O(4)#7	128.7(4)	O(2)-S(1)-N(1)	103.8(4)
O(1)#2-Cs(1)-O(4)#8	128.7(4)	O(2)#1-Cs(1)-O(4)#6	168.9(3)
O(1)#1-Cs(1)-O(4)#7	67.5(4)	O(2)#3-Cs(1)-O(4)#7	118.2(4)
O(1)#2-Cs(1)-O(4)#6	67.5(4)	O(2)-Cs(1)-O(4)#7	168.9(3)
O(1)#1-Cs(1)-O(4)#6	128.7(4)	O(2)-Cs(1)-O(4)#8	71.0(4)
O(1)-Cs(1)-O(4)#8	67.5(4)	O(2)#2-Cs(1)-O(4)#6	71.0(4)
O(1)#2-Cs(1)-O(4)#7	109.4(4)	O(4)#11-S(2)-O(4)#12	113.6(6)
O(2)#3-Cs(1)-O(1)#2	120.0(2)	O(4)#11-S(2)-O(4)#8	40.8(15)
O(2)#4-Cs(1)-O(1)	120.0(2)	O(4)-S(2)-O(4)#3	40.8(15)
O(2)#5-Cs(1)-O(1)#2	105.7(2)	O(2)#4-Cs(1)-O(4)#8	61.0(4)
O(2)#4-Cs(1)-O(1)#1	105.7(2)	O(2)#5-Cs(1)-O(4)#6	58.2(4)
O(2)#5-Cs(1)-O(1)	167.3(2)	O(2)#3-Cs(1)-O(4)#6	61.0(4)
O(2)#3-Cs(1)-O(1)	105.7(2)	O(4)-S(2)-O(4)#10	95.5(19)
O(2)#4-Cs(1)-O(1)#2	167.3(2)	O(4)-S(2)-O(4)#11	113.6(6)
O(2)#5-Cs(1)-O(2)	126.30(12)	O(4)#8-S(2)-O(4)#12	95.5(19)
O(2)#5-Cs(1)-O(2)#2	72.9(2)	O(3)#10-S(2)-O(3)	180.0
O(2)#3-Cs(1)-O(2)#4	71.3(2)	O(3)#10-S(2)-O(4)	104.9(8)
O(2)#2-Cs(1)-O(2)#1	103.24(15)	O(3)-S(2)-O(4)#12	75.1(8)
O(2)#5-Cs(1)-O(2)#4	71.3(2)	O(3)-S(2)-O(4)	75.1(8)
O(2)#2-Cs(1)-O(2)	103.24(15)	O(3)#10-S(2)-O(4)#10	75.1(8)
O(2)#4-Cs(1)-O(2)	130.17(7)	O(3)#10-S(2)-O(4)#8	75.1(8)
O(2)#3-Cs(1)-O(2)#2	130.17(7)	O(3)-S(2)-O(4)#11	75.1(8)
O(2)#4-Cs(1)-O(2)#1	72.9(2)	O(3)-S(2)-O(4)#3	104.9(8)
O(2)#3-Cs(1)-O(2)#1	126.30(11)	O(3)#10-S(2)-O(4)#12	104.9(8)
O(2)#5-Cs(1)-O(2)#3	71.3(2)	O(3)-S(2)-O(4)#10	104.9(8)
O(2)#5-Cs(1)-O(2)#1	130.17(7)	O(3)#10-S(2)-O(4)#11	104.9(8)
O(2)#4-Cs(1)-O(2)#2	126.30(12)	O(3)-S(2)-O(4)#8	104.9(8)
O(2)#3-Cs(1)-O(2)	72.9(2)	O(3)#10-S(2)-O(4)#3	75.1(8)
O(2)-Cs(1)-O(2)#1	103.24(15)	O(4)#8-S(2)-O(4)#3	113.6(6)
O(2)#5-Cs(1)-O(4)#7	61.0(4)	O(4)#10-S(2)-O(4)#8	113.6(6)
O(2)#5-Cs(1)-O(4)#8	118.2(4)	O(4)#10-S(2)-O(4)#3	113.6(6)
O(2)-Cs(1)-O(4)#6	69.8(4)	O(4)#11-S(2)-O(4)#10	149.3(18)
O(2)#1-Cs(1)-O(4)#7	71.0(4)	O(4)-S(2)-O(4)#12	113.6(6)
O(2)#4-Cs(1)-O(4)#7	58.2(4)	O(4)#11-S(2)-O(4)#3	95.5(19)
O(2)#2-Cs(1)-O(4)#7	69.8(4)	O(4)#3-S(2)-O(4)#12	149.3(19)
O(2)#4-Cs(1)-O(4)#6	118.2(4)	O(4)-S(2)-O(4)#8	149.3(18)
O(2)#2-Cs(1)-O(4)#8	168.9(3)	O(4)#10-S(2)-O(4)#12	40.8(15)

Symmetry transformations used to generate equivalent atoms:
#1-y+1,x-y,z #2 -x+y+1,-x+1,z#3 -x+2,-x+y+1,-z+2#4x-y,-y+1,-z+2#5y,x-1,-z+2#6 -x+2,-x+y,-z+2#7y-1,x-1,-z+2#8x-y+1,-y+2,-z+2#9 x,y,-z+3/2#10 y,x,-z+2#11-y+2,x-y+1,z#12-x+y+1,-x+2,z#13y+1,x,-z+2#14y+1,x+1,-z+2.

Table S5. Anisotropic displacement parameters ($Å^2 \times 10^3$) for $K_2SO_4(NH_3SO_3)_2$. The anisotropic displacement factor exponent takes the form: $-2\pi^2 [h^2 a^{*2} U11 + ... + 2hka^* b^* U12]$

	U11	U22	U33	U23	U13	U12

K(1)	70(1)	74(1)	51(1)	9(1)	0(1)	2(1)
K(2)	31(1)	58(1)	48(1)	-13(1)	0(1)	1(1)
S(1)	22(1)	38(1)	25(1)	-6(1)	2(1)	-2(1)
S(2)	27(1)	28(1)	29(1)	-6(1)	1(1)	-1(1)
S(3)	34(1)	53(1)	28(1)	-14(1)	2(1)	-4(1)
O(1)	108(5)	94(4)	67(4)	23(3)	12(3)	53(4)
O(2)	127(6)	149(6)	70(4)	-64(4)	54(4)	-98(5)
O(3)	46(3)	69(3)	32(2)	4(2)	5(2)	11(2)
O(4)	28(3)	103(4)	49(3)	-2(3)	5(2)	-5(3)
O(5)	73(4)	85(4)	43(3)	0(3)	9(3)	34(3)
O(6)	63(3)	99(4)	46(3)	-1(3)	2(2)	-43(3)
O(7)	66(3)	81(4)	35(3)	-29(2)	6(2)	-8(3)
O(8)	50(3)	48(3)	32(2)	-16(2)	4(2)	-5(2)
O(9)	46(3)	34(2)	57(3)	-4(2)	-17(2)	4(2)
O(10)	44(3)	37(2)	50(3)	-6(2)	19(2)	-11(2)
N(1)	26(3)	46(3)	38(3)	-12(2)	5(2)	-3(2)
N(2)	25(2)	27(2)	26(2)	0(2)	2(2)	-3(2)

Table S6. Anisotropic displacement parameters ($Å^2×10^3$) for $Cs_2SO_4(NH_3SO_3)_3$. The anisotropic displacement factor exponent takes the form: $-2\pi^2$ [h^2 a^{*2} U11 + ... + 2hka* b*U12].

	U11	U22	U33	U23	U13	U12
Cs(1)	52(1)	52(1)	22(1)	0	0	26(1)
S(1)	20(1)	22(2)	36(2)	0	0	10(1)
S(2)	132(5)	132(5)	17(3)	0	0	66(3)
O(1)	22(5)	35(5)	49(5)	0	0	16(4)
O(2)	41(4)	43(5)	72(5)	26(4)	-20(4)	11(3)
O(3)	80(10)	80(10)	18(9)	0	0	40(5)
O(4)	117(12)	111(11)	112(11)	14(6)	6(6)	58(7)
N(1)	29(7)	20(7)	35(5)	0	0	12(4)

Table S7. Hydrogen coordinates (Å×104) and isotropic displacement parameters (Å2×103) for $K_2SO_4(NH_3SO_3)_2$

	x	y	z	U(eq)
H(1A)	13618	3363	7572	44
H(1B)	11081	3465	7647	44
H(1C)	12682	3965	8271	44
H(2A)	7769	2933	-393	31
H(2B)	5853	2959	493	31
H(2C)	8287	2932	948	31

Table S8. Hydrogen coordinates (Å×104) and isotropic displacement parameters (Å2×103) for $Cs_2SO_4(NH_3SO_3)_3$.

	x	y	z	U(eq)
H(1A)	11432	8975	8152	34
H(1B)	11132	9018	6982	34
H(1C)	12593	8810	7366	34

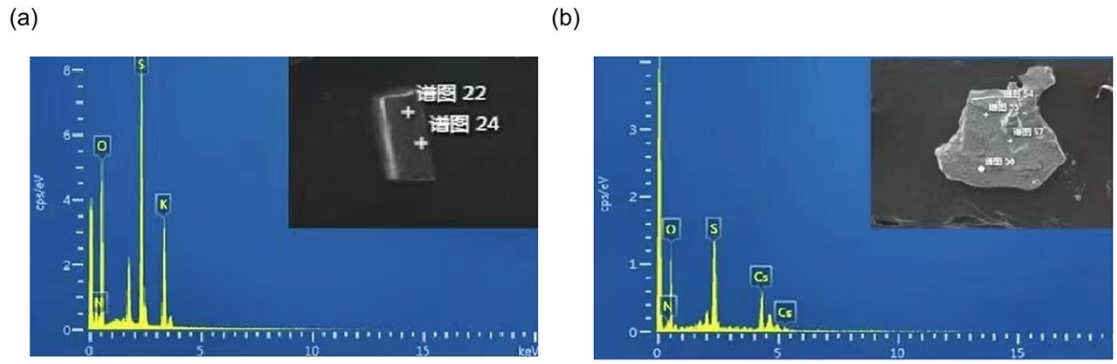

Figure S1. Energy-dispersive X-ray spectroscope (EDS) analysis for $K_2SO_4(NH_3SO_3)_2$ (a) and $Cs_2SO_4(NH_3SO_3)_3$ (b).

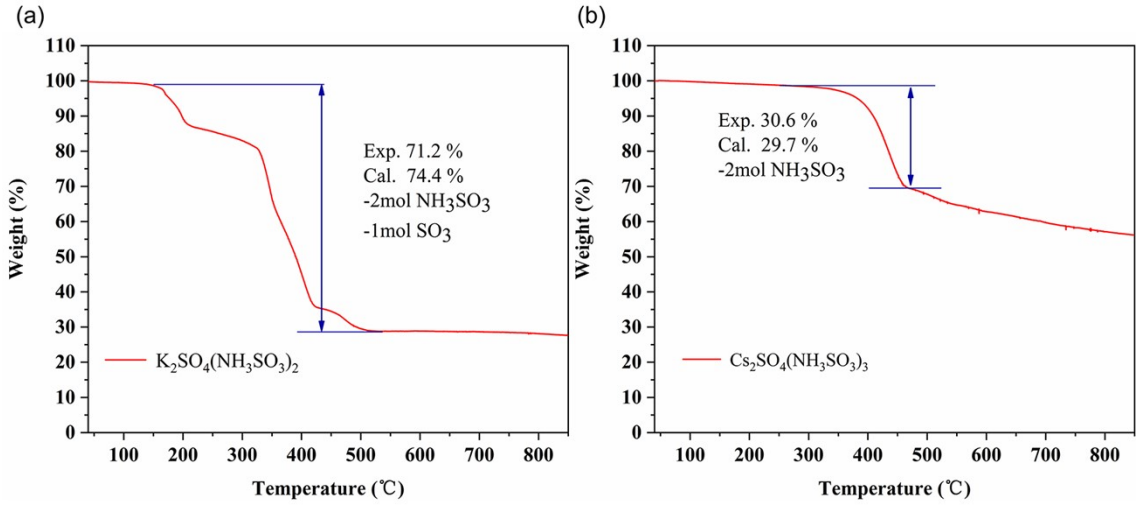

Figure S2. TG analysis for $K_2SO_4(NH_3SO_3)_2$ (a) and $Cs_2SO_4(NH_3SO_3)_3$ (b).

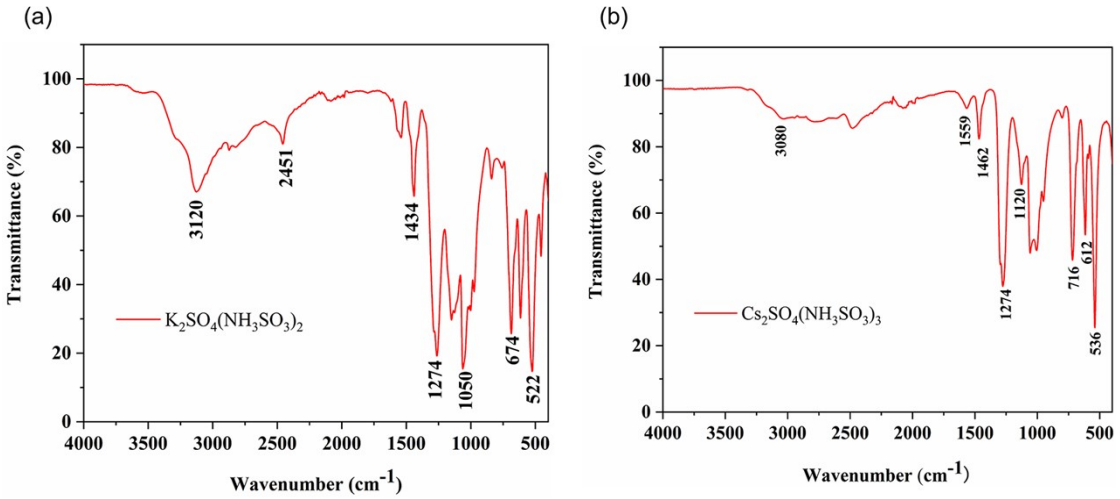

Figure S3. IR spectra of $K_2SO_4(NH_3SO_3)_2$ (a) and $Cs_2SO_4(NH_3SO_3)_3$ (b).

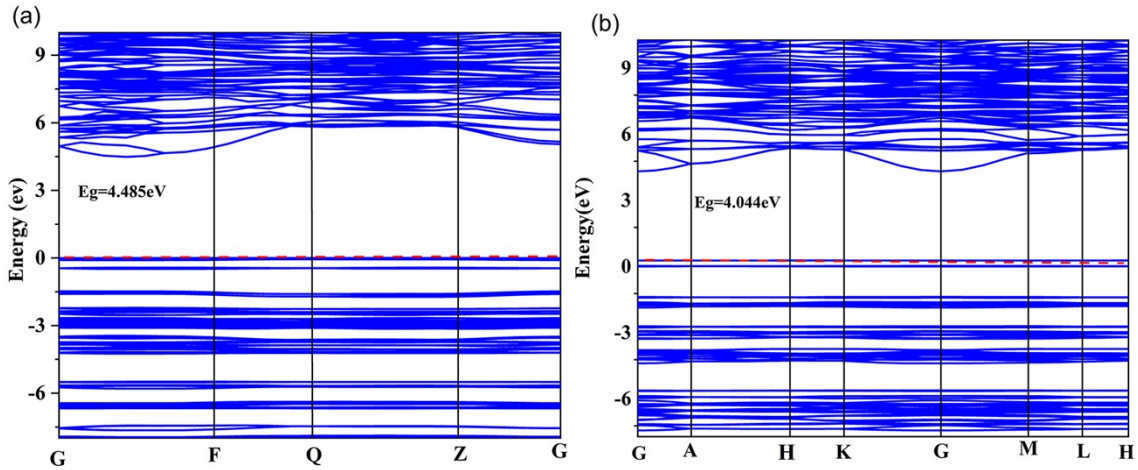

Figure S4. Band gaps of $K_2SO_4(NH_3SO_3)_2$ (a) and $Cs_2SO_4(NH_3SO_3)_3$ (b).