Supplementary Information

Increased hole mobility in anti-$ThCr_2Si_2$-type La_2O_2Bi co-sintered with alkaline earth metal oxides for oxygen intercalation and hole carrier doping

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Table S1 Crystal structural parameters for La_2O_xBi with different nominal composition (R_{wp}: *R*-factor, R_e: expected *R*-factor, *S*: goodness-of-fit indicator).

La_2O_xBi	x = 1.0	x = 1.1	x = 1.2	x = 1.3	x = 1.4	x = 1.5	x = 1.6	x = 1.7	x = 1.8	x = 1.9	x = 2.0
Phase	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi	La_2O_2Bi
Space group	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*	*I*4/*mmm*
a (Å)	4.1064(2)	4.1004(1)	4.0930(1)	4.0895(2)	4.0860(1)	4.0871(1)	4.0845(1)	4.0844(2)	4.0850(1)	4.0867(1)	4.0885(1)
c (Å)	13.4018(12)	13.5187(7)	13.6298(7)	13.7177(9)	13.7612(5)	13.8370(6)	13.9698(6)	13.9800(10)	13.9856(6)	13.9911(7)	13.9953(8)
c/*a*	3.2636	3.2969	3.33	3.3543	3.3679	3.3855	3.4203	3.4223	3.4236	3.4236	3.4231
La_2O_2Bi (mol%)	51.1	76.8	86.3	89.6	100	100	100	67.9	77.2	54	42.4
LaBi (mol%)	23.8	15.9	13.7	10.4	0	0	0	0	0	0	0
La_2Bi (mol%)	25	7.3	0	0	0	0	0	0	0	0	0
La_2O_3 (mol%)	0	0	0	0	0	0	0	32.1	22.8	46	47.6
R_{wp}	2.358	2.323	2,133	1.736	1.647	1.672	1.742	1.589	1.684	2.025	2.282
R_e	1.401	1.445	1.44	1.436	1.46	1.449	1.407	1.43	1.411	1.781	1.797
S	1.6832	1.6073	1.4809	1.2087	1.1278	1.1545	1.2379	1.114	1.1935	1.2929	1.2694

Table S2 Lattice parameters of Bi impurity phase in La_2O_2Bi (x_{Sr} = 0.16, 0.20, x_{Ba} = 0.10, 0.20). The longer *a*- and *c*- axis lengths for Ba doping were possibly due to the formation of Bi-Ba alloy.[1]

	x_{Sr} = 0.16	x_{Sr} = 0.20	x_{Ba} = 0.10	x_{Ba} = 0.20
Phase	Bi	Bi	Bi	Bi
Space group	*R*-3*m*	*R*-3*m*	*R*-3*m*	*R*-3*m*
a (Å)	4.5508(18)	4.5506(13)	4.5548(12)	4.5504(4)
c (Å)	11.8404(77)	11.8438(49)	11.8586(52)	11.8677(18)

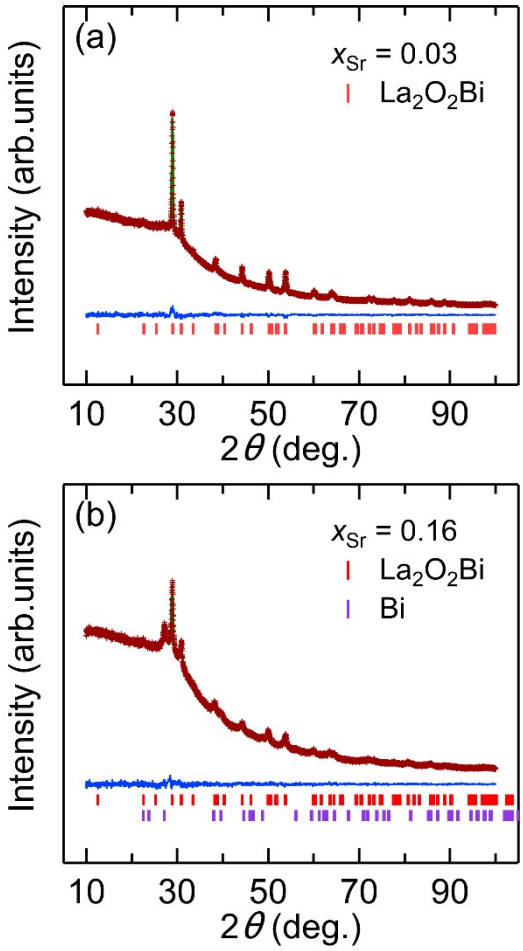

Fig. S1 XRD patterns and the fitting results of Rietveld refinement for (a) x_{Sr} = 0.03, and (b) x_{Sr} = 0.16. Brown, green, and blue curves denote the measurement data, simulation pattern, and their difference, respectively.

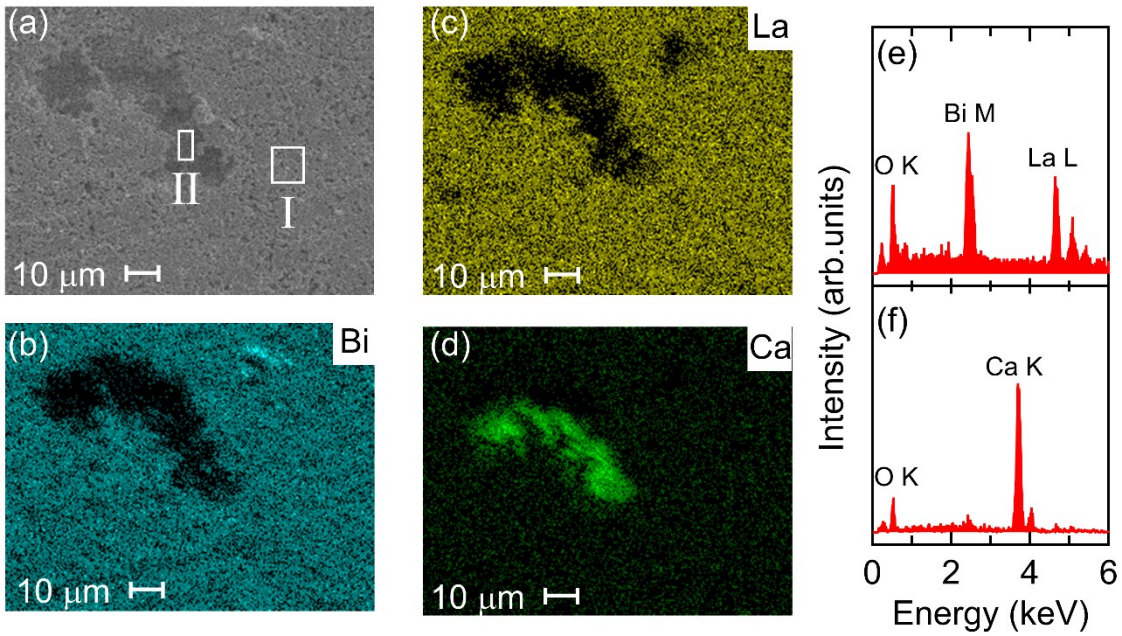

Fig. S2 (a) SEM image and the EDX mappings of (b) Bi, (c) La, (d) Ca for La_2O_2Bi (x_{Ca} = 0.20). EDX spectra in the selected areas of (e) I and (f) II in (a). Ca was segregated locally, and La and Bi were distributed homogeneously.

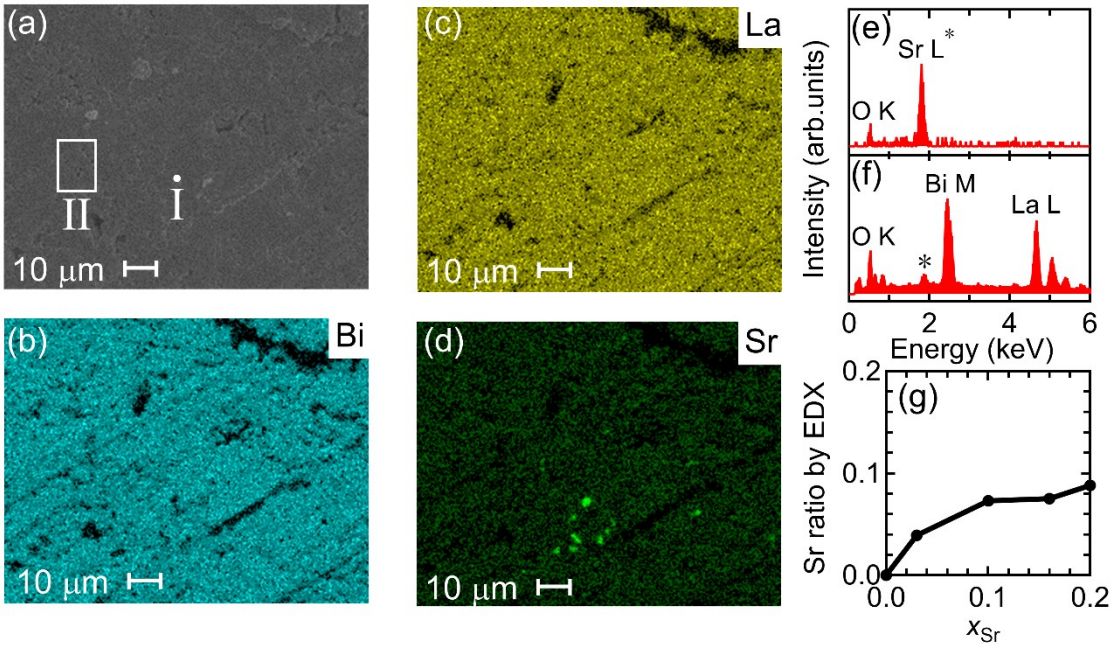

Fig. S3 (a) SEM image and the EDX mappings of (b) Bi, (c) La, (d) Sr for La_2O_2Bi (x_{Sr} = 0.10). EDX spectra (e) at the point I on a SrO grain and (f) in the area II displayed in (a). Bi and La peaks were absent in (e) because only the SrO grain was excited by the point focused measurement.[2] (g) Sr substitution ratio calculated by Sr/(Sr + La) from EDX as a function of x_{Sr}. Sr was distributed almost homogeneously with a tiny segregation. The solubility limit of Sr was about 0.09.

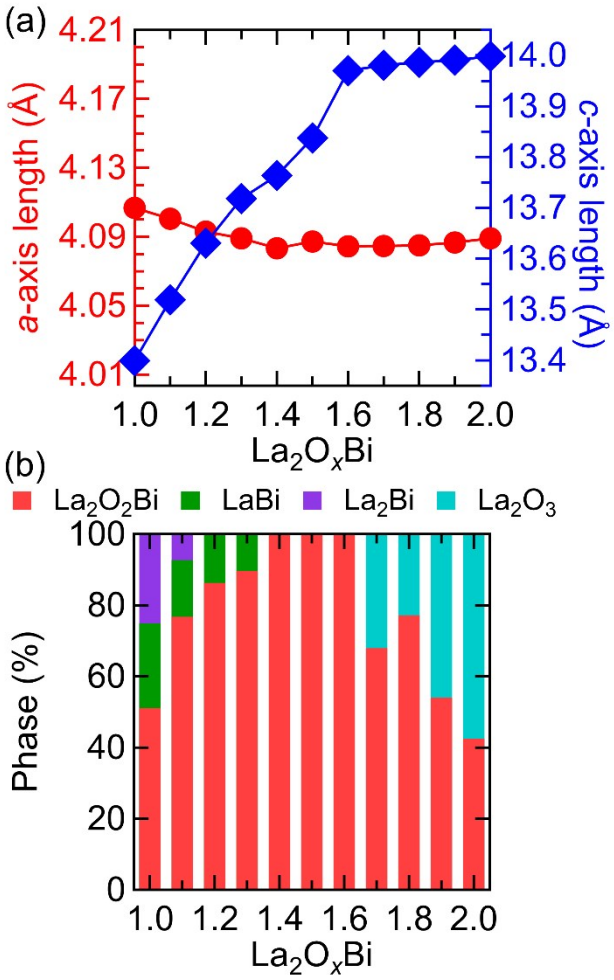

Fig. S4 (a) *a*- and *c*-axis lengths and (b) molar fractions of constituent phases for La_2O_xBi synthesized with different nominal composition.

Supplementary reference

1 T. Lichtenstein, N. D. Smith, J. Gesualdi, K. Kumar and H. Kim, *Electrochim. Acta*, 2017, **228**, 628–635.

2 J. I. Goldstein, C. E. Lyman, D. E. Newbury, E. Lifshin, P. Echlin, L. Sawyer, D. C. Joy and J. R. Michael, *Scanning Electron Microscopy and X-Ray Microanalysis*, Springer, New York, 3rd ed., 2003.