Supporting Information

Entropy-mediated Pb doping in tungsten bronze $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ oxides for synergistic electron-phonon transport optimization

Jiacheng Cao[a], Min Zhu[a,b*], Xuefeng Lu[a,b], Xiaonan Chen[a]

[a] *School of materials science and engineering, Lanzhou University of Technology, Lanzhou, Gansu, 730050, China*

[b] *State key Laboratory of Advance Processing and Recycling of Non-ferrous Metals, Lanzhou University of Technology, Lanzhou, Gansu, 730050, China*

Table of Contents

Supplementary Notes

The absorption spectrum in UV spectroscopy is represented using the Kubelka-Munk function, denoted as $F(R_\infty)$, and the corresponding formula is presented below [1]:

$$F(R_\infty) = \frac{(1-2R_\infty)^2}{2R_\infty} \quad \text{(Supplementary Equation 1)}$$

where $R_\infty = \frac{R_{sample}}{R_{standard}}$ represents the reflectivity of a semi-infinite thick experiment ($R \to \infty$). The optical band gap can be determined using the following formula:

$$(F(R_\infty)hv)^n = B(hv - E_g) \quad \text{(Supplementary Equation 2)}$$

In this equation, h denotes the Planck constant, v represents the frequency of the emitted photon, B is a proportionality constant, and Eg denotes the band gap energy. The exponent n is determined by the energy band structure associated with the electron transition. Specifically, for direct and indirect band gap materials, n takes the value of 2 or 1/2, respectively.

Calculate the m* value of the sample using the following formulas (S3, S4, and S5) [2]:

$$F_r(\xi) = \int_0^\infty \frac{x^r}{1 + e^{x-\xi}} dx \quad \text{(Supplementary Equation 3)}$$

$$S = -\frac{k_B}{e}\left[\frac{(r+2)F_{r+1}(\xi)}{(r+1)F_r(\xi)} - \xi\right] \quad \text{(Supplementary Equation 4)}$$

$$m^* = \frac{\hbar^2}{2\,k_B T}\left[\frac{n}{4\pi^2 F_{\frac{1}{2}}(\xi)}\right]^{\frac{2}{3}} \quad \text{(Supplementary Equation 5)}$$

where $F_r(\xi)$, ξ and r represent the Fermi integral, chemical formula and scattering parameter respectively.

The weighted mobility for samples is calculated based on their conductivity and Seebeck coefficient using the following formula μ_w [3]:

$$\mu_w = 311 \frac{cm^2}{Vs}\left(\frac{m\Omega\ cm}{\rho}\right)\left(\frac{T}{300K}\right)^{-3/2}\left[\frac{exp^{[}\frac{|S|}{k_B}-2]}{1+exp\left[-5\left(\frac{|S|}{k_{B/e}}-1\right)\right]}+\frac{\frac{3\ |S|}{\pi^2 k_{B/e}}}{1+exp\left[5\left(\frac{|S|}{k_{B/e}}-1\right)\right]}\right]$$

(Supplementary Equation 6)

Supplementary Figures

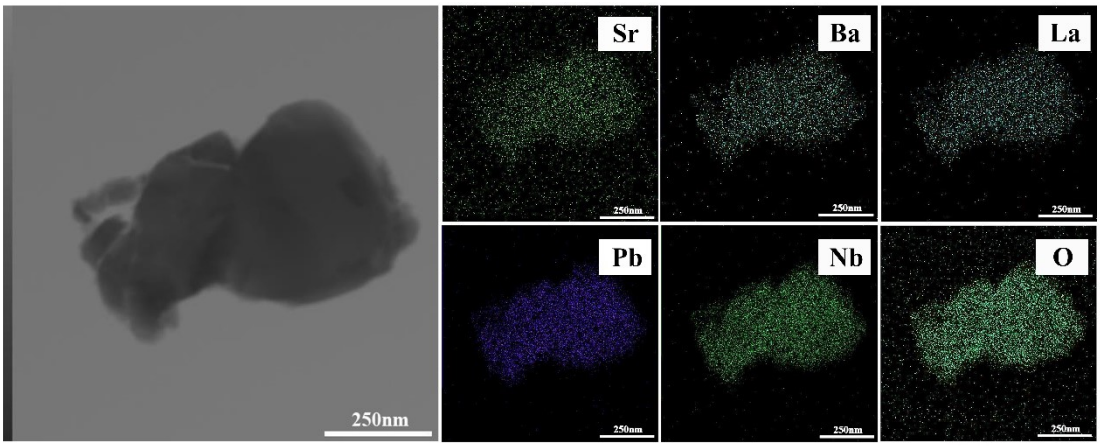

Fig. S1. TEM-EDS of SBLP10 sample.

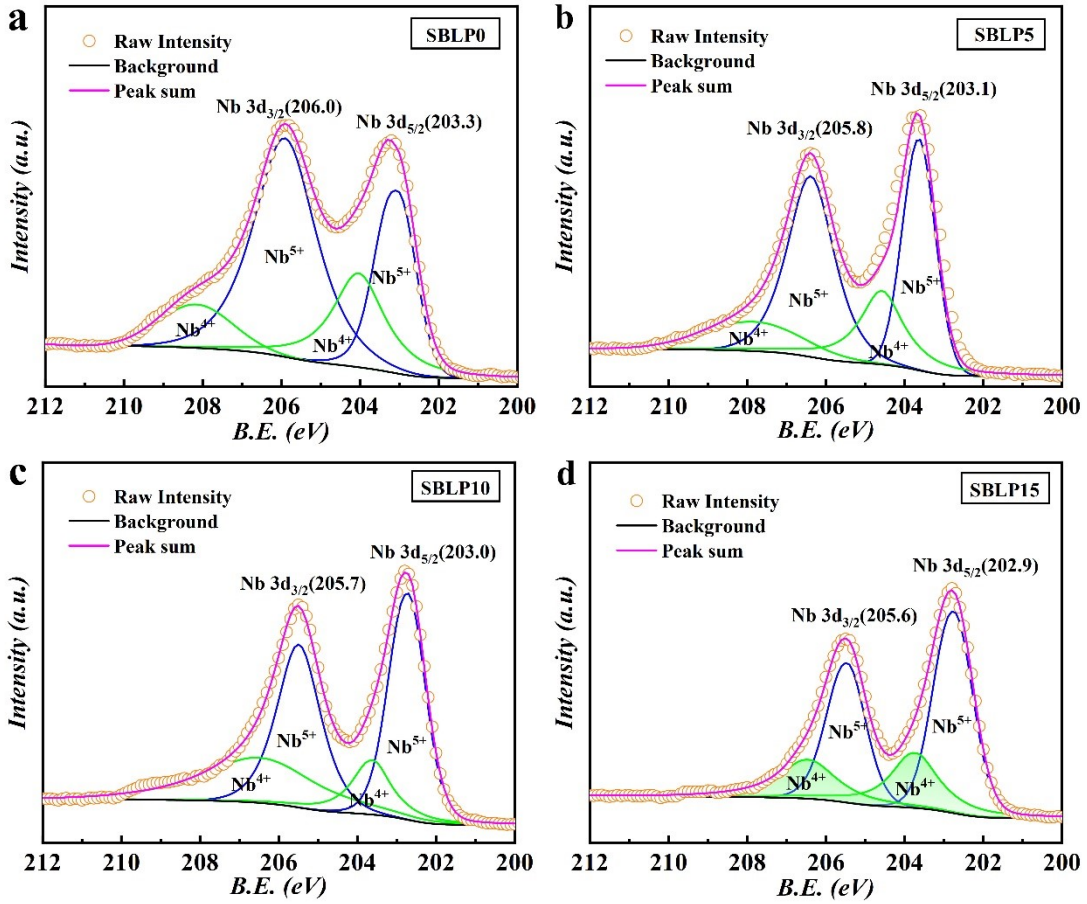

Fig. S2. XPS Nb 3d spectra of $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramic samples: (a) SBLP0; (b) SBLP5; (c) SBLP10; (d) SBLP15.

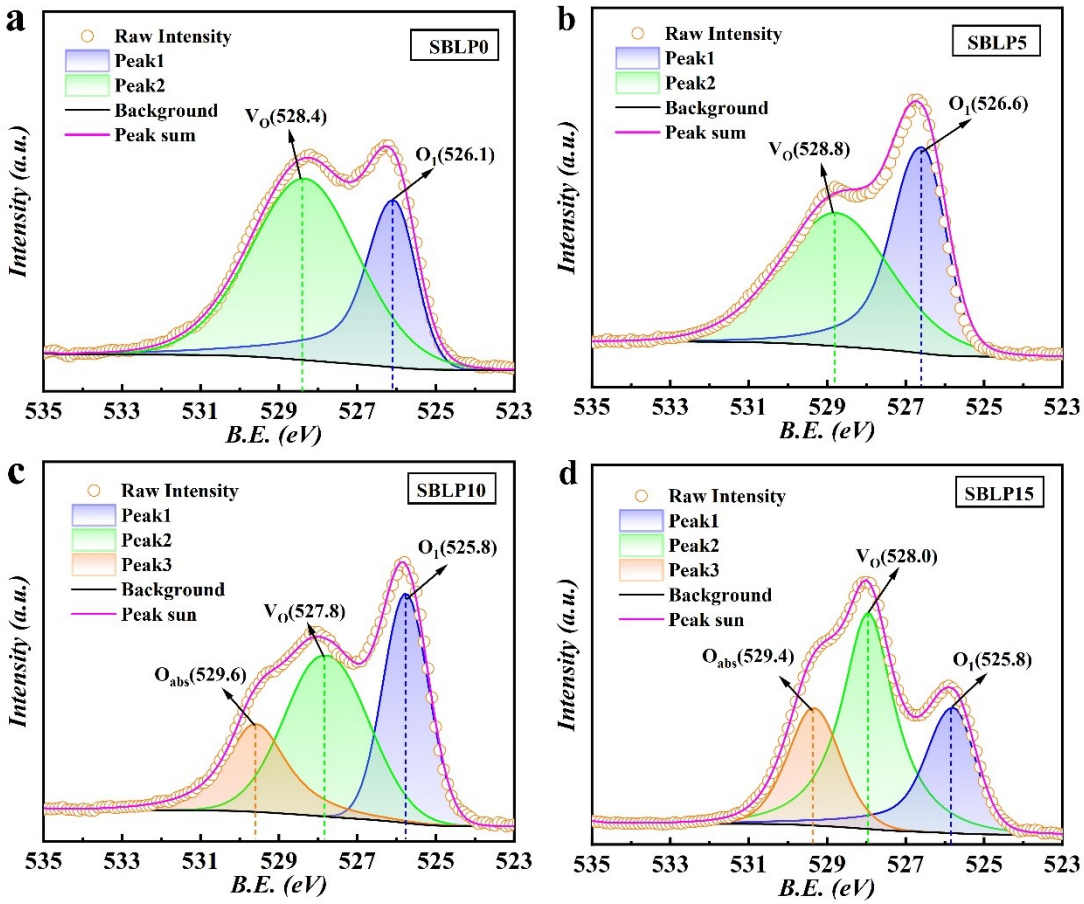

Fig. S3. XPS O 1S spectra of $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramic samples: (a) SBLP0; (b) SBLP5; (c) SBLP10; (d) SBLP15.

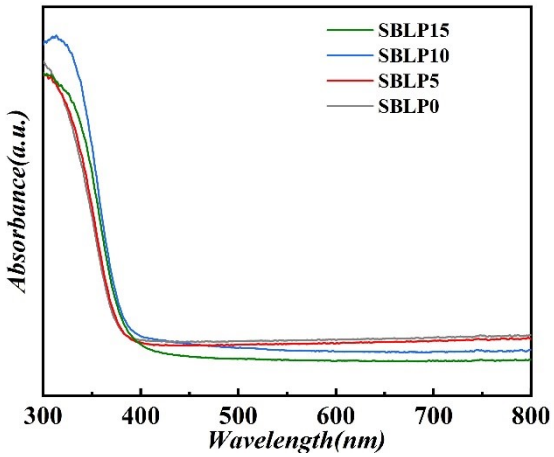

Fig. S4. Diffuse reflectance spectra of $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramic samples.

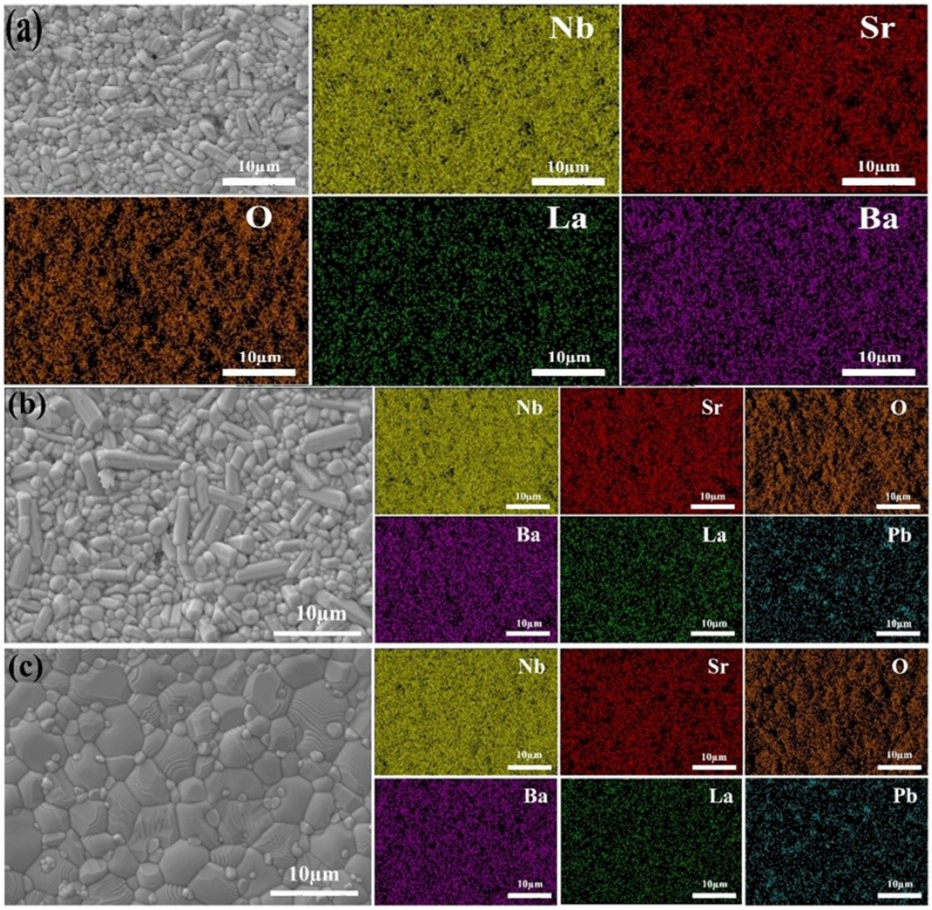

Fig. S5. Surface and elemental distribution maps of $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramic samples: (a) SBLP0; (b) SBLP5; (c) SBLP15.

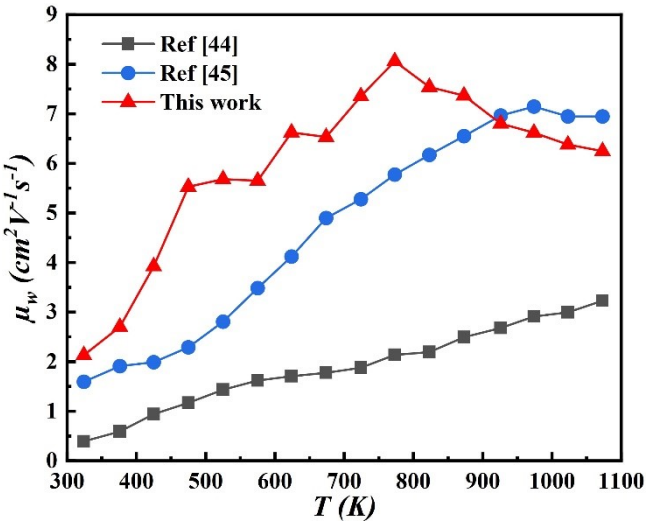

Fig. S6. Comparison of weighted mobility (μ_w) of SBLP10 ceramic sample with others.

Supplementary Tables

Table S1. Refined structure parameters (a, b, c, α, β, γ), Rwp and Rp for $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramics extracted from Rietveld refinement.

Sample	a=b(Å)	C(Å)	α=β=γ	Rwp(%)	Rp(%)
SBLP0	12.43068	3.91676	90°	5.75	5.3
SBLP5	12.42632	3.89754	90°	6.97	5.8
SBLP10	12.42223	3.8967	90°	8.69	6.5
SBLP15	12.44347	3.89469	90°	9.44	7.6

Table S2. SEM-EDS for $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ Ceramic Samples.

element	SBLP0;at. %	SBLP5;at. %	SBLP10;at. %	SBL15;at. %
Sr	8.33	8.24	9.45	9.03
Ba	4.94	4.99	5.27	4.84
La	2.88	3.15	4.66	5.06
Pb	0	0.83	0.70	0.83
Nb	23.52	23.82	22.92	23.33
O	60.33	58.97	57.00	56.92

Table S3. Carrier transport parameters in $(Sr_{0.5}Ba_{0.3}La_{0.2})_{1-x}Pb_xNb_2O_{6-\delta}$ ceramic samples under ambient temperature conditions.

Sample	ρ(ohm-cm)	μ_H ($cm^2/V\cdot S$)	n($10^{18}cm^{-3}$)	Hall(cm^{-3}/C)	f	m*（m_0）
SBLP0	0.08175	92.03671	0.0830645	-7.52376	0.90100	2.35
SBLP5	0.05536	25.57804	0.44135	-1.41641	0.92620	2.43
SBLP10	0.04230	20.51064	0.720401	-0.86759	0.88130	2.51
SBLP15	0.03579	15.69992	1.112290	-0.56186	0.88700	2.38

Table S4. The entropy value according to the nominal composition.

Sample	Nominal composition	ΔSconf (J /(mol·K))	Entropy (R)
SBLP0	$(Sr_{0.5}Ba_{0.3}La_{0.2})Nb_2O_6$	8.561	1.03
SBLP5	$(Sr_{0.5}Ba_{0.3}La_{0.2})_{0.95}Pb_{0.05}Nb_2O_6$	9.782	1.18
SBLP10	$(Sr_{0.5}Ba_{0.3}La_{0.2})_{0.9}Pb_{0.1}Nb_2O_6$	10.407	1.25
SBLP15	$(Sr_{0.5}Ba_{0.3}La_{0.2})_{0.85}Pb_{0.15}Nb_2O_6$	10.791	1.30

Supplementary References

[1] P. Makuła, M. Pacia, W. Macyk, How to correctly determine the band gap energy of modified semiconductor photocatalysts based on UV-Vis spectra, J. Phys. Chem. Lett. 9 (2018) 6814e7.

[2] T. Okuda, K. Nakanishi, S. Miyasaka, Y. Tokura, Large thermoelectric response of metallic perovskites: $Sr_{1-x}La_xTiO_3$ ($0 \leq x \leq 0.1$), Phys. Rev. B 63 (2001) 113104

[3] G.J. Snyder, A.H. Snyder, M. Wood, R. Gurunathan, B.H. Snyder, C. Niu, Weighted mobility, Adv. Mater. 32 (25) (2020) 2001537.