Supporting Information

Multi-element mixing boosts exfoliation of layered hexaniobate single crystals

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Table S1. Flux growth conditions for KNO and multicomponent KNO single crystals.

Run	Solute												cooling rate
	K_2CO_3	Nb_2O_5	Ta_2O_5	TiO_2	NH_4VO_3	$ZrO(NO_3)_2 \cdot 2H_2O$	Sb_2O_5	HfO_2	SnO_2	CeO_2	$Bi(NO_3)_3 \cdot 5H_2O$	WO_3	
	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ mmol	/ °C·h-1
1	2	3	-	-	-	-	-	-	-	-	-	-	200
2	2	2.4	0.15	0.3	0.3	0.3	-	-	-	-	-	-	200
3	2	2.76	0.06	0.12	-	-	0.12	0.12	-	-	-	-	200
4	2	2.4	0.15	0.3	-	-	0.3	0.3	-	-	-	-	200
5	2	2.4	0.15	0.3	-	-	0.3	0.3	-	-	-	-	quench
6	2	2.1	0.225	0.45	-	-	0.45	0.45	-	-	-	-	200
7	2	2.1	0.225	0.45	-	-	0.45	0.45	-	-	-	-	quench
8	2	1.8	0.3	0.6	-	-	0.6	0.6	-	-	-	-	200
9	2	1.2	0.45	0.9	-	-	0.9	0.9	-	-	-	-	200
10	2	0.6	0.6	1.2	-	-	1.2	1.2	-	-	-	-	200
11	2	2.4	0.15	0.3	-	-	0.3	-	0.3	-	-	-	200
12	2	2.4	0.15	0.3	-	-	0.3	-	-	0.3	-	-	200
13	2	2.4	0.15	0.3	-	-	0.3	-	-	-	0.3	-	200
14	2	2.4	0.15	0.3	-	-	0.3	-	-	-	-	0.3	200
15	2	2.55	0.15	0.3	-	-	0.3	-	-	-	-	-	quench
16	2	2.325	0.225	0.45	-	-	0.45	-	-	-	-	-	quench
17	2	2.325	0.225	0.45	-	-	0.45	-	-	-	-	-	200

Table S2. Crystallographic parameters for (a) $K_{3.8}H_{0.2}Nb_6O_{17}$ and (b) $K_{3.3}H_{0.7}Nb_{4.6}Ti_{0.4}Ta_{0.5}Sb_{0.5}O_{16.8}$ ($TiTaSb_{0.5}$-Q) prepared through quenching, determined from SXRD refinements.

(a) $K_4Nb_6O_{17}$

Atom	Wyckoff	*x*	*y*	*z*	*g*	*U* (Å^2)
K1	4a	0.0243(4)	0.7856(20)	0.7632	0.950	0.0026
K2	4a	0.0823(4)	0.3210(29)	0.7557	0.950	0.0026
K3	4a	0.2168(4)	0.6531(31)	0.7538	0.950	0.0026
K4	4a	0.2153(4)	0.6401(30)	0.2629	0.950	0.0026
Nb1	4a	0.0506(2)	0.6823(10)	0.2673	1.000	0.0034
Nb2	4a	0.0948(2)	0.1388(12)	0.2716	1.000	0.0034
Nb3	4a	0.1271(3)	0.7734(16)	-0.0017	1.000	0.0034
Nb4	4a	0.1239(4)	0.7536(18)	0.5069	1.000	0.0034
Nb5	4a	0.1756(4)	0.2128(20)	0.0009	1.000	0.0034
Nb6	4a	0.1733(4)	0.2036(20)	0.5044	1.000	0.0034
O1	4a	0.0017(9)	0.3602(49)	0.7621	1.000	0.0034
O2	4a	0.0353(11)	1.0504(53)	0.2677	1.000	0.0034
O3	4a	0.0790(15)	0.7250(88)	-0.0019	1.000	0.0034
O4	4a	0.0701(13)	0.6930(89)	0.5249	1.000	0.0034
O5	4a	0.0732(11)	0.3708(63)	-0.2566	1.000	0.0034
O6	4a	0.1042(16)	0.0736(76)	-0.0087	1.000	0.0034
O7	4a	0.1155(15)	0.1185(73)	0.5107	1.000	0.0034
O8	4a	0.1121(13)	0.7767(75)	0.2162	1.000	0.0003
O9	4a	0.1161(11)	0.8233(57)	0.7743	1.000	0.0003
O10	4a	0.1512(17)	0.4917(91)	0.0099	1.000	0.0003
O11	4a	0.1585(15)	0.5255(88)	0.4952	1.000	0.0003
O12	4a	0.1615(10)	0.2247(56)	0.2626	1.000	0.0003
O13	4a	0.1711(10)	0.2140(64)	0.7555	1.000	0.0003
O14	4a	0.1652(13)	0.9067(78)	0.0049	1.000	0.0003
O15	4a	0.1872(14)	0.9019(76)	0.5105	1.000	0.0003
O16	4a	0.2237(15)	0.2237(15)	0.0124	1.000	0.0003
O17	4a	0.2283(14)	0.2283(14)	0.5018	1.000	0.0003

Space group: $P2_1nb$; a = 0.78159(3) nm, b = 3.2962(2) nm, and c = 0.64734(3) nm, R_{wp}= 6.25%, R_p= 4.82%, S = 0.789. The primary profile parameter (PPP) for 020 reflections is refined.

(b) $TiTaSb_{0.5}$-Q

Atom	Wyckoff	*x*	*y*	*z*	*g*	*U* (Å^2)
K1	4a	0.0351(9)	0.7651(47)	0.7544	0.825	0.0026
K2	4a	0.0843(9)	0.2773(50)	0.7762	0.825	0.0026
K3	4a	0.2119(9)	0.6764(60)	0.7662	0.825	0.0026

K4	4a	0.2137(10)	0.6597(62)	0.2691	0.825	0.0026
Nb1	4a	0.0488(3)	0.6813(16)	0.2478	0.767	0.0034
Nb2	4a	0.0924(4)	0.1163(18)	0.2795	0.767	0.0034
Nb3	4a	0.1270(7)	0.7671(33)	0.0041	0.767	0.0034
Nb4	4a	0.1251(8)	0.7551(35)	0.5215	0.767	0.0034
Nb5	4a	0.1756(7)	0.2255(33)	0.0189	0.790	0.0034
Nb6	4a	0.1729(7)	0.2181(35)	0.5120	0.790	0.0034
Ti1	4a	0.0488(3)	0.6813(16)	0.2478	0.067	0.0034
Ti2	4a	0.0924(4)	0.1163(18)	0.2795	0.067	0.0034
Ti3	4a	0.1270(7)	0.7671(33)	0.0041	0.067	0.0034
Ti4	4a	0.1251(8)	0.7551(35)	0.5215	0.067	0.0034
Ti5	4a	0.1756(7)	0.2255(33)	0.0189	0.067	0.0034
Ti6	4a	0.1729(7)	0.2181(35)	0.5120	0.067	0.0034
Ta1	4a	0.0488(3)	0.6813(16)	0.2478	0.083	0.0034
Ta2	4a	0.0924(4)	0.1163(18)	0.2795	0.083	0.0034
Ta3	4a	0.1270(7)	0.7671(33)	0.0041	0.083	0.0034
Ta4	4a	0.1251(8)	0.7551(35)	0.5215	0.083	0.0034
Ta5	4a	0.1756(7)	0.2255(33)	0.0189	0.083	0.0034
Ta6	4a	0.1729(7)	0.2181(35)	0.5120	0.083	0.0034
Sb1	4a	0.0488(3)	0.6813(16)	0.2478	0.083	0.0034
Sb2	4a	0.0924(4)	0.1163(18)	0.2795	0.083	0.0034
Sb3	4a	0.1270(7)	0.7671(33)	0.0041	0.083	0.0034
Sb4	4a	0.1251(8)	0.7551(35)	0.5215	0.083	0.0034
Sb5	4a	0.1756(7)	0.2255(33)	0.0189	0.083	0.0034
Sb6	4a	0.1729(7)	0.2181(35)	0.5120	0.083	0.0034
O1	4a	0.00145	0.3602	0.7621	0.970	0.0034
O2	4a	0.03496	1.0504	0.2677	0.970	0.0034
O3	4a	0.07370	0.7250	-0.0019	0.970	0.0034
O4	4a	0.07667	0.6930	0.5249	0.970	0.0034
O5	4a	0.0732	0.3708	0.2586	0.970	0.0034
O6	4a	0.1042	0.0736	-0.0087	0.970	0.0034
O7	4a	0.1155	0.1155	0.5107	0.970	0.0034
O8	4a	0.1121	0.7767	0.2162	0.970	0.0034
O9	4a	0.1161	0.8233	0.7743	0.970	0.0034
O10	4a	0.1512	0.4917	0.0099	0.970	0.0034
O11	4a	0.1585	0.5255	0.4952	0.970	0.0034
O12	4a	0.1615	0.23688	0.2626	0.970	0.0034
O13	4a	0.1711	0.22718	0.7556	0.970	0.0034
O14	4a	0.1652	0.91376	0.0049	0.970	0.0034
O15	4a	0.1872	0.88808	0.5105	0.970	0.0034
O16	4a	0.2237	0.22624	0.0124	0.970	0.0034
O17	4a	0.2283	0.26943	0.5018	0.970	0.0034

Space group: $P2_1nb$; $a = 0.78222(4)$ nm, $b = 3.2917(2)$ nm, and $c = 0.64633(5)$ nm, $R_{wp} = 9.35\%$, $R_p = 6.65\%$, $S = 1.07$. The primary profile parameter (PPP) for 020, 202, and 402 reflections are refined.

Table S3. Experimental and calculated lattice parameters of KNO and MKNO-rdm-a1.

formula	KNO		MKNO	MKNO-rdm-a1
	Exp.	Theo.	Exp.	Theo.
space group	$P2_1nb$	$P2_1nb$	$P2_1nb$*	$P1$
a	0.78167 nm	0.787638 nm	0.78222 nm	0.78461 nm
b	3.2970 nm	3.310688 nm	3.29173 nm	3.3226 nm
c	0.64727 nm	0.650944 nm	0.64633 nm	0.64711 nm
α	90 º	90 º	90 º	90.039 º
β	90 º	90 º	90 º	89.985 º
γ	90 º	90 º	90 º	90.073 º

*Analyzed based on tha asumption that the introduced Ti, Ta, and Sb species partially occupy the Nb site of $K_4Nb_6O_{17}$.

Table S4. The distortion index *D* (Baur's distortion index),[S5] the quadratic elongation $\langle\lambda_{oct}\rangle$,[S6] and bond angle variance σ_{oct}^2 of the O-M-O bond angles[S6] for MO_6 octahedra (M = Nb, Ti, Ta, Sb) in pristine $K_4Nb_6O_{17}$ and MKNO-rdm-a1–10. The index values for $K_4Nb_6O_{17}$ are taken as the average value in the unit cell. For crystallographically equivalent sites, the oxygen-metal bond lengths, *D*, $\langle\lambda_{oct}\rangle$, and σ_{oct}^2 of MKNO model that are shorter compared to those of $K_4Nb_6O_{17}$ are indicated in blue, while these index values for MKNO are higher are indicated in red.

Sample	Nb-site	d_{Nb-O} / Å	*D* (10^{-2})	$\langle\lambda_{oct}\rangle$	σ_{oct}^2 (deg^2)
KNO	Nb-1	2.038	6.4	1.03	95.5
	Nb-2	2.038	6.3	1.03	95.0
	Nb-3	2.009	3.9	1.02	59.4
	Nb-4	2.009	3.9	1.02	60.2
	Nb-5	2.028	6.8	1.05	137.5
	Nb-6	2.044	7.0	1.06	164.7
MKNO-rdm-a1 layer 4	Nb-1	2.024	5.8	1.03	84.7
	Nb-2	2.020	5.6	1.03	84.4
	Sb-3	1.988	0.9	1.01	22.4
	Ta-4	1.987	2.7	1.01	37.0
	Ti-5	2.016	8.3	1.06	150.9
	Nb-6	2.027	6.8	1.06	154.7
MKNO-rdm-a1 layer3	Nb-1	2.030	6.7	1.04	105.0
	Nb-2	2.029	6.6	1.04	104.3
	Nb-3	1.996	3.9	1.02	62.0
	Nb-4	1.996	3.9	1.02	62.5
	Nb-5	2.012	6.6	1.05	133.8
	Nb-6	2.022	6.8	1.06	158.4
MKNO-rdm-a1 layer 2	Nb-1	2.025	6.5	1.04	95.7
	Nb-2	2.025	6.5	1.04	95.9
	Nb-3	1.995	3.9	1.02	58.4
	Nb-4	1.995	3.9	1.02	58.9
	Nb-5	2.014	6.8	1.05	133.1
	Nb-6	2.029	7.1	1.06	164.1
MKNO-rdm-a1 layer 1	Nb-1	2.023	6.4	1.03	90.6
	Nb-2	2.022	6.4	1.03	91.0
	Nb-3	1.995	4.3	1.02	57.6
	Nb-4	1.995	4.3	1.02	58.3
	Nb-5	2.015	7.0	1.05	133.2
	Nb-6	2.034	7.3	1.06	169.2
MKNO-rdm-a2 layer4	Nb-1	2.033	6.5	1.04	98.1
	Nb-2	2.028	6.9	1.04	104.1
	Sb-3	1.991	1.3	1.01	31.3
	Nb-4	1.995	4.6	1.02	68.4
	Nb-5	2.014	6.5	1.05	136.3
	Nb-6	2.028	6.8	1.06	166.0
MKNO-rdm-a2 layer3	Nb-1	2.023	6.4	1.03	89.2
	Nb-2	2.022	6.3	1.03	88.6
	Nb-3	1.996	4.2	1.02	58.0

	Nb-4	1.995	4.2	1.02	58.3
	Nb-5	2.016	7.0	1.05	133.9
	Nb-6	2.035	7.3	1.06	171.3
MKNO-rdm-a2 layer2	Nb-1	2.004	6.1	1.03	80.0
	Nb-2	1.988	6.1	1.03	83.6
	Ti-3	2.018	4.3	1.02	66.3
	Nb-4	2.019	5.1	1.02	64.7
	Nb-5	2.008	6.0	1.04	117.2
	Nb-6	2.019	6.1	1.05	139.6
MKNO-rdm-a2 layer1	Nb-1	2.032	6.9	1.04	108.5
	Nb-2	2.033	6.7	1.04	108.8
	Nb-3	1.994	4.2	1.02	63.1
	Ta-4	1.996	3.2	1.02	53.7
	Nb-5	2.013	6.5	1.05	135.7
	Nb-6	2.024	6.9	1.06	157.8
MKNO-rdm-a3 layer 4	Nb-1	2.025	6.5	1.04	95.7
	Nb-2	2.025	6.5	1.04	96.0
	Nb-3	1.995	4.0	1.02	59.0
	Nb-4	1.995	3.9	1.02	58.4
	Nb-5	2.014	6.8	1.05	132.8
	Nb-6	2.030	7.1	1.06	165.4
MKNO-rdm-a3 layer 3	Nb-1	2.024	6.3	1.03	90.1
	Nb-2	2.023	6.4	1.03	90.2
	Ta-3	1.993	3.2	1.02	48.7
	Nb-4	1.995	4.5	1.02	60.1
	Nb-5	2.016	7.1	1.05	135.6
	Nb-6	2.036	7.5	1.06	174.8
MKNO-rdm-a3 layer 2	Nb-1	2.020	6.2	1.03	79.6
	Nb-2	2.018	6.1	1.03	82.8
	Ti-3	1.988	4.3	1.02	66.4
	Nb-4	2.004	**5.1**	1.02	65.5
	Nb-5	2.008	6.0	1.04	115.1
	Nb-6	2.019	6.1	1.05	139.3
MKNO-rdm-a3 layer 1	Nb-1	2.041	7.0	1.04	109.1
	Nb-2	2.035	7.2	1.04	116.1
	Sb-3	1.992	1.5	1.01	34.2
	Nb-4	1.996	4.9	1.02	73.0
	Nb-5	2.012	6.1	1.05	137.5
	Nb-6	2.021	6.5	1.06	156.3
MKNO-rdm-a4 layer 4	Nb-1	2.020	6.2	1.03	81.3
	Nb-2	2.019	5.9	1.03	85.4
	Ti-3	1.989	4.9	1.02	66.8
	Ta-4	2.000	3.8	1.02	52.8
	Nb-5	2.008	6.0	1.04	118.4
	Nb-6	2.020	6.3	1.05	145.8
MKNO-rdm-a4 layer 3	Nb-1	2.031	6.8	1.04	107.0
	Nb-2	2.030	6.6	1.04	104.7
	Nb-3	1.996	4.0	1.02	61.6
	Nb-4	1.996	4.0	1.02	61.5

	Nb-5	2.011	6.5	1.04	131.8
	Nb-6	2.024	6.8	1.06	156.7
MKNO-rdm-a4 layer 2	Nb-1	1.994	6.5	1.03	94.7
	Nb-2	1.995	6.4	1.03	95.4
	Nb-3	2.025	4.0	1.02	59.4
	Nb-4	2.024	3.8	1.02	57.5
	Nb-5	2.013	6.9	1.05	132.8
	Nb-6	2.031	7.1	1.06	166.6
MKNO-rdm-a4 layer 1	Nb-1	2.024	6.7	1.04	96.2
	Nb-2	2.029	6.0	1.04	90.3
	Nb-3	1.989	4.9	1.02	67.3
	Sb-4	1.995	1.3	1.01	29.4
	Nb-5	2.016	6.7	1.05	136.7
	Nb-6	2.034	7.1	1.06	174.2
KNO-TTS-a5 layer4	Nb-1	2.019	6.1	1.03	78.3
	Nb-2	2.018	6.1	1.03	83.7
	Ti-3	1.988	4.3	1.02	66.1
	Nb-4	2.004	5.1	1.02	64.8
	Nb-5	2.007	6.0	1.04	114.9
	Nb-6	2.019	6.1	1.05	139.5
KNO-TTS-a5 layer3	Nb-1	2.031	6.8	1.04	106.6
	Nb-2	2.030	6.7	1.04	105.0
	Nb-3	1.996	3.9	1.02	61.3
	Nb-4	1.996	3.9	1.02	62.5
	Nb-5	2.012	6.5	1.04	133.5
	Nb-6	2.023	6.7	1.06	155.3
KNO-TTS-a5 layer2	Nb-1	2.024	6.5	1.03	94.5
	Nb-2	2.025	6.5	1.04	95.7
	Nb-3	1.995	4.0	1.02	59.3
	Nb-4	1.995	3.9	1.02	58.1
	Nb-5	2.014	6.8	1.05	133.3
	Nb-6	2.030	7.1	1.06	164.8
KNO-TTS-a5 layer1	Nb-1	2.028	6.6	1.04	98.8
	Nb-2	2.030	6.2	1.04	91.8
	Ta-3	1.990	4.0	1.02	58.2
	Sb-4	1.993	1.4	1.01	29.8
	Nb-5	2.017	6.8	1.05	139.1
	Nb-6	2.034	7.2	1.06	174.2
KNO-TTS-a6 layer4	Nb-1	2.025	6.6	1.04	95.7
	Nb-2	2.025	6.4	1.03	94.9
	Nb-3	1.995	4.0	1.02	59.2
	Nb-4	1.994	3.9	1.02	58.6
	Nb-5	2.014	6.9	1.05	133.3
	Nb-6	2.030	7.1	1.06	165.7
KNO-TTS-a6 layer3	Nb-1	2.022	6.4	1.03	88.2
	Nb-2	2.021	6.3	1.03	87.5
	Nb-3	1.995	4.3	1.02	58.3
	Nb-4	1.995	4.2	1.02	57.8
	Nb-5	2.016	7.1	1.05	134.6

	Nb-6	2.036	7.4	1.06	173.5
KNO-TTS-a6 layer2	Nb-1	2.022	6.4	1.03	84.8
	Nb-2	2.024	5.8	1.03	85.5
	Ti-3	1.994	6.3	1.03	81.4
	Sb-4	1.997	1.4	1.01	33.7
	Nb-5	2.010	6.4	1.04	121.0
	Nb-6	2.018	6.5	1.05	143.3
KNO-TTS-a6 layer1	Nb-1	2.033	6.8	1.04	108.3
	Nb-2	2.031	6.8	1.04	107.4
	Ta-3	1.995	3.2	1.02	54.1
	Nb-4	1.996	4.1	1.02	63.6
	Nb-5	2.012	6.5	1.05	134.7
	Nb-6	2.024	6.8	1.06	157.3
MKNO-rdm-a7 layer 4	Nb-1	2.030	6.3	1.04	92.4
	Nb-2	2.027	6.6	1.04	99.0
	Sb-3	1.989	1.4	1.01	30.0
	Ta-4	1.993	4.0	1.02	58.0
	Nb-5	2.015	6.7	1.05	138.3
	Nb-6	2.033	7.1	1.06	173.6
KNO-TTS-a7 layer3	Nb-1	2.025	6.5	1.04	96.0
	Nb-2	2.024	6.4	1.03	95.1
	Nb-3	1.995	3.9	1.02	58.4
	Nb-4	1.995	3.9	1.02	58.8
	Nb-5	2.013	6.8	1.05	133.0
	Nb-6	2.029	7.1	1.06	164.8
KNO-TTS-a7 layer 2	Nb-1	2.030	6.7	1.04	105.4
	Nb-2	2.029	6.6	1.04	105.1
	Nb-3	1.996	3.9	1.02	62.2
	Nb-4	1.996	3.9	1.02	61.9
	Nb-5	2.011	6.5	1.04	133.3
	Nb-6	2.022	6.7	1.06	155.0
KNO-TTS-a7 layer 1	Nb-1	2.017	6.1	1.03	72.5
	Nb-2	2.016	6.0	1.03	72.4
	Nb-3	1.989	4.0	1.02	39.4
	Nb-4	1.989	3.9	1.02	39.7
	Ti-5	2.016	8.6	1.06	149.1
	Nb-6	2.027	6.8	1.05	145.9
KNO-TTS-a8 layer4	Nb-1	2.022	6.4	1.03	90.1
	Nb-2	2.022	6.3	1.03	88.1
	Nb-3	1.995	4.2	1.02	58.0
	Nb-4	1.995	4.2	1.02	58.9
	Nb-5	2.016	7.0	1.05	134.3
	Nb-6	2.035	7.3	1.06	171.3
KNO-TTS-a8 layer3	Nb-1	2.034	6.5	1.04	97.7
	Nb-2	2.028	6.8	1.04	103.1
	Sb-3	1.990	1.3	1.01	30.5
	Nb-4	1.995	4.6	1.02	69.1
	Nb-5	2.014	6.5	1.05	136.3
	Nb-6	2.028	6.8	1.06	166.4

KNO-TTS-a8 layer2	Nb-1	2.030	6.8	1.04	106.0
	Nb-2	2.030	6.7	1.04	106.3
	Nb-3	1.996	4.0	1.02	63.1
	Nb-4	1.996	3.9	1.02	61.7
	Nb-5	2.012	6.5	1.04	133.3
	Nb-6	2.023	6.7	1.06	155.7
KNO-TTS-a8 layer1	**Ta-1**	2.015	5.2	1.03	76.4
	Nb-2	2.019	6.3	1.03	79.7
	Nb-3	1.990	5.0	1.02	65.8
	Ti-4	2.004	4.5	1.02	68.5
	Nb-5	2.008	6.0	1.04	118.1
	Nb-6	2.019	6.0	1.05	141.1
MKNO-rdm-a9 layer 4	Nb-1	2.026	6.5	1.03	95.2
	Nb-2	2.025	6.4	1.03	95.0
	Nb-3	1.995	4.0	1.02	58.9
	Nb-4	1.996	3.9	1.02	59.1
	Nb-5	2.015	6.9	1.05	133.7
	Nb-6	2.031	7.1	1.06	164.5
MKNO-rdm-a9 layer 3	Nb-1	2.023	6.4	1.03	88.5
	Nb-2	2.022	6.3	1.03	88.1
	Nb-3	1.995	4.3	1.02	58.2
	Nb-4	1.996	4.2	1.02	58.5
	Nb-5	2.016	7.1	1.05	134.3
	Nb-6	2.036	7.3	1.06	172.0
MKNO-rdm-a9 layer 2	Nb-1	2.026	6.1	1.03	88.2
	Nb-2	2.022	6.4	1.03	83.7
	Sb-3	1.997	1.4	1.01	33.7
	Ti-4	1.994	6.3	1.03	79.3
	Ta-5	2.009	5.6	1.04	112.1
	Nb-6	2.021	6.3	1.05	147.8
MKNO-rdm-a10 layer 1	Nb-1	2.031	6.8	1.04	105.4
	Nb-2	2.030	6.7	1.04	105.2
	Nb-3	1.996	3.9	1.02	62.2
	Nb-4	1.997	4.0	1.02	62.3
	Nb-5	2.012	6.5	1.04	133.6
	Nb-6	2.024	6.7	1.06	155.4
MKNO-rdm-a10 layer 4	Nb-1	2.035	7.3	1.04	115.4
	Nb-2	2.042	7.1	1.04	110.0
	Nb-3	1.995	4.9	1.02	72.1
	Sb-4	1.991	1.5	1.01	34.9
	Ta-5	2.010	5.4	1.04	126.6
	Nb-6	2.024	6.7	1.06	162.0
MKNO-rdm-a10 layer 3	Nb-1	2.018	6.1	1.03	83.5
	Nb-2	2.019	6.1	1.03	78.5
	Nb-3	1.988	5.0	1.02	64.3
	Ti-4	2.004	4.3	1.02	65.5
	Nb-5	2.007	6.0	1.04	113.7
	Nb-6	2.019	6.1	1.05	139.8
	Nb-1	2.022	6.4	1.03	89.2

MKNO-rdm-a10 layer 2	Nb-2	2.022	6.3	1.03	88.0
	Nb-3	1.995	4.2	1.02	57.4
	Nb-4	1.995	4.2	1.02	57.6
	Nb-5	2.016	7.0	1.05	133.2
	Nb-6	2.036	7.4	1.06	172.7
MKNO-rdm-a10 layer 1	Nb-1	2.025	6.5	1.04	96.0
	Nb-2	2.025	6.4	1.03	95.2
	Nb-3	1.995	3.9	1.02	57.8
	Nb-4	1.995	3.9	1.02	58.2
	Nb-5	2.013	6.8	1.04	131.6
	Nb-6	2.031	7.1	1.06	166.2

Supplementary Figures

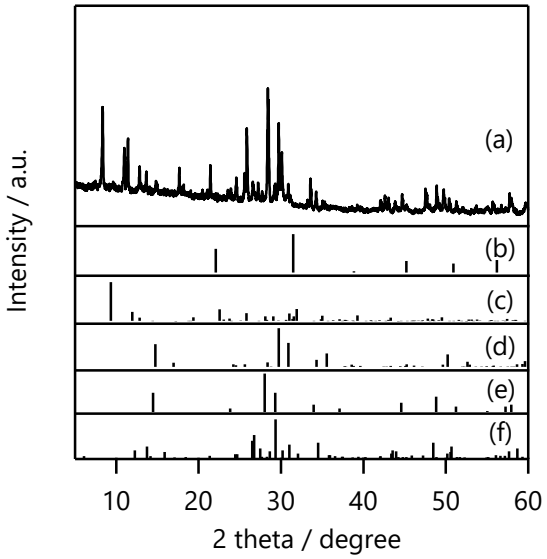


Figure S1. XRD patterns of (a) $(K_4Nb_6O_{17})_{0.5}(Rb_4Ta_6O_{17})_{0.5}$ at 1300 ºC together with those of the references (b) $KTa_{0.77}Nb_{0.23}O_3$ (PDF 01-070-2011), (c) $Rb_6(H_2Nb_6O_{19})\cdot 19H_2O$ (PDF 01-077-4941), (d) Ta_2O_5 (PDF 00-054-0514), (e) $Rb_4Ta_{10}O_{27}$ (PDF 00-032-0951), and (f) $Rb_{4.9}Ta_{11.02}O_{30}$ (PDF 01-076-0977). The holding temperature, 1300 °C; solute concentration, 5 mol %.

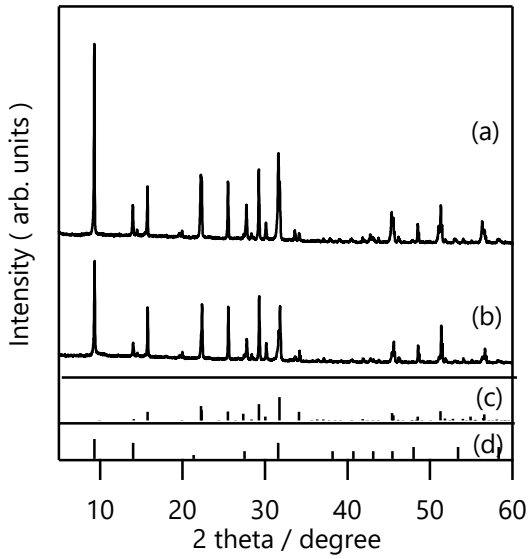


Figure S2. XRD patterns of $K_4Nb_3Ta_3O_{17}$ crystals grown from K_2MoO_4 at 1100 ºC prepared through (a) the flux cooling method, and (b) the quenching method, together with those of the reference (c) $K_6Nb_{10.88}O_{30}$ (PDF 01-087-1856) and (d) $K_4Nb_6O_{17}$ $3H_2O$ (PDF 00-021-1297)**.**

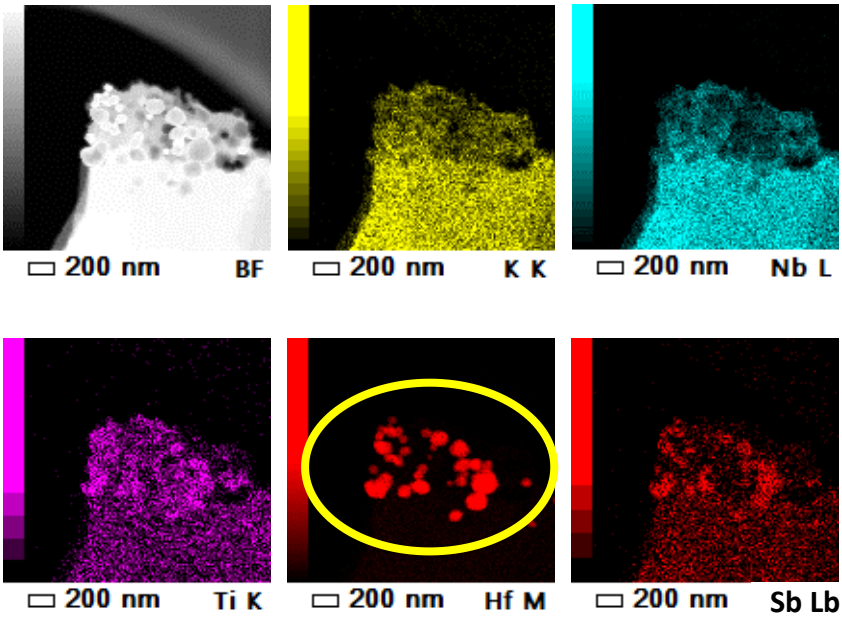


Figure S3. Dark field TEM image and its EDX mapping of $TiTaSbHf_{0.5}$-Q prepared through quenching for K, Nb, Ti, Hf, and Sb using the *K* and *L* line characteristic X-ray. Only Hf species is segrigated in the $TiTaSb_{0.5}$-Q.

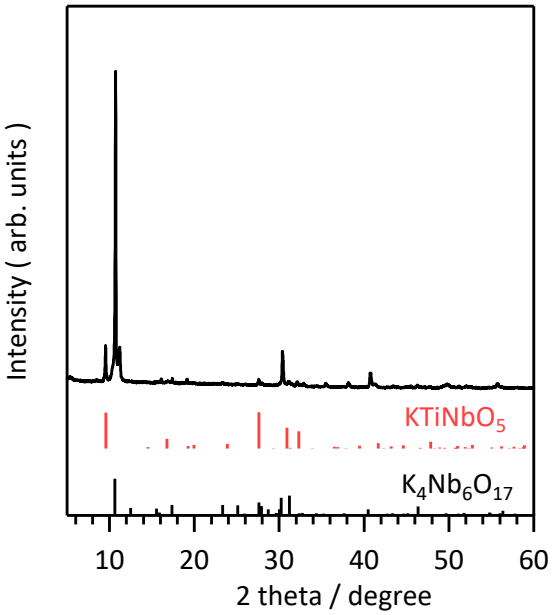
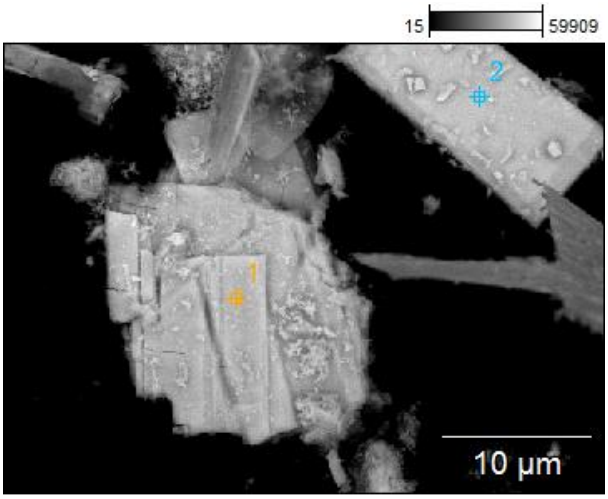
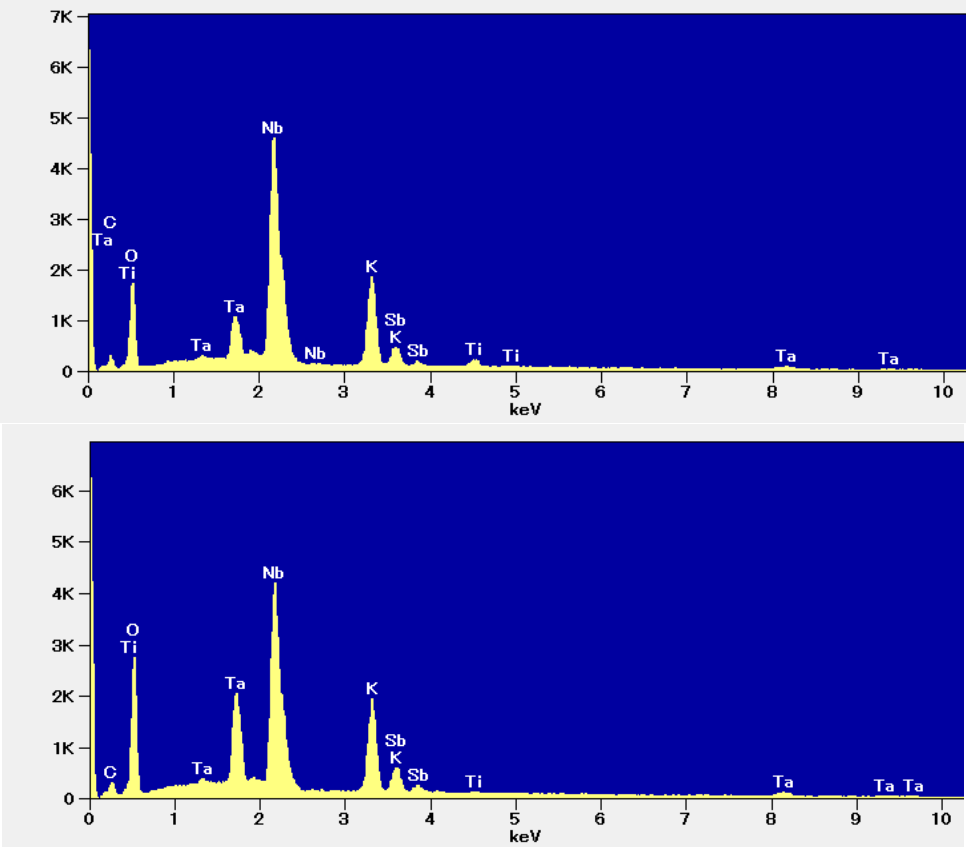


Figure S4. XRD pattern of $K_4Nb_{4.2}Ti_{0.6}Ta_{0.6}Sb_{0.6}O_{17}$ prepared through quenching ($TiTaSb_{0.6}$-Q).

a)



b)



c)

	Atomic ratio (at %)					
	O-K	***K-K***	***Ti-K***	***Nb-L***	***Sb-L***	***Ta-L***
$TiTaSb_{0.5}$-Q_*point1*	53.60	8.18	1.12	10.54	0.89	1.21
$TiTaSb_{0.5}$-Q_*point2*	64.18	7.89	0.22	9.27	1.37	2.09

Figure S5. (a) FE-SEM image and (b,c) EDX mapping results of $TiTaSb_{0.5}$-Q prepared through quenching.

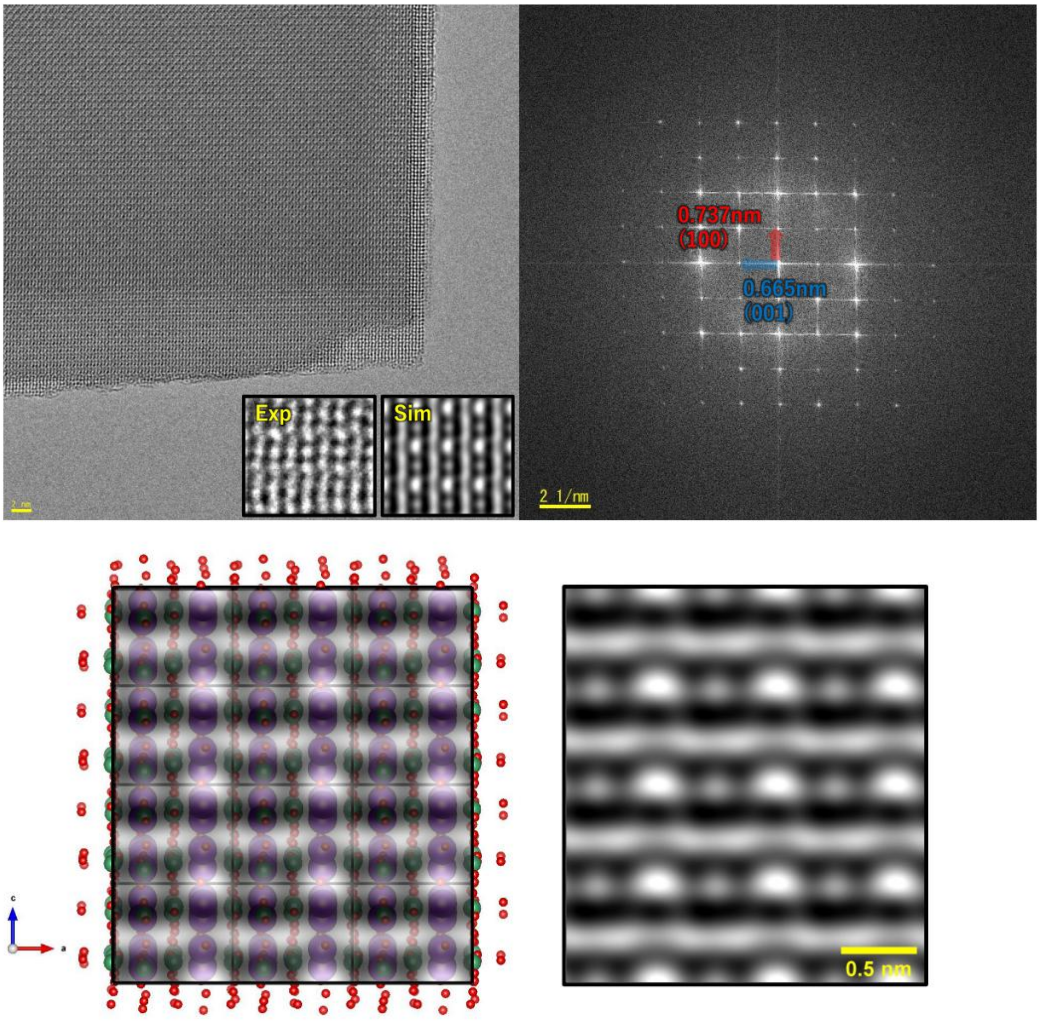
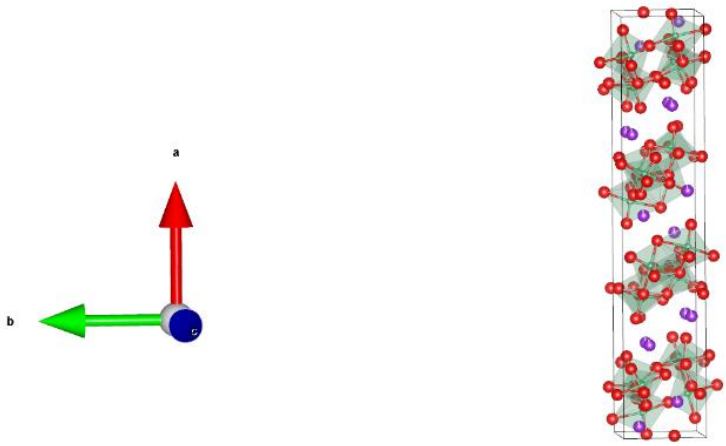


Figure S6. TEM image of pristine $K_4Nb_6O_{17}$ crystals grown from a K_2MoO_4 flux at 1100 ºC taken by irradiating from the <010> direction. The experimental TEM image matched well with the simulated TEM image, showing that $K_4Nb_6O_{17}$ crystal grows in the *ac*-plane direction.

a)



b)

Figure S7. Structural model of (a) $K_4Nb_6O_{17}$ and (b) $TiTaSb_{0.5}$-Q crystal used for Rietveld analysis. Purple, green, orange, light blue, dark brown, and red spheres represent K, Nb, Ta, Ti, Sb, and O, respectively. Ta, Ti, and Sb species occupy the same sites as Nb species.

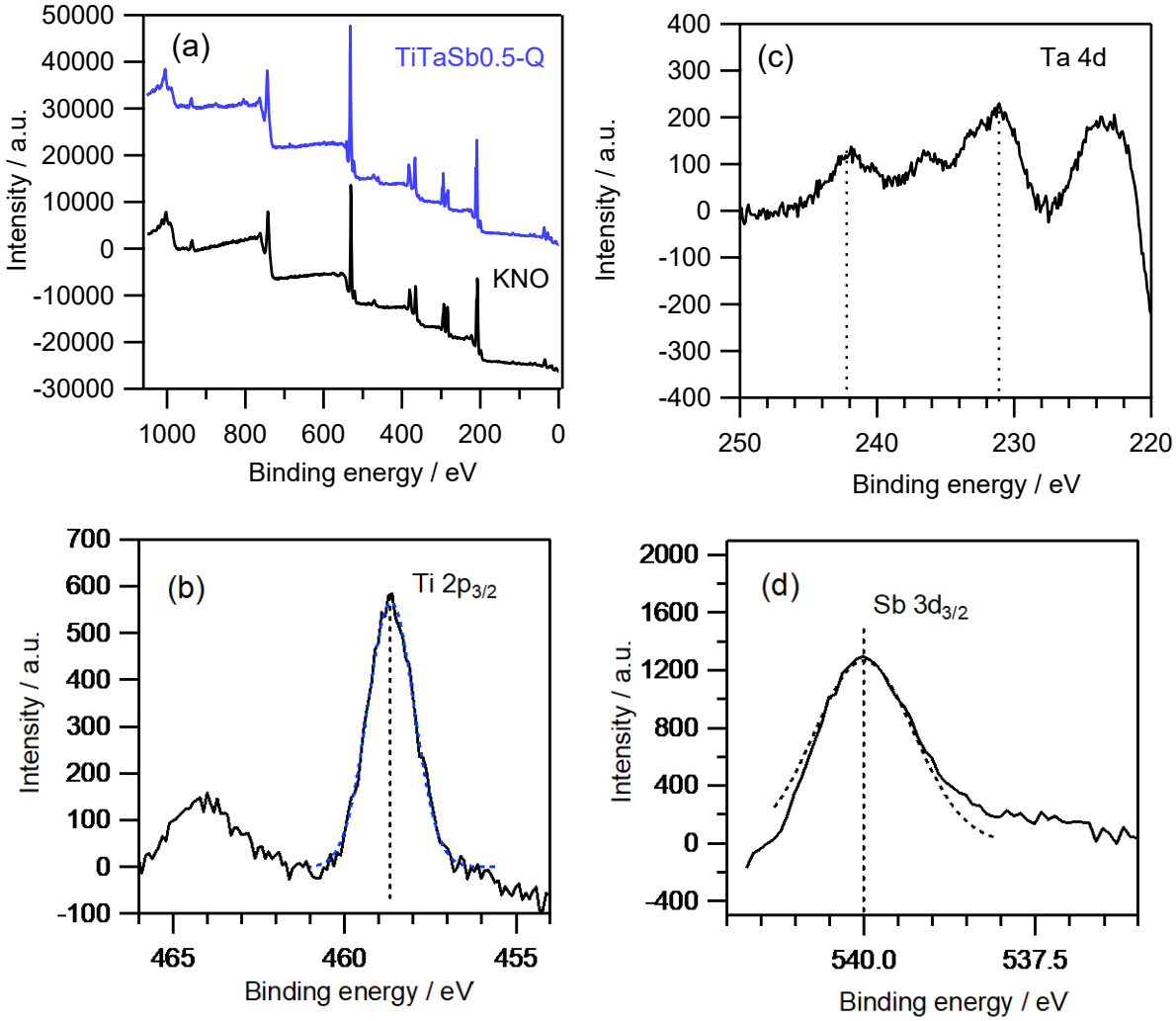


Figure S8. (a) Wide-scan XPS spectra of KNO and $TiTaSb_{0.5}$-Q, and (b) Ti 2p, (c) Ta 4d, and (d) Sb 3d XPS spectra of $TiTaSb_{0.5}$-Q.

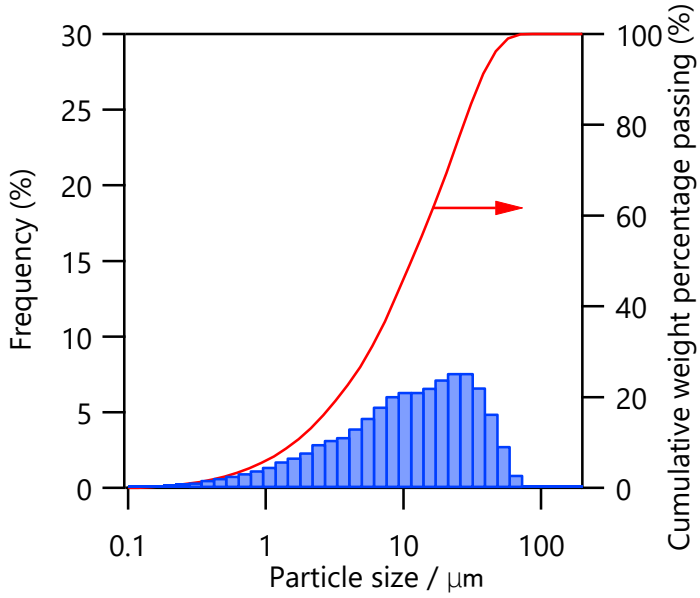


Figure S9. Dynamic light scattering (DLS) particle size distribution of $TiTaSb_{0.5}$-Q nanosheet colloid.

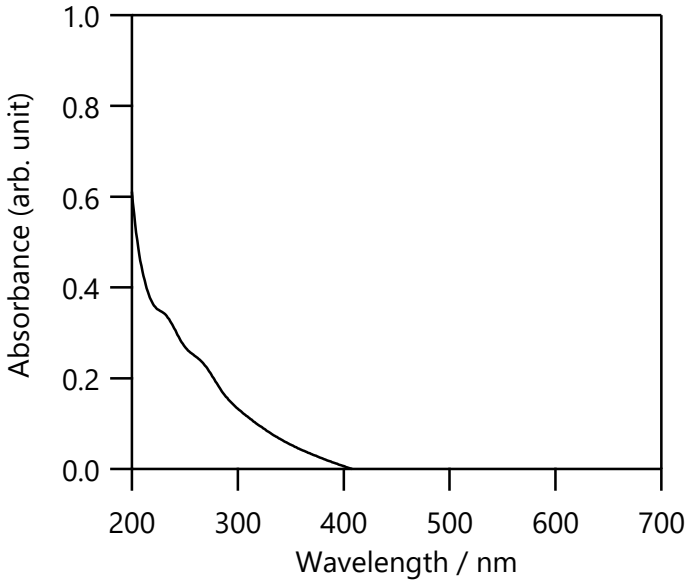


Figure S10. UV-Vis spectrum of $TiTaSb_{0.5}$-Q nanosheet colloid.

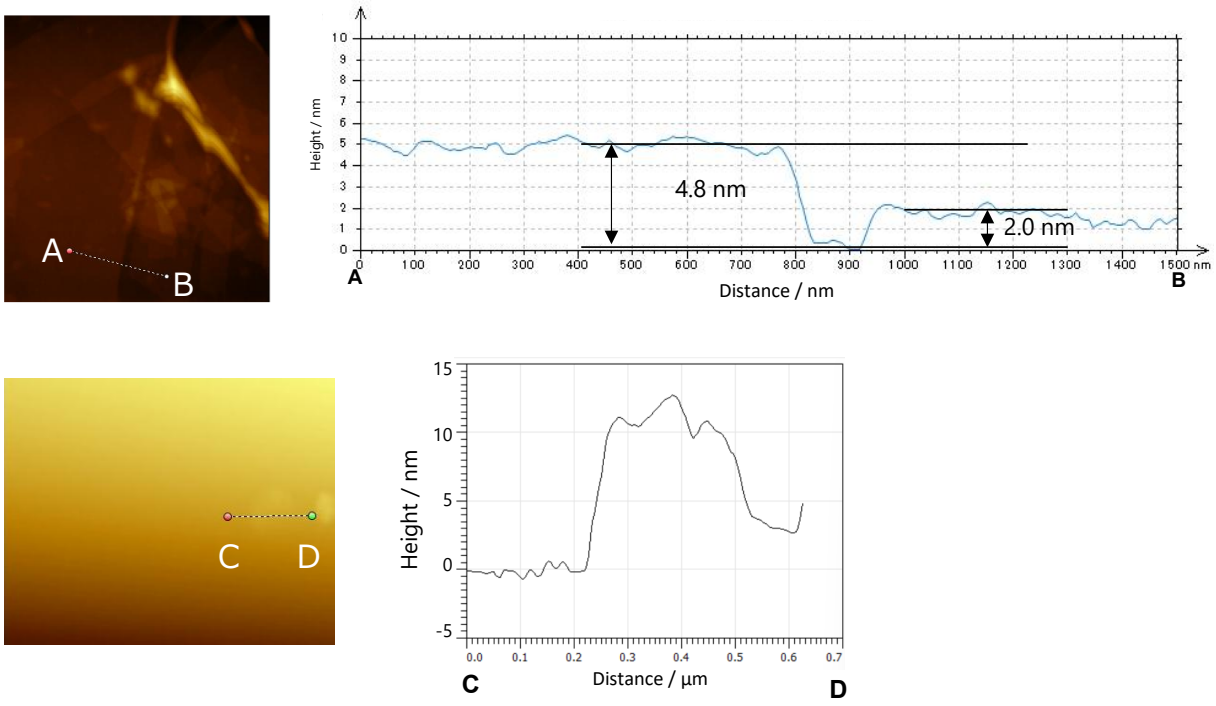


Figure S11. AFM image of $TiTaSb_{0.5}$-Q nanosheet prepared without use of exfoliation reagent and the corresponding thickness profile.

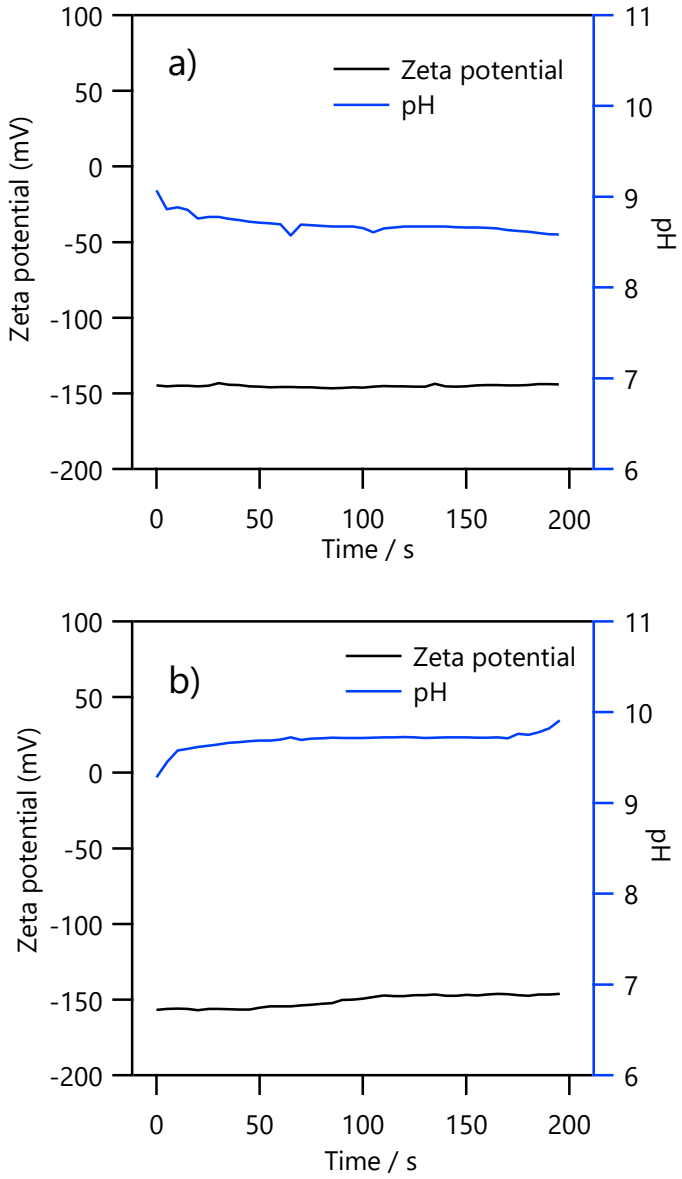


Figure S12. Zeta potential profiles with pH changes for (a) the pristine KNO and (b) $TiTaSb_{0.5}$-Q nanosheet sanples.

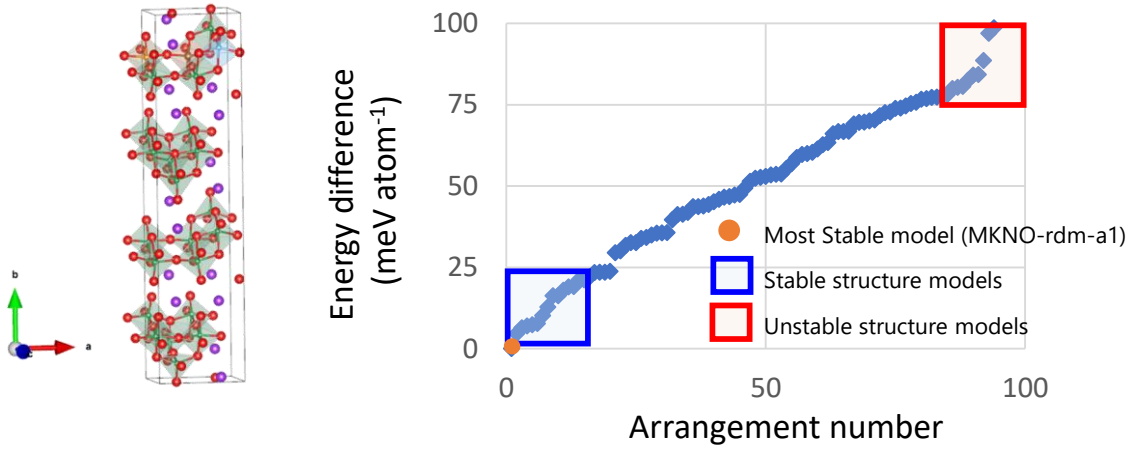


Figure S13. (Left) Crystal structure of MKNO-rdm models ($K_{16}Nb_{21}TiTaSbO_{68}$, Z=4). MKNO-rdm-a1 is shown as a typical example, the structure of which is drawn using the VESTA program.[S1] Purple, green, orange, light blue, dark brown, and red spheres represent K, Nb, Ta, Ti, Sb, and O, respectively. (Right) Total energy differences between stable and unstable structure models of MKNO-rdm.

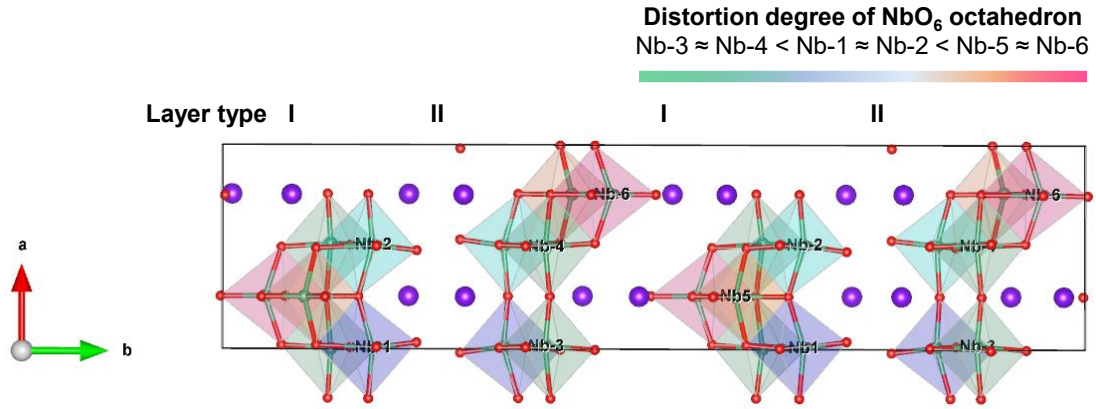


Figure S14. Crystal structure of $K_4Nb_6O_{17}$ in unit cell. The unit cell shows different intraoctahedral distortion degrees, represented in a continuous color scale, according to the literature.[S2] Distorted octahedra centered on niobium (Nb-1~6) are highlighted and their ditortion degrees are shown in color bar.

References:

[S1] K. Momma and F. Izumi, VESTA 3 for three dimensional visualization of crystal, volumetric and morphology, *J. Appl., Crystallogr.* **2011**, 44, 1272–1276.

[S2] Souza, J. K. D.; Duarte, T. M.; dos Santos, I. M. G.; Sambrano, J. R.; Maia, A. d. S.; dos Reis Albuquerque, A. Intra-octahedral distortion on lamellar potassium niobate $K_4Nb_6O_{17}$: a periodic DFT study of structural, electronic and vibrational properties. *Phys. Chem. Chem. Phys.* **2020**, 22, 16562–16570.