Supplementary Information (SI)

$M_3MoO_4(HCO_3)$ (M = Rb, Cs): Cation-tuned synthesis of two noncentrosymmetric molybdate-carbonates with isolated $[MoO_4]$ and $[HCO_3]$ groups

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Table S1. Crystal data and structure refinement for $Rb_3MoO_4(HCO_3)$ and $Cs_3MoO_4(HCO_3)$.

Formula	$Rb_3MoO_4(HCO_3)$	$Cs_3MoO_4(HCO_3)$
Formula weight	477.37	619.69
Temperature / K	298.15	293.0
Crystal system	Orthorhombic	Orthorhombic
Space group	$Pna2_1$	$Pna2_1$
a / Å	16.4966(7)	17.2074(7)
b / Å	6.2117(3)	6.4605(2)
c / Å	17.7267(10)	18.3641(6)
α / °	90	90
β / °	90	90
γ / °	90	90
Volume / Å^3	1816.49(16)	2041.51(12)
Z	8	8
ρ_{calc} / Mg cm^{-3}	3.491	4.032
μ / mm^{-1}	17.414	11.826
F (000)	1728	2160
Radiation	Mo Kα (λ = 0.71073)	Mo Kα (λ = 0.71073)
2θ range for data collection/ °	4.596 to 55.012	4.436 to 55.018
Index ranges	$-21 \le h \le 20$, $-8 \le k \le 8$, $-23 \le l \le 22$	$-22 \le h \le 22$, $-8 \le k \le 8$, $-23 \le l \le 23$
Reflections collected / unique	33213 / 4164 [R(int) = 0.0988]	49951 / 4642 [R(int) = 0.0663]
Completeness / %	99.9	99.9
Data / restraints / parameters	4164 / 3 / 227	4642 / 9 / 227
Goodness-of-fit on F^2	1.035	1.130
Final R indices [$I \ge 2\sigma(I)$][a]	R_1= 0.0340, wR_2= 0.0759	R_1= 0.0196, wR_2= 0.0429
Final R indices (all data)[a]	R_1= 0.0421, wR_2= 0.0807	R_1= 0.0227, wR_2= 0.0452
Largest diff. peak / hole /e Å^{-3}	1.319 / - 0.743	0.971 / - 0.730

[a] $R_1 = \Sigma||F_o| - |F_c||/\Sigma|F_o|$ and $wR_2 = [\Sigma w (F_o^2 - F_c^2)^2 / \Sigma w F_o^4]^{1/2}$ for $F_o^2 > 2\sigma(F_o^2)$

Table S2 Atomic coordinates (× 10^4), equivalent isotropic displacement parameters (Å^2 × 10^3), and bond valence sums (BVS) for $Rb_3MoO_4(HCO_3)$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor.

Atoms	x	y	z	U_{eq}	BVS
Rb(1)	8740(1)	2940(2)	3633(1)	31(1)	0.88
Rb(2)	7286(1)	2669(2)	5410(2)	31(1)	0.95
Rb(3)	4636(1)	-2615(2)	2295(1)	25(1)	1.19
Rb(4)	5199(1)	-2531(2)	4594(2)	31(1)	1.07
Rb(5)	2867(1)	2248(2)	7720(1)	24(1)	1.20
Rb(6)	-1250(1)	7985(2)	6368(1)	30(1)	0.89
Mo(1)	6062(1)	2463(1)	3583(1)	18(1)	6.08
Mo(2)	1433(1)	7285(1)	6437(1)	18(1)	6.38
C(1)	4212(7)	1882(19)	5745(6)	20(2)	4.12
C(2)	-1731(8)	8134(18)	4270(6)	22(2)	3.97
O(1)	7104(7)	1962(15)	3538(9)	50(3)	1.98
O(2)	5815(7)	4706(16)	3031(8)	54(3)	2.00
O(3)	5536(6)	192(14)	3250(7)	42(3)	2.00
O(4)	5779(10)	2911(17)	4522(11)	48(3)	1.86
O(5)	4881(6)	3131(14)	5717(5)	27(2)	2.39
O(6)	4073(4)	618(12)	5213(4)	30(2)	1.98
O(7)	3806(7)	2086(14)	6324(9)	33(3)	2.15
O(8)	1902(8)	5123(17)	6860(9)	57(3)	2.12
O(9)	1755(8)	9667(18)	6833(9)	67(4)	2.07
O(10)	1650(13)	7276(16)	5458(13)	62(4)	1.78
O(11)	397(8)	7030(17)	6527(11)	64(4)	2.03
O(12)	-1566(4)	9397(12)	4791(4)	30(2)	1.99
O(13)	-1302(7)	7805(13)	3678(10)	31(3)	1.98
O(14)	-2404(6)	6929(15)	4320(5)	27(2)	2.39
H(5)	5110(70)	2760(140)	5310(40)	0(30)	/
H(14)	-2660(70)	7080(170)	4730(40)	10(30)	/

Table S3 Atomic coordinates (× 10^4), equivalent isotropic displacement parameters (Å^2 × 10^3), and bond valence sums (BVS) for $Cs_3MoO_4(HCO_3)$. U_{eq} is defined as one-third of the trace of the orthogonalized U_{ij} tensor.

Atoms	x	y	z	U_{eq}	BVS
Cs(1)	7094(1)	7672(1)	2256(1)	26(1)	1.16
Cs(2)	11241(1)	2172(1)	3601(1)	29(1)	0.91
Cs(3)	7729(1)	7626(1)	4596(1)	28(1)	1.02
Cs(4)	9774(1)	2469(1)	5397(1)	29(1)	0.97
Cs(5)	1272(1)	7158(1)	6401(1)	30(1)	0.85
Cs(6)	5404(1)	2624(1)	7727(1)	26(1)	1.15
Mo(1)	8524(1)	2682(1)	3565(1)	19(1)	6.26
Mo(2)	3978(1)	7550(1)	6417(1)	19(1)	6.03
C(1)	6743(6)	3063(15)	5709(5)	26(2)	3.97
C(2)	5786(5)	8042(14)	4293(5)	20(2)	4.05
O(1)	6169(7)	7746(10)	3728(8)	33(3)	2.09
O(2)	5117(4)	6890(12)	4340(5)	32(2)	2.31
O(3)	5940(3)	9273(9)	4787(3)	30(1)	2.02
O(4)	8109(6)	4850(14)	3172(7)	43(2)	2.10
O(5)	8188(6)	430(14)	3175(7)	50(2)	2.11
O(6)	9538(7)	2809(12)	3532(8)	47(3)	1.92
O(7)	8237(12)	2643(10)	4489(9)	50(4)	1.79
O(8)	6555(3)	4299(9)	5211(3)	32(1)	1.92
O(9)	6368(7)	2732(9)	6297(8)	28(2)	1.98
O(10)	7387(4)	1938(13)	5641(4)	32(2)	2.43
O(11)	2988(7)	7836(13)	6508(9)	58(3)	2.03
O(12)	4244(9)	7373(10)	5494(7)	38(3)	1.82
O(13)	4420(5)	9739(14)	6830(7)	42(2)	1.91
O(14)	4271(6)	5316(14)	6900(7)	49(2)	1.93
H(2)	4840(80)	7470(130)	4670(60)	30(40)	/
H(10)	7530(120)	2590(180)	5260(50)	80(90)	/

Table S4 Bond distances [Å] and angles [°] for $Rb_3MoO_4(HCO_3)$.

C(1)-O(5)	1.351(14)	O(1)-Rb(1)-O(3)#4	162.2(4)
C(1)-O(6)	1.248(12)	O(1)-Rb(1)-O(6)#4	107.6(3)
C(1)-O(7)	1.231(18)	O(1)-Rb(1)-O(8)#10	74.2(4)
C(2)-O(12)	1.243(13)	O(1)-Rb(1)-O(12)#16	73.9(3)
C(2)-O(13)	1.281(19)	O(1)-Rb(1)-O(13)#17	101.5(3)
C(2)-O(14)	1.342(15)	O(1)-Rb(1)-O(13)#16	76.2(3)
Mo(1)-O(1)	1.750(12)	O(1)-Rb(1)-O(14)#17	68.6(3)
Mo(1)-O(2)	1.751(11)	O(3)#4-Rb(1)-O(8)#10	88.0(3)
Mo(1)-O(3)	1.758(9)	O(3)#4-Rb(1)-O(14)#17	109.0(2)
Mo(1)-O(4)	1.751(18)	O(6)#4-Rb(1)-O(3)#4	85.5(3)
Mo(2)-O(8)	1.721(10)	O(6)#4-Rb(1)-O(8)#10	141.9(2)
Mo(2)-O(9)	1.722(10)	O(6)#4-Rb(1)-O(12)#16	67.4(2)
Mo(2)-O(10)	1.77(2)	O(6)#4-Rb(1)-O(13)#16	106.2(3)
Mo(2)-O(11)	1.725(13)	O(6)#4-Rb(1)-O(13)#17	71.3(4)
Rb(1)-O(1)	2.770(12)	O(6)#4-Rb(1)-O(14)#17	62.7(2)
Rb(1)-O(3)#4	3.254(13)	O(12)#16-Rb(1)-O(3)#4	123.2(2)
Rb(1)-O(6)#4	2.993(14)	O(12)#16-Rb(1)-O(8)#10	142.7(3)
Rb(1)-O(8)#10	3.528(60)	O(12)#16-Rb(1)-O(13)#16	42.1(3)
Rb(1)-O(12)#16	3.052(14)	O(12)#16-Rb(1)-O(14)#17	101.3(2)
Rb(1)-O(13)#17	3.024(11)	O(13)#16-Rb(1)-O(3)#4	112.4(3)
Rb(1)-O(13)#16	3.192(13)	O(13)#17-Rb(1)-O(3)#4	70.7(3)
Rb(1)-O(14)#17	3.344(10)	O(13)#17-Rb(1)-O(8)#10	71.1(4)
Rb(2)-O(1)	3.361(11)	O(13)#16-Rb(1)-O(8)#10	110.9(4)
Rb(2)-O(4)	2.947(12)	O(13)#17-Rb(1)-O(12)#16	134.4(3)
Rb(2)-O(6)#4	3.153(10)	O(13)#17-Rb(1)-O(13)#16	176.1(5)
Rb(2)-O(7)#4	2.989(02)	O(13)#17-Rb(1)-O(14)#17	40.3(3)
Rb(2)-O(8)#4	3.164(11)	O(13)#16-Rb(1)-O(14)#17	136.0(3)
Rb(2)-O(9)#3	3.141(12)	O(14)#17-Rb(1)-O(8)#10	84.4(2)
Rb(2)-O(10)#4	3.247(15)	O(4)-Rb(2)-O(1)	53.4(4)
Rb(2)-O(12)#16	2.987(10)	O(4)-Rb(2)-O(6)#4	135.4(4)
Rb(2)-O(14)#17	3.316(10)	O(4)-Rb(2)-O(7)#4	174.1(3)
Rb(3)-O(2)#19	2.873(8)	O(4)-Rb(2)-O(8)#4	107.0(4)
Rb(3)-O(3)	2.848(7)	O(4)-Rb(2)-O(9)#3	99.6(4)
Rb(3)-O(5)#9	2.927(7)	O(4)-Rb(2)-O(10)#4	77.8(5)
Rb(3)-O(7)#9	3.111(7)	O(4)-Rb(2)-O(10)#3	72.4(4)
Rb(3)-O(8)#11	3.150(14)	O(4)-Rb(2)-O(12)#16	111.9(3)
Rb(3)-O(9)#18	2.964(14)	O(4)-Rb(2)-O(14)#17	77.2(3)
Rb(3)-O(11)#11	3.191(16)	O(6)#4-Rb(2)-O(1)	91.0(3)
Rb(3)-O(13)#4	2.901(9)	O(6)#4-Rb(2)-O(8)#4	117.5(3)
Rb(4)-O(2)#19	3.414(16)	O(6)#4-Rb(2)-O(9)#3	99.9(3)
Rb(4)-O(3)	2.975(16)	O(6)#4-Rb(2)-O(10)#4	128.6(4)
Rb(4)-O(4)#19	2.991(13)	O(6)#4-Rb(2)-O(10)#3	88.8(4)
Rb(4)-O(4)	3.516(11)	O(6)#4-Rb(2)-O(14)#17	61.5(2)

Rb(4)-O(5)#19	3.391(13)	O(7)#4-Rb(2)-O(1)	128.1(4)
Rb(4)-O(6)	2.912(17)	O(7)#4-Rb(2)-O(6)#4	42.1(3)
Rb(4)-O(10)#4	2.846(14)	O(7)#4-Rb(2)-O(8)#4	75.9(4)
Rb(4)-O(11)#4	3.456(11)	O(7)#4-Rb(2)-O(9)#3	76.8(4)
Rb(4)-O(12)#4	3.153(12)	O(7)#4-Rb(2)-O(10)#4	107.8(4)
Rb(4)-O(13)#4	2.966(12)	O(7)#4-Rb(2)-O(10)#3	101.8(4)
Rb(5)-O(1)#2	2.991(2)	O(7)#4-Rb(2)-O(12)#16	72.6(3)
Rb(5)-O(2)#12	2.934(14)	O(7)#4-Rb(2)-O(14)#17	98.4(3)
Rb(5)-O(3)#2	3.181(11)	O(8)#4-Rb(2)-O(1)	135.4(3)
Rb(5)-O(7)	2.92(16)	O(8)#4-Rb(2)-O(10)#4	52.8(4)
Rb(5)-O(8)	2.836(13)	O(8)#4-Rb(2)-O(10)#3	115.8(4)
Rb(5)-O(9)#19	2.899(8)	O(8)#4-Rb(2)-O(14)#17	160.3(3)
Rb(5)-O(13)#20	3.091(7)	O(9)#3-Rb(2)-O(1)	146.8(3)
Rb(5)-O(14)#20	2.982(7)	O(9)#3-Rb(2)-O(8)#4	65.2(4)
Rb(6)-O(2)#15	3.217(8)	O(9)#3-Rb(2)-O(10)#4	112.8(4)
Rb(6)-O(5)#13	3.261(8)	O(9)#3-Rb(2)-O(10)#3	52.5(4)
Rb(6)-O(6)#1	3.079(8)	O(9)#3-Rb(2)-O(14)#17	95.2(3)
Rb(6)-O(7)#13	3.064(16)	O(10)#4-Rb(2)-O(1)	82.7(4)
Rb(6)-O(8)#13	3.382(14)	O(10)#3-Rb(2)-O(1)	96.9(4)
Rb(6)-O(11)	2.796(15)	O(10)#4-Rb(2)-O(10)#3	142.5(7)
Rb(6)-O(12)	2.976(10)	O(10)#3-Rb(2)-O(14)#17	46.2(4)
O(6)-C(1)-O(5)	118.9(10)	O(10)#4-Rb(2)-O(14)#17	145.1(4)
O(7)-C(1)-O(5)	114.6(11)	O(12)#16-Rb(2)-O(1)	66.7(2)
O(7)-C(1)-O(6)	126.5(12)	O(12)#16-Rb(2)-O(6)#4	66.2(2)
O(12)-C(2)-O(13)	126.0(12)	O(12)#16-Rb(2)-O(8)#4	93.0(3)
O(12)-C(2)-O(14)	119.0(11)	O(12)#16-Rb(2)-O(9)#3	146.1(3)
O(13)-C(2)-O(14)	115.0(11)	O(12)#16-Rb(2)-O(10)#4	64.6(4)
O(1)-Mo(1)-O(2)	110.2(6)	O(12)#16-Rb(2)-O(10)#3	148.8(4)
O(1)-Mo(1)-O(3)	109.1(5)	O(12)#16-Rb(2)-O(14)#17	103.4(2)
O(1)-Mo(1)-O(4)	109.4(8)	O(2)#19-Rb(3)-O(5)#9	100.7(3)
O(2)-Mo(1)-O(3)	109.7(7)	O(2)#19-Rb(3)-O(7)#9	75.7(3)
O(4)-Mo(1)-O(2)	110.1(6)	O(2)#19-Rb(3)-O(8)#11	165.8(4)
O(4)-Mo(1)-O(3)	108.4(6)	O(2)#19-Rb(3)-O(9)#18	108.7(3)
O(8)-Mo(2)-O(9)	110.8(9)	O(2)#19-Rb(3)-O(11)#11	136.8(3)
O(8)-Mo(2)-O(10)	109.5(7)	O(2)#19-Rb(3)-O(11)#18	69.3(3)
O(8)-Mo(2)-O(11)	109.5(6)	O(2)#19-Rb(3)-O(13)#4	87.4(4)
O(9)-Mo(2)-O(10)	109.8(7)	O(3)-Rb(3)-O(2)#19	74.5(4)
O(11)-Mo(2)-O(9)	110.3(7)	O(3)-Rb(3)-O(5)#9	119.5(3)
O(11)-Mo(2)-O(10)	106.8(10)	O(3)-Rb(3)-O(7)#9	80.4(3)
O(5)#9-Rb(3)-O(7)#9	42.1(3)	O(3)-Rb(3)-O(8)#11	103.6(3)
O(5)#9-Rb(3)-O(8)#11	92.5(3)	O(3)-Rb(3)-O(9)#18	156.5(4)
O(5)#9-Rb(3)-O(9)#18	83.3(3)	O(3)-Rb(3)-O(11)#18	143.1(3)
O(5)#9-Rb(3)-O(11)#11	72.3(4)	O(3)-Rb(3)-O(11)#11	73.1(4)
O(5)#9-Rb(3)-O(11)#18	62.6(3)	O(3)-Rb(3)-O(13)#4	78.5(3)

O(7)#9-Rb(3)-O(8)#11	118.2(3)	O(10)#4-Rb(4)-O(5)#19	81.9(3)
O(7)#9-Rb(3)-O(11)#11	71.5(3)	O(10)#4-Rb(4)-O(6)	107.2(4)
O(7)#9-Rb(3)-O(11)#18	84.3(3)	O(10)#4-Rb(4)-O(11)#4	51.8(5)
O(8)#11-Rb(3)-O(11)#11	52.7(3)	O(10)#4-Rb(4)-O(12)#4	137.5(4)
O(8)#11-Rb(3)-O(11)#18	113.2(3)	O(10)#4-Rb(4)-O(13)#4	179.3(5)
O(9)#18-Rb(3)-O(7)#9	123.1(4)	O(11)#4-Rb(4)-O(4)	85.6(4)
O(9)#18-Rb(3)-O(8)#11	67.4(4)	O(12)#4-Rb(4)-O(2)#19	100.4(2)
O(9)#18-Rb(3)-O(11)#11	112.6(4)	O(12)#4-Rb(4)-O(4)	127.5(3)
O(9)#18-Rb(3)-O(11)#18	50.0(3)	O(12)#4-Rb(4)-O(5)#19	60.0(2)
O(11)#11-Rb(3)-O(11)#18	132.4(6)	O(12)#4-Rb(4)-O(11)#4	90.6(3)
O(13)#4-Rb(3)-O(5)#9	161.5(3)	O(13)#4-Rb(4)-O(2)#19	77.0(4)
O(13)#4-Rb(3)-O(7)#9	155.9(5)	O(13)#4-Rb(4)-O(3)	75.5(3)
O(13)#4-Rb(3)-O(8)#11	78.4(4)	O(13)#4-Rb(4)-O(4)	105.2(4)
O(13)#4-Rb(3)-O(9)#18	78.3(4)	O(13)#4-Rb(4)-O(4)#19	100.9(4)
O(13)#4-Rb(3)-O(11)#18	106.0(3)	O(13)#4-Rb(4)-O(5)#19	98.4(3)
O(13)#4-Rb(3)-O(11)#11	112.9(4)	O(13)#4-Rb(4)-O(11)#4	128.8(4)
O(2)#19-Rb(4)-O(4)	111.9(3)	O(13)#4-Rb(4)-O(12)#4	43.0(3)
O(2)#19-Rb(4)-O(11)#4	145.3(3)	O(1)#2-Rb(5)-O(3)#2	55.0(3)
O(3)-Rb(4)-O(2)#19	65.2(3)	O(1)#2-Rb(5)-O(13)#20	74.8(3)
O(3)-Rb(4)-O(4)#19	116.4(4)	O(2)#12-Rb(5)-O(1)#2	117.5(4)
O(3)-Rb(4)-O(4)	51.2(4)	O(2)#12-Rb(5)-O(3)#2	68.8(3)
O(3)-Rb(4)-O(5)#19	162.0(3)	O(2)#12-Rb(5)-O(13)#20	121.5(4)
O(3)-Rb(4)-O(11)#4	136.5(3)	O(2)#12-Rb(5)-O(14)#20	84.3(3)
O(3)-Rb(4)-O(12)#4	118.1(2)	O(7)-Rb(5)-O(1)#2	111.9(3)
O(4)#19-Rb(4)-O(2)#19	52.7(4)	O(7)-Rb(5)-O(2)#12	77.8(3)
O(4)#19-Rb(4)-O(4)	145.2(6)	O(7)-Rb(5)-O(3)#2	78.2(3)
O(4)#19-Rb(4)-O(5)#19	47.4(4)	O(7)-Rb(5)-O(13)#20	155.3(4)
O(4)#19-Rb(4)-O(11)#4	95.6(4)	O(7)-Rb(5)-O(14)#20	161.4(3)
O(4)#19-Rb(4)-O(12)#4	87.3(3)	O(8)-Rb(5)-O(1)#2	145.0(4)
O(5)#19-Rb(4)-O(2)#19	97.1(2)	O(8)-Rb(5)-O(2)#12	96.4(3)
O(5)#19-Rb(4)-O(4)	145.9(4)	O(8)-Rb(5)-O(3)#2	157.5(4)
O(5)#19-Rb(4)-O(11)#4	60.3(2)	O(8)-Rb(5)-O(7)	82.2(4)
O(6)-Rb(4)-O(2)#19	146.5(3)	O(8)-Rb(5)-O(9)#19	72.6(5)
O(6)-Rb(4)-O(3)	92.2(2)	O(8)-Rb(5)-O(13)#20	80.4(4)
O(6)-Rb(4)-O(4)#19	148.9(4)	O(8)-Rb(5)-O(14)#20	105.0(4)
O(6)-Rb(4)-O(4)	62.7(3)	O(9)#19-Rb(5)-O(1)#2	77.8(4)
O(6)-Rb(4)-O(5)#19	102.4(2)	O(9)#19-Rb(5)-O(2)#12	158.0(5)
O(6)-Rb(4)-O(11)#4	68.0(3)	O(9)#19-Rb(5)-O(3)#2	114.9(3)
O(6)-Rb(4)-O(12)#4	67.4(2)	O(9)#19-Rb(5)-O(7)	81.8(4)
O(6)-Rb(4)-O(13)#4	73.3(3)	O(9)#19-Rb(5)-O(13)#20	76.3(4)
O(10)#4-Rb(4)-O(2)#19	102.4(4)	O(9)#19-Rb(5)-O(14)#20	116.6(4)
O(10)#4-Rb(4)-O(3)	104.0(4)	O(13)#20-Rb(5)-O(3)#2	121.6(4)
O(10)#4-Rb(4)-O(4)	74.8(4)	O(14)#20-Rb(5)-O(1)#2	72.1(4)
O(10)#4-Rb(4)-O(4)#19	78.8(4)	O(14)#20-Rb(5)-O(3)#2	90.7(3)

O(14)#20-Rb(5)-O(13)#20	42.7(3)	O(7)#1-Rb(6)-O(8)#13	112.4(3)
O(2)#15-Rb(6)-O(5)#13	87.2(3)	O(11)-Rb(6)-O(2)#15	76.2(4)
O(2)#15-Rb(6)-O(8)#13	81.3(3)	O(11)-Rb(6)-O(5)#13	68.7(3)
O(5)#13-Rb(6)-O(8)#13	110.5(2)	O(11)-Rb(6)-O(6)#1	75.2(4)
O(6)#1-Rb(6)-O(2)#15	144.3(2)	O(11)-Rb(6)-O(7)#13	100.7(3)
O(6)#1-Rb(6)-O(5)#13	101.7(2)	O(11)-Rb(6)-O(7)#1	76.2(3)
O(6)#1-Rb(6)-O(7)#1	41.6(3)	O(11)-Rb(6)-O(8)#13	157.5(5)
O(6)#1-Rb(6)-O(8)#13	125.5(2)	O(11)-Rb(6)-O(12)	109.1(4)
O(7)#13-Rb(6)-O(2)#15	71.6(3)	O(12)-Rb(6)-O(2)#15	143.3(2)
O(7)#1-Rb(6)-O(2)#15	110.4(4)	O(12)-Rb(6)-O(5)#13	63.2(2)
O(7)#1-Rb(6)-O(5)#13	135.5(3)	O(12)-Rb(6)-O(6)#1	67.7(2)
O(7)#13-Rb(6)-O(5)#13	40.0(3)	O(12)-Rb(6)-O(7)#1	106.0(3)
O(7)#13-Rb(6)-O(6)#1	134.8(3)	O(12)-Rb(6)-O(7)#13	71.8(3)
O(7)#13-Rb(6)-O(7)#1	175.5(5)	O(12)-Rb(6)-O(8)#13	89.0(3)
O(7)#13-Rb(6)-O(8)#13	71.7(3)		

Symmetry transformations used to generate equivalent atoms:

#1 x-1/2,-y+1/2,z #2 -x+1,-y,z+1/2 #3 x+1/2,-y+3/2,z #4 x+1/2,-y+1/2,z

#5 x-1,y,z #6 x-1,y+1,z #7 -x,-y+1,z-1/2 #8 x,y+1,z #9 -x+1,-y,z-1/2

#10 -x+1,-y+1,z-1/2 #11 -x+1/2,y-1/2,z-1/2 #12 -x+1,-y+1,z+1/2 #13 x-1/2,-y+3/2,z

#14 -x+1/2,y+3/2,z+1/2 #15 -x+1/2,y+1/2,z+1/2 #16 x+1,y-1,z #17 x+1,y,z

#18 -x+1/2,y-3/2,z-1/2 #19 x,y-1,z #20 -x,-y+1,z+1/2

Table S5 Bond distances [Å] and angles [°] for $Cs_3MoO_4(HCO_3)$.

C(1)-O(8)	1.256(11)	O(9)#1-Cs(5)-O(9)#9	171.1(5)
C(1)-O(9)	1.277(17)	O(9)#1-Cs(5)-O(10)#1	37.5(3)
C(1)-O(10)	1.332(12)	O(9)#9-Cs(5)-O(10)#1	133.8(3)
C(2)-O(1)	1.244(16)	O(9)#9-Cs(5)-O(4)#10	112.9(3)
C(2)-O(2)	1.373(10)	O(9)#1-Cs(5)-O(4)#10	71.6(3)
C(2)-O(3)	1.236(10)	O(9)#1-Cs(5)-O(8)#9	131.6(3)
Mo(1)-O(4)	1.730(9)	O(9)#1-Cs(5)-O(13)#9	73.3(3)
Mo(1)-O(5)	1.721(9)	O(9)#9-Cs(5)-O(13)#9	114.0(3)
Mo(1)-O(6)	1.747(12)	O(9)#9-Cs(5)-O(5)#10	67.7(3)
Mo(1)-O(7)	1.768(17)	O(9)#1-Cs(5)-O(5)#10	117.1(3)
Mo(2)-O(11)	1.721(13)	O(11)-Cs(5)-O(9)#9	78.9(3)
Mo(2)-O(12)	1.760(12)	O(11)-Cs(5)-O(9)#1	95.6(3)
Mo(2)-O(13)	1.776(9)	O(11)-Cs(5)-O(10)#1	66.5(2)
Mo(2)-O(14)	1.768(9)	O(11)-Cs(5)-O(3)#9	106.4(3)
Cs(1)-O(9)#4	3.178(13)	O(11)-Cs(5)-O(4)#10	72.9(3)
Cs(1)-O(11)#3	3.214(8)	O(11)-Cs(5)-O(8)#9	77.9(3)
Cs(1)-O(10)#4	3.134(9)	O(11)-Cs(5)-O(13)#9	159.1(3)
Cs(1)-O(4)	3.033(9)	O(11)-Cs(5)-O(5)#10	68.6(3)
Cs(1)-O(13)#3	3.194(9)	O(10)#1-Cs(5)-O(4)#10	85.9(2)
Cs(1)-O(5)#5	3.093(9)	O(10)#1-Cs(5)-O(5)#10	121.3(2)
Cs(1)-O(14)#2	3.110(10)	O(3)#9-Cs(5)-O(9)#1	70.2(3)
Cs(1)-O(1)	3.138(14)	O(3)#9-Cs(5)-O(9)#9	104.3(3)
Cs(2)-O(2)#6	3.531(8)	O(3)#9-Cs(5)-O(10)#1	60.71(17)
Cs(2)-O(3)#7	3.208(6)	O(3)#9-Cs(5)-O(4)#10	141.55(18)
Cs(2)-O(4)#6	3.558(10)	O(3)#9-Cs(5)-O(8)#9	66.19(17)
Cs(2)-O(8)#6	3.152(6)	O(3)#9-Cs(5)-O(13)#9	86.8(2)
Cs(2)-O(5)#6	3.774(11)	O(3)#9-Cs(5)-O(5)#10	171.03(17)
Cs(2)-O(14)#8	3.459(13)	O(4)#10-Cs(5)-O(5)#10	45.5(3)
Cs(2)-O(6)	2.962(12)	O(8)#9-Cs(5)-O(9)#9	40.6(3)
Cs(2)-O(7)#6	3.81(2)	O(8)#9-Cs(5)-O(10)#1	100.48(18)
Cs(2)-O(1)#7	3.294(7)	O(8)#9-Cs(5)-O(4)#10	144.61(19)
Cs(2)-O(1)#6	3.188(7)	O(8)#9-Cs(5)-O(13)#9	122.84(19)
Cs(3)-O(11)#7	3.551(16)	O(8)#9-Cs(5)-O(5)#10	105.05(17)
Cs(3)-O(10)#5	3.433(9)	O(13)#9-Cs(5)-O(10)#1	108.6(2)
Cs(3)-O(12)#7	3.083(17)	O(13)#9-Cs(5)-O(4)#10	86.6(3)
Cs(3)-O(3)	3.276(6)	O(13)#9-Cs(5)-O(5)#10	100.1(3)
Cs(3)-O(4)	3.238(12)	O(9)-Cs(6)-O(2)#10	163.1(3)
Cs(3)-O(8)	3.158(6)	O(9)-Cs(6)-O(4)#12	78.6(3)
Cs(3)-O(5)#5	3.274(13)	O(9)-Cs(6)-O(5)#11	78.3(3)
Cs(3)-O(7)#5	3.363(9)	O(9)-Cs(6)-O(6)#11	107.8(2)
Cs(3)-O(7)	3.341(8)	O(9)-Cs(6)-O(6)#12	111.5(3)
Cs(3)-O(1)	3.123(14)	O(9)-Cs(6)-O(1)#10	156.3(4)
Cs(4)-O(9)#6	3.206(13)	O(2)#10-Cs(6)-O(4)#12	92.5(3)

Cs(4)-O(2)#6	3.471(9)	O(2)#10-Cs(6)-O(5)#11	85.1(3)
Cs(4)-O(12)#7	3.459(7)	O(2)#10-Cs(6)-O(6)#11	62.1(3)
Cs(4)-O(12)#6	3.263(8)	O(2)#10-Cs(6)-O(6)#12	72.0(3)
Cs(4)-O(3)#7	3.116(6)	O(2)#10-Cs(6)-O(1)#10	40.2(3)
Cs(4)-O(8)#6	3.290(6)	O(4)#12-Cs(6)-O(6)#11	112.6(3)
Cs(4)-O(13)#7	3.248(13)	O(4)#12-Cs(6)-O(6)#12	50.8(3)
Cs(4)-O(14)#6	3.407(13)	O(4)#12-Cs(6)-O(1)#10	118.2(3)
Cs(4)-O(6)	3.456(14)	O(13)#13-Cs(6)-O(9)	81.4(3)
Cs(4)-O(7)	3.13(2)	O(13)#13-Cs(6)-O(2)#10	114.9(3)
Cs(5)-O(9)#9	3.311(6)	O(13)#13-Cs(6)-O(4)#12	103.9(2)
Cs(5)-O(9)#1	3.169(6)	O(13)#13-Cs(6)-O(5)#11	159.5(4)
Cs(5)-O(11)	2.993(13)	O(13)#13-Cs(6)-O(14)	73.6(4)
Cs(5)-O(10)#1	3.555(8)	O(13)#13-Cs(6)-O(6)#11	143.4(3)
Cs(5)-O(3)#9	3.155(6)	O(13)#13-Cs(6)-O(6)#12	72.1(3)
Cs(5)-O(4)#10	3.661(12)	O(13)#13-Cs(6)-O(1)#10	78.3(3)
Cs(5)-O(8)#9	3.202(6)	O(5)#11-Cs(6)-O(4)#12	69.1(3)
Cs(5)-O(13)#9	3.504(9)	O(5)#11-Cs(6)-O(6)#12	112.8(3)
Cs(5)-O(5)#10	3.730(13)	O(5)#11-Cs(6)-O(6)#11	49.0(3)
Cs(6)-O(9)	3.107(14)	O(5)#11-Cs(6)-O(1)#10	122.2(3)
Cs(6)-O(2)#10	3.110(9)	O(14)-Cs(6)-O(9)	84.6(3)
Cs(6)-O(4)#12	3.229(10)	O(14)-Cs(6)-O(2)#10	103.6(3)
Cs(6)-O(13)#13	3.010(9)	O(14)-Cs(6)-O(4)#12	163.3(4)
Cs(6)-O(5)#11	3.135(11)	O(14)-Cs(6)-O(5)#11	107.3(2)
Cs(6)-O(14)	3.022(10)	O(14)-Cs(6)-O(6)#11	72.2(3)
Cs(6)-O(6)#12	3.446(9)	O(14)-Cs(6)-O(6)#12	138.9(3)
Cs(6)-O(6)#11	3.662(9)	O(14)-Cs(6)-O(1)#10	77.9(3)
Cs(6)-O(1)#10	3.282(14)	O(6)#12-Cs(6)-O(6)#11	130.7(4)
O(9)#4-Cs(1)-O(11)#3	77.8(3)	O(1)#10-Cs(6)-O(6)#12	73.6(3)
O(9)#4-Cs(1)-O(13)#3	122.5(3)	O(1)#10-Cs(6)-O(6)#11	82.1(2)
O(10)#4-Cs(1)-O(9)#4	40.6(3)	O(4)-Mo(1)-O(6)	111.1(5)
O(10)#4-Cs(1)-O(11)#3	75.2(3)	O(4)-Mo(1)-O(7)	107.2(6)
O(10)#4-Cs(1)-O(13)#3	94.5(3)	O(5)-Mo(1)-O(4)	111.8(7)
O(10)#4-Cs(1)-O(1)	164.2(3)	O(5)-Mo(1)-O(6)	111.1(5)
O(4)-Cs(1)-O(9)#4	80.5(3)	O(5)-Mo(1)-O(7)	107.1(6)
O(4)-Cs(1)-O(11)#3	143.7(3)	O(6)-Mo(1)-O(7)	108.2(8)
O(4)-Cs(1)-O(10)#4	105.7(3)	O(11)-Mo(2)-O(12)	111.0(8)
O(4)-Cs(1)-O(13)#3	157.0(3)	O(11)-Mo(2)-O(13)	107.3(4)
O(4)-Cs(1)-O(5)#5	72.2(4)	O(11)-Mo(2)-O(14)	108.8(5)
O(4)-Cs(1)-O(14)#2	100.3(2)	O(12)-Mo(2)-O(13)	110.6(5)
O(4)-Cs(1)-O(1)	79.8(3)	O(12)-Mo(2)-O(14)	110.9(5)
O(13)#3-Cs(1)-O(11)#3	52.1(3)	O(14)-Mo(2)-O(13)	108.3(7)
O(5)#5-Cs(1)-O(9)#4	77.8(3)	O(5)#5-Cs(1)-O(13)#3	109.2(2)
O(5)#5-Cs(1)-O(11)#3	74.9(3)	O(5)#5-Cs(1)-O(14)#2	158.8(4)
O(5)#5-Cs(1)-O(10)#4	115.5(3)	O(5)#5-Cs(1)-O(1)	80.2(3)

O(14)#2-Cs(1)-O(9)#4	121.3(3)	O(1)#6-Cs(2)-O(4)#6	71.6(3)
O(14)#2-Cs(1)-O(11)#3	115.9(3)	O(1)#7-Cs(2)-O(4)#6	114.5(3)
O(14)#2-Cs(1)-O(10)#4	85.5(3)	O(1)#7-Cs(2)-O(5)#6	68.8(3)
O(14)#2-Cs(1)-O(13)#3	69.9(3)	O(1)#6-Cs(2)-O(5)#6	117.3(3)
O(14)#2-Cs(1)-O(1)	78.9(3)	O(1)#7-Cs(2)-O(14)#8	113.6(3)
O(1)-Cs(1)-O(9)#4	154.1(4)	O(1)#6-Cs(2)-O(14)#8	73.2(3)
O(1)-Cs(1)-O(11)#3	109.4(3)	O(1)#6-Cs(2)-O(7)#6	92.0(3)
O(1)-Cs(1)-O(13)#3	77.8(3)	O(1)#7-Cs(2)-O(7)#6	88.4(3)
O(2)#6-Cs(2)-O(4)#6	107.9(2)	O(1)#6-Cs(2)-O(1)#7	170.6(5)
O(2)#6-Cs(2)-O(5)#6	150.56(18)	C(2)#7-Cs(2)-O(2)#6	113.7(2)
O(2)#6-Cs(2)-O(7)#6	110.6(2)	C(2)#7-Cs(2)-O(4)#6	128.2(2)
O(3)#7-Cs(2)-O(2)#6	100.53(18)	C(2)#7-Cs(2)-O(5)#6	84.8(2)
O(3)#7-Cs(2)-O(4)#6	123.96(18)	C(2)#6-Cs(2)-O(5)#6	129.44(19)
O(3)#7-Cs(2)-O(5)#6	89.4(2)	C(2)#7-Cs(2)-O(14)#8	126.1(2)
O(3)#7-Cs(2)-O(14)#8	145.14(19)	C(2)#7-Cs(2)-O(7)#6	91.1(2)
O(3)#7-Cs(2)-O(7)#6	80.3(2)	C(2)#6-Cs(2)-O(7)#6	94.15(19)
O(3)#7-Cs(2)-O(1)#7	39.8(3)	O(10)#5-Cs(3)-O(11)#7	62.5(2)
O(4)#6-Cs(2)-O(5)#6	45.8(3)	O(12)#7-Cs(3)-O(11)#7	50.6(3)
O(4)#6-Cs(2)-O(7)#6	44.8(3)	O(12)#7-Cs(3)-O(10)#5	81.1(2)
O(8)#6-Cs(2)-O(2)#6	60.62(17)	O(12)#7-Cs(3)-O(3)	137.5(2)
O(8)#6-Cs(2)-O(3)#7	66.81(17)	O(12)#7-Cs(3)-O(4)	105.2(3)
O(8)#6-Cs(2)-O(4)#6	86.7(2)	O(12)#7-Cs(3)-O(8)	110.5(2)
O(8)#6-Cs(2)-O(5)#6	99.5(2)	O(12)#7-Cs(3)-O(5)#5	102.8(3)
O(8)#6-Cs(2)-O(14)#8	141.81(18)	O(12)#7-Cs(3)-O(7)#5	79.1(4)
O(8)#6-Cs(2)-O(7)#6	56.8(3)	O(12)#7-Cs(3)-O(7)	79.1(4)
O(8)#6-Cs(2)-O(1)#7	103.9(3)	O(12)#7-Cs(3)-O(1)	177.8(3)
O(8)#6-Cs(2)-O(1)#6	68.7(3)	O(12)#7-Cs(3)-C(2)	156.7(3)
O(5)#6-Cs(2)-O(7)#6	43.5(3)	O(3)-Cs(3)-O(11)#7	92.2(2)
O(14)#8-Cs(2)-O(2)#6	87.2(2)	O(3)-Cs(3)-O(10)#5	61.01(17)
O(14)#8-Cs(2)-O(4)#6	84.4(3)	O(3)-Cs(3)-O(7)#5	86.4(3)
O(14)#8-Cs(2)-O(5)#6	100.4(3)	O(3)-Cs(3)-O(7)	124.4(3)
O(14)#8-Cs(2)-O(7)#6	128.9(3)	O(4)-Cs(3)-O(11)#7	136.6(2)
O(6)-Cs(2)-O(2)#6	65.0(2)	O(4)-Cs(3)-O(10)#5	159.3(2)
O(6)-Cs(2)-O(3)#7	76.7(2)	O(4)-Cs(3)-O(3)	117.1(2)
O(6)-Cs(2)-O(4)#6	159.3(3)	O(4)-Cs(3)-O(5)#5	67.3(3)
O(6)-Cs(2)-O(8)#6	104.6(3)	O(4)-Cs(3)-O(7)#5	115.7(3)
O(6)-Cs(2)-O(5)#6	144.4(2)	O(4)-Cs(3)-O(7)	50.7(3)
O(6)-Cs(2)-O(14)#8	76.0(3)	O(8)-Cs(3)-O(11)#7	70.7(2)
O(6)-Cs(2)-O(7)#6	155.1(4)	O(8)-Cs(3)-O(10)#5	104.06(18)
O(6)-Cs(2)-O(1)#7	80.1(3)	O(8)-Cs(3)-O(3)	65.27(17)
O(6)-Cs(2)-O(1)#6	95.9(3)	O(8)-Cs(3)-O(4)	92.3(2)
O(1)#7-Cs(2)-O(2)#6	133.9(3)	O(8)-Cs(3)-O(5)#5	144.7(2)
O(1)#6-Cs(2)-O(2)#6	37.8(3)	O(8)-Cs(3)-O(7)#5	147.5(3)
O(1)#6-Cs(2)-O(3)#7	131.1(3)	O(8)-Cs(3)-O(7)	62.1(3)

O(5)#5-Cs(3)-O(11)#7	143.5(2)	O(3)#7-Cs(4)-O(8)#6	66.22(17)
O(5)#5-Cs(3)-O(10)#5	92.2(2)	O(3)#7-Cs(4)-O(13)#7	92.1(2)
O(5)#5-Cs(3)-O(3)	97.6(2)	O(3)#7-Cs(4)-O(14)#6	144.2(2)
O(5)#5-Cs(3)-O(7)	115.0(3)	O(3)#7-Cs(4)-O(6)	71.2(2)
O(5)#5-Cs(3)-O(7)#5	50.0(3)	O(3)#7-Cs(4)-O(7)	109.2(3)
O(7)-Cs(3)-O(11)#7	86.8(3)	O(8)#6-Cs(4)-O(2)#6	60.08(16)
O(7)#5-Cs(3)-O(11)#7	96.1(3)	O(8)#6-Cs(4)-O(12)#7	125.8(3)
O(7)#5-Cs(3)-O(10)#5	45.2(3)	O(8)#6-Cs(4)-O(14)#6	97.9(2)
O(7)-Cs(3)-O(10)#5	149.3(3)	O(8)#6-Cs(4)-O(6)	91.6(2)
O(7)-Cs(3)-O(7)#5	149.0(6)	O(13)#7-Cs(4)-O(2)#6	159.3(2)
O(1)-Cs(3)-O(11)#7	127.9(3)	O(13)#7-Cs(4)-O(12)#6	115.8(3)
O(1)-Cs(3)-O(10)#5	96.7(2)	O(13)#7-Cs(4)-O(12)#7	51.3(3)
O(1)-Cs(3)-O(3)	40.4(3)	O(13)#7-Cs(4)-O(8)#6	116.85(19)
O(1)-Cs(3)-O(4)	77.0(3)	O(13)#7-Cs(4)-O(14)#6	65.7(3)
O(1)-Cs(3)-O(8)	69.5(2)	O(13)#7-Cs(4)-O(6)	138.2(2)
O(1)-Cs(3)-O(5)#5	77.7(3)	O(14)#6-Cs(4)-O(2)#6	93.9(2)
O(1)-Cs(3)-O(7)	102.6(4)	O(14)#6-Cs(4)-O(12)#7	113.6(3)
O(1)-Cs(3)-O(7)#5	99.8(4)	O(14)#6-Cs(4)-O(6)	143.8(2)
O(9)#6-Cs(4)-O(2)#6	96.3(2)	O(6)-Cs(4)-O(2)#6	61.15(18)
O(9)#6-Cs(4)-O(12)#6	99.9(3)	O(6)-Cs(4)-O(12)#7	87.6(3)
O(9)#6-Cs(4)-O(12)#7	103.8(3)	O(7)-Cs(4)-O(9)#6	178.8(4)
O(9)#6-Cs(4)-O(8)#6	40.7(2)	O(7)-Cs(4)-O(2)#6	82.8(2)
O(9)#6-Cs(4)-O(13)#7	76.4(3)	O(7)-Cs(4)-O(12)#6	80.0(3)
O(9)#6-Cs(4)-O(14)#6	77.2(3)	O(7)-Cs(4)-O(12)#7	76.7(3)
O(9)#6-Cs(4)-O(6)	127.9(3)	O(7)-Cs(4)-O(8)#6	138.2(3)
O(12)#6-Cs(4)-O(2)#6	45.6(2)	O(7)-Cs(4)-O(13)#7	104.7(3)
O(12)#7-Cs(4)-O(2)#6	148.8(3)	O(7)-Cs(4)-O(14)#6	103.7(3)
O(12)#6-Cs(4)-O(12)#7	147.9(5)	O(7)-Cs(4)-O(6)	51.0(4)
O(12)#6-Cs(4)-O(8)#6	86.2(3)	O(9)-C(1)-O(10)	114.1(9)
O(12)#6-Cs(4)-O(14)#6	51.6(3)	O(8)-C(1)-O(9)	126.4(10)
O(12)#6-Cs(4)-O(6)	94.7(3)	O(8)-C(1)-O(10)	119.5(9)
O(3)#7-Cs(4)-O(9)#6	70.2(2)	O(3)-C(2)-O(2)	118.9(8)
O(3)#7-Cs(4)-O(2)#6	103.73(18)	O(3)-C(2)-O(1)	126.7(9)
O(3)#7-Cs(4)-O(12)#7	62.5(3)	O(1)-C(2)-O(2)	114.4(9)
O(3)#7-Cs(4)-O(12)#6	147.9(3)		

Symmetry transformations used to generate equivalent atoms:

#1 x-1/2,-y+1/2,z #2 -x+1,-y+1,z-1/2 #3 -x+1,-y+2,z-1/2 #4 -x+3/2,y+1/2,z-1/2

#5 x,y+1,z #6 x+1/2,-y+1/2,z #7 x+1/2,-y+3/2,z #8 -x+3/2,y-1/2,z-1/2

#9 x-1/2,-y+3/2,z #10 -x+1,-y+1,z+1/2 #11 -x+3/2,y+1/2,z+1/2

#12 -x+3/2,y-1/2,z+1/2 #13 x,y-1,z #14 -x+1,-y+2,z+1/2

Table S6 Carbonates with cut-off edge reported.

Compound	Space group	Noncentrosymmetric	Cut-off edge (nm)
Na_2CO_3	*C*2/*m*	No (N)	221
K_2CO_3	$P2_1/c$	N	225
Rb_2CO_3	$P2_1/c$	N	237
Cs_2CO_3	$P2_1/c$	N	328
$NaHCO_3$	$P2_1/c$	N	< 190
$KHCO_3$	$P2_1/a$	N	< 190
$RbHCO_3$	$C\bar{1}$	N	< 190
$CsHCO_3$	$P2_1/n$	N	< 190
$LiKCO_3$	$P2_1/c$	N	196
$LiRbCO_3$	$P2_1/n$	N	197
$LiCsCO_3$	$P2_1/n$	N	200
$LiZn(OH)CO_3$	$Pmn2_1$	Yes(Y)	< 190
$NaY(CO_3)_2$	$P2_1/c$	N	< 200
$NaZnCO_3(OH)$	*Pc*	Y	200
$Na_4Zn(CO_3)_3$	$P2_1/c$	N	305
$KY(CO_3)_2$	*C*2/*c*	N	< 200
$KEu(CO_3)_2$	*C*2/*c*	N	297
$KGd(CO_3)_2$	*C*2/*c*	N	273
$KTb(CO_3)_2$	*C*2/*c*	N	249
$Na_3Gd(CO_3)_3$	*Ama*2	Y	220
$Na_3Y(CO_3)_3$	*Ama*2	Y	220
$Na_4La_2(CO_3)_5$	$P6_3mc$	Y	235
$Na_5CsCa_5(CO_3)_8$	$P6_3mc$	Y	210

Figure S1 Experimental and theoretical powder XRD patterns for $Rb_3MoO_4(HCO_3)$ (a) and $Cs_3MoO_4(HCO_3)$ (b).

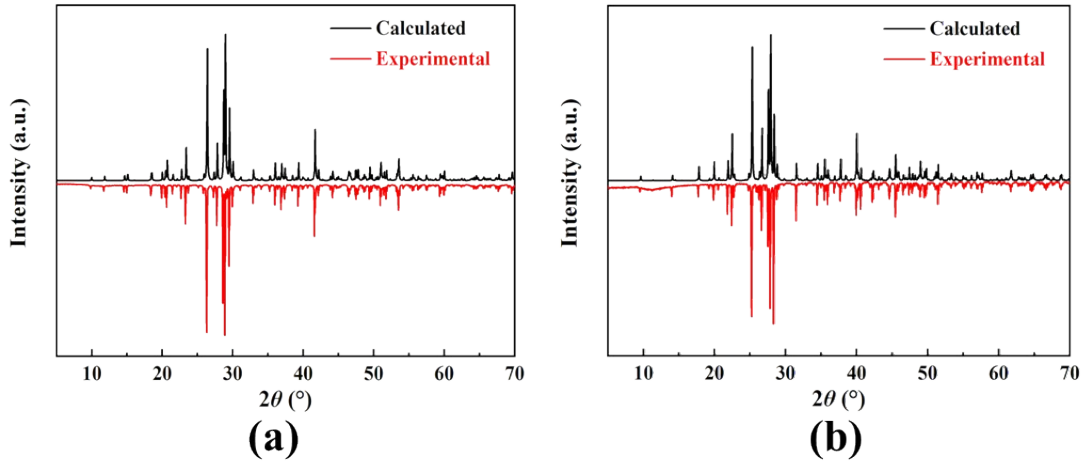


Figure S2 The arrangement of [MoO_4] tetrahedral groups of $Rb_3MoO_4(HCO_3)$ (a) (b) and $Cs_3MoO_4(HCO_3)$ (c) (d).

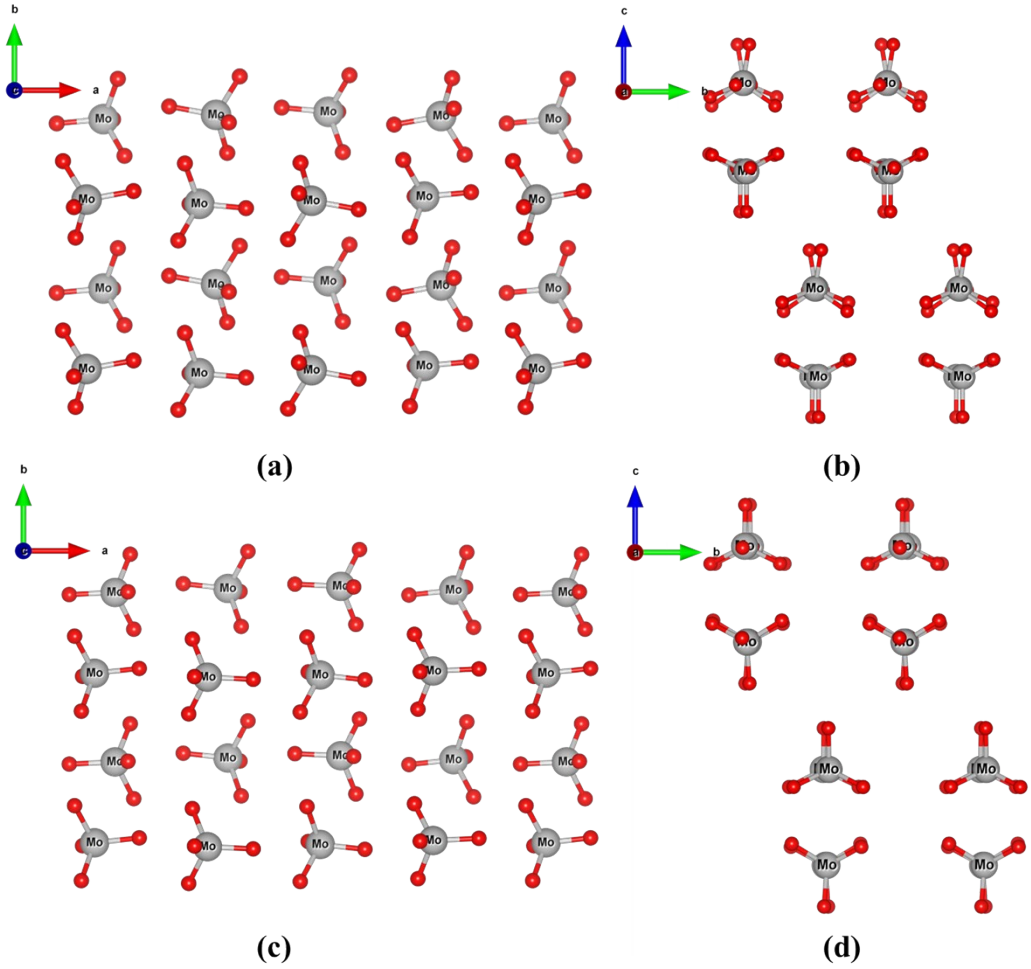


Figure S3 The cationic coordination environment of $M_3MoO_4(HCO_3)$ (M = Rb, Cs).

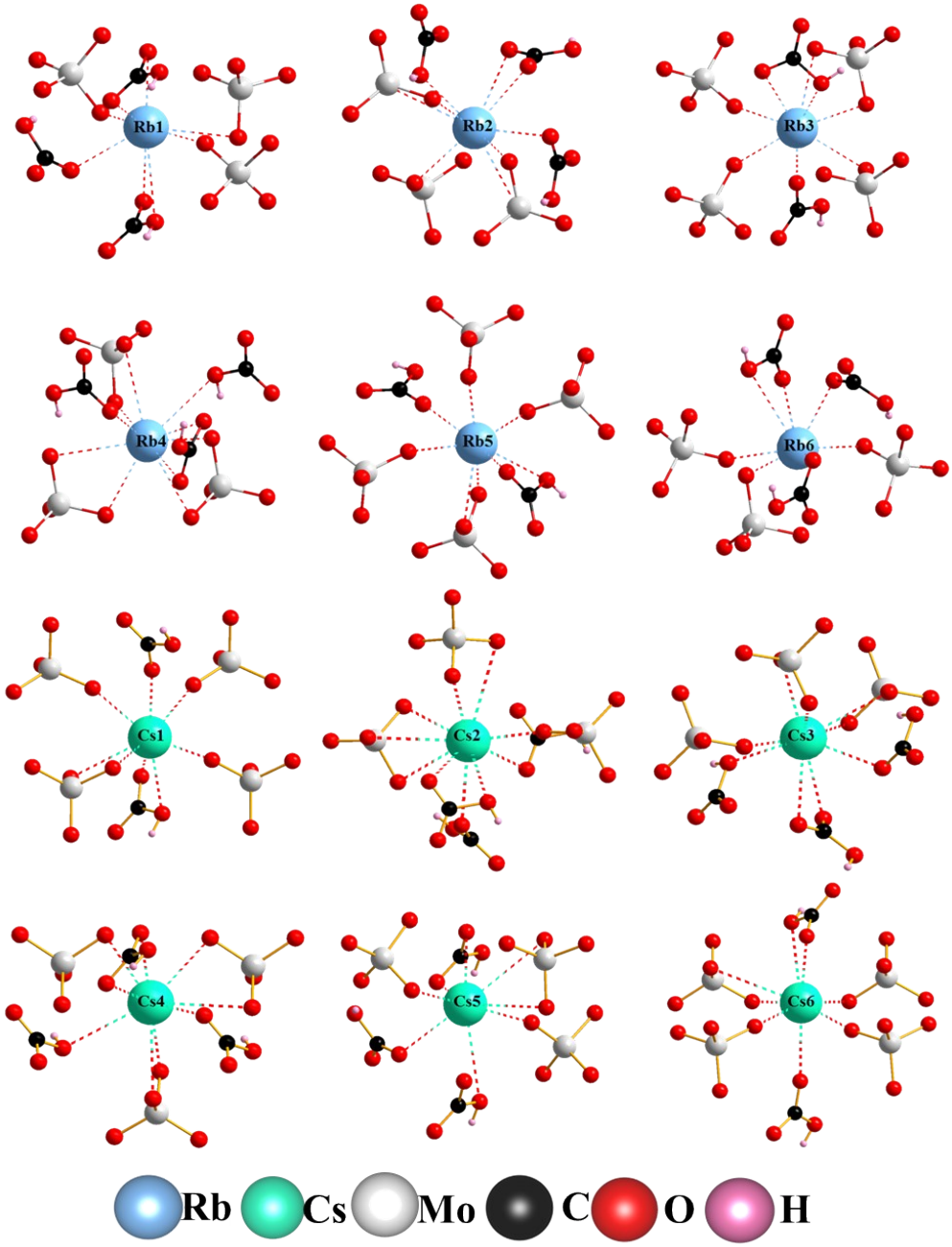


Figure S4 The crystal structure of $Cs_3MoO_4(HCO_3)$.

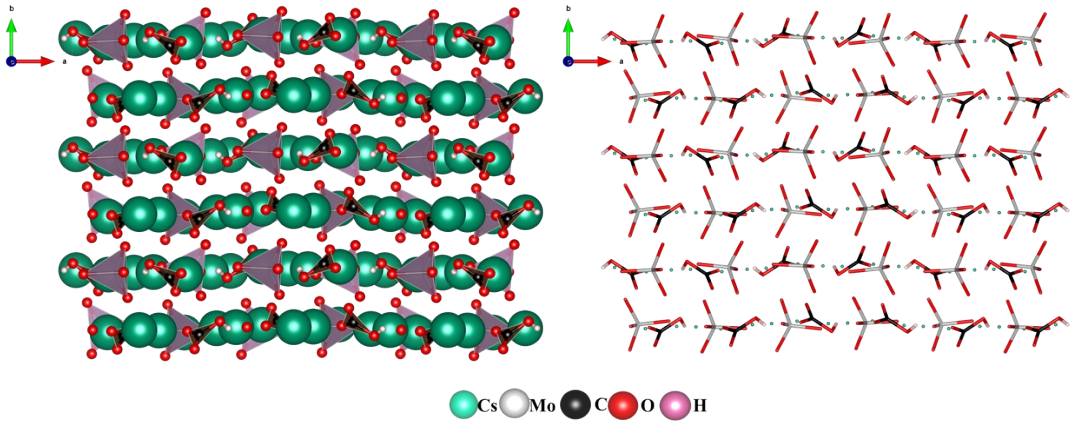
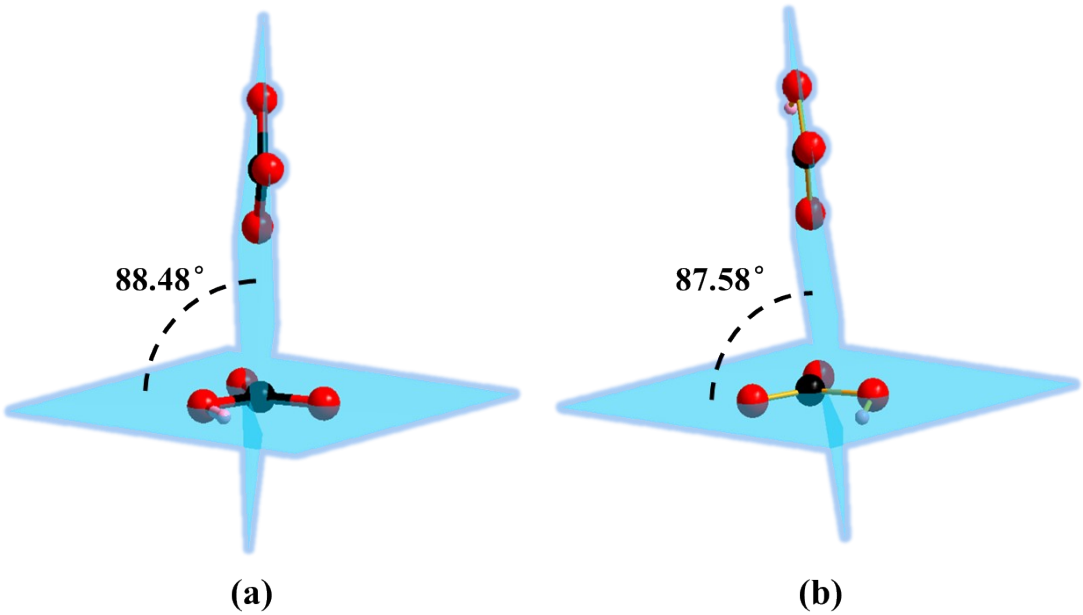
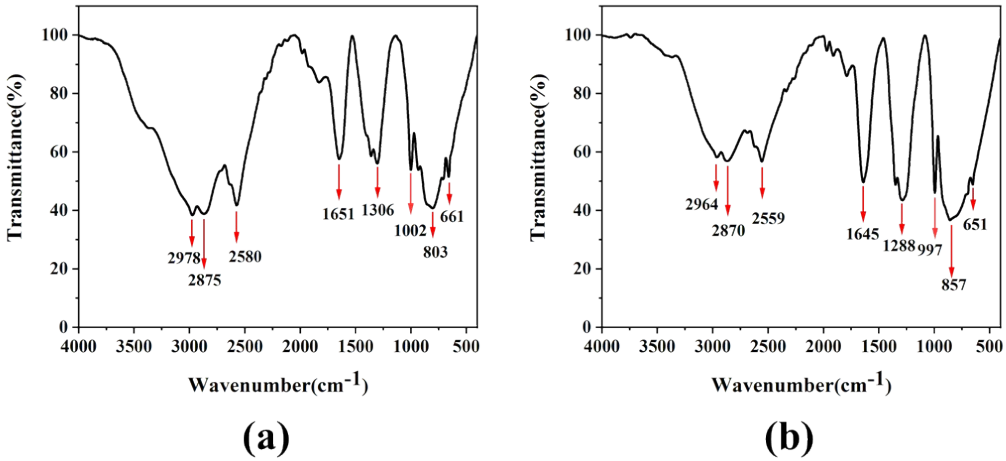


Figure S5 The adjacent plane [HCO_3] anion groups' dihedral angle of $Rb_3MoO_4(HCO_3)$ (a) and $Cs_3MoO_4(HCO_3)$ (b).



(a) **(b)**

Figure S6 FTIR spectra for $Rb_3MoO_4(HCO_3)$ (a) and $Cs_3MoO_4(HCO_3)$ (b).



(a) **(b)**

Figure S7 The UV-vis absorption spectra for $Rb_3MoO_4(HCO_3)$ (a) and $Cs_3MoO_4(HCO_3)$ (b).

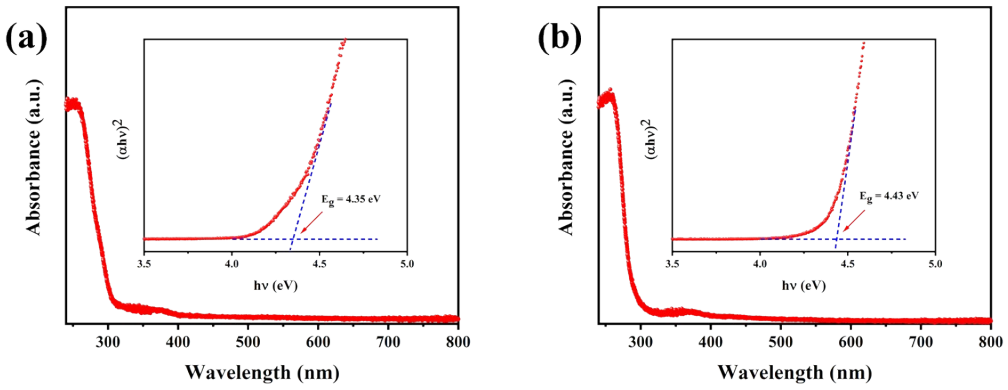


Figure S8 SHG density of the virtual electron occupied and unoccupied states for $Rb_3MoO_4(HCO_3)$ (a) (b) and $Cs_3MoO_4(HCO_3)$ (c) (d).