Table S1. The calculated mixing enthalpy ΔH_{mix}(eV/Cell) and free energy ΔG_{mix}(eV/Cell) for $Sn_{2-x}Pb_xNb_2O_7$ (x=0, 0.5, 1.0, 1.5, 2.0) under 300, 400, 600, 800, 1000 K.

	T(K)	x=0	x=0.5	x=1.0	x=1.5	x=2.0
ΔH_{mix}	-	0	-0.09	-0.12	-0.09	0
ΔG_{mix}	300	0	-0.10	-0.13	-0.11	0
	400	0	-0.11	-0.14	-0.11	0
	600	0	-0.12	-0.15	-0.12	0
	800	0	-0.13	-0.16	-0.13	0
	1000	0	-0.14	-0.18	-0.14	0

Table S2. The calculated branch point energy E_{BP}, band edge potential E_{NHE}, E_{vac} and ionization energy difference ΔE for $Sn_{2-x}Pb_xNb_2O_7$ (x=0, 0.5, 1.0, 1.5, 2.0).

	x=0	x=0.5	x=1.0	x=1.5	x=2.0
E_{BP}	0.76	0.54	0.27	0.28	0.93
E_{NHE}	2.61	2.48	2.38	2.30	2.66
E_{vac}	-7.11	-6.98	-6.87	-6.80	-7.16
ΔE	0.33	0.26	0.08	0.14	0.77

Table S3. Calculated Mulliken electronegativity of each element in $Sn_{2-x}Pb_xNb_2O_7$ (x=0, 0.5, 1.0, 1.5, 2.0).

element	I(eV)	A(eV)	χ(eV)
Sn	7.34	1.11	4.23
Pb	7.42	0.36	3.89
Nb	6.76	0.92	3.84
O	13.62	1.46	7.54

Table S4. The calculated elastic constant C_{ij}(GPa) and deformation potential constant E(eV)of $Sn_{2-x}Pb_xNb_2O_7$ (x=0, 0.5, 1.0, 1.5, 2.0).

	x=0	x=0.5	x=1.0	x=1.5	x=2.0
C_{11}	139.06	260.23	264.91	257.76	236.09
C_{44}	66.55	74.33	90.73	75.34	88.56
C_{12}	-6.40	97.51	77.94	94.86	103.64
$E_{[100]}$	-	6.20	1.52	16.01	-
$E_{[010]}$	-	6.20	3.01	14.34	-
$E_{[001]}$	7.17	1.34	0.51	3.69	11.42
$E_{[111]}$	2.00	1.34	1.01	0.67	8.45