

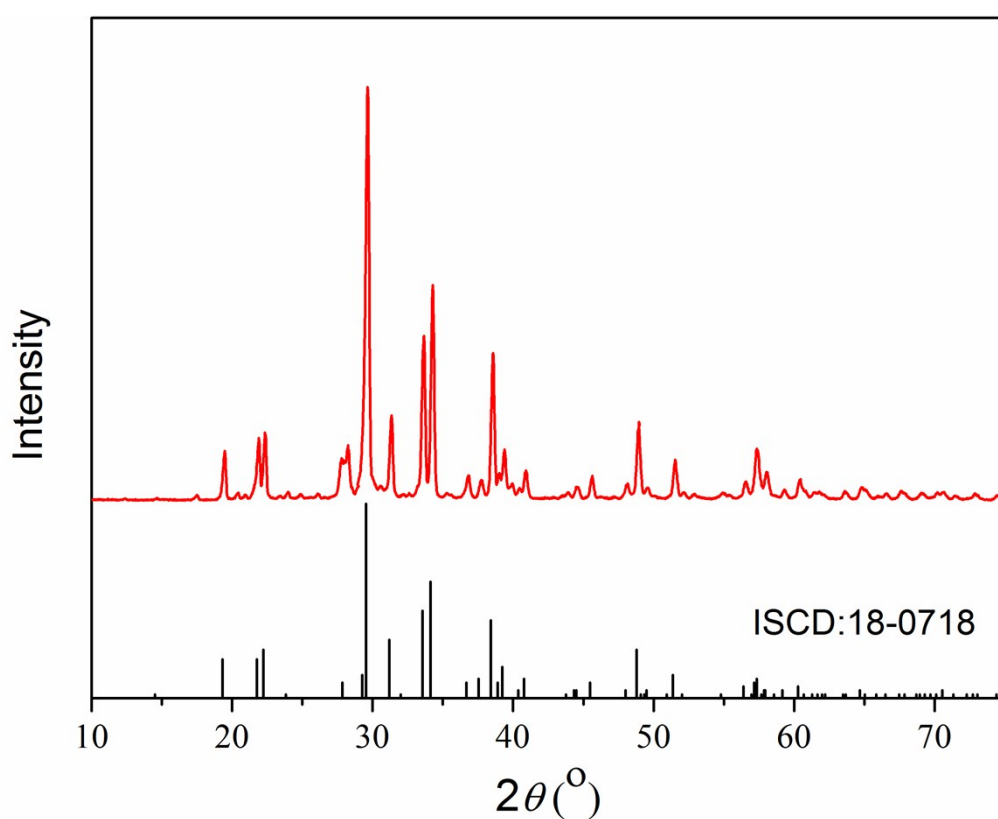


Figure S1. X-ray diffraction patterns of the as-synthesized LBO

It is obvious that main peaks are good in accordance to the standard pattern of LBO (ICSD#180718), indicating that LBO is prepared successfully.

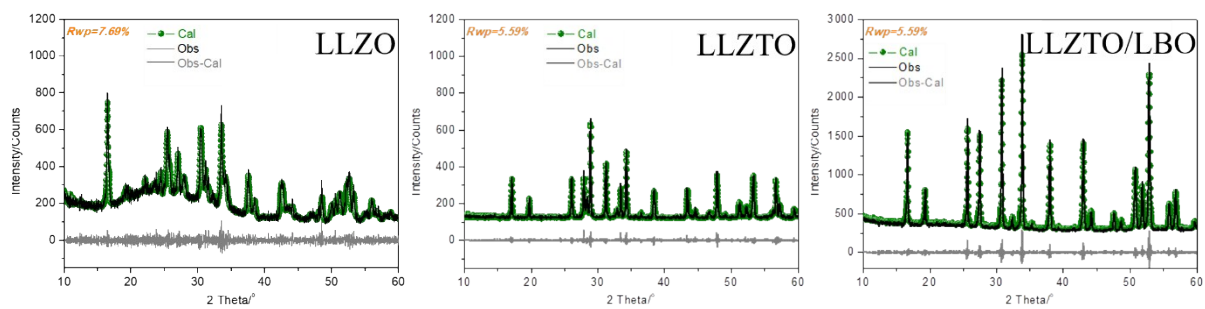


Figure S2. The refinement of XRD plots for LLZO, LLZTO, LLZTO/LBO

Table S1. Crystallographic information for composites of LBO/LLZTO with a space group of *Ia-3d* for (a) 5 h, (b) 10 h, and (c) 20 h milling time.

LBO/LLZTO with a space group of <i>Ia-3d</i> for 5 h, a = 12.9310(1) Å, wRp = 3.90%				
Atom	Position	Uiso*100	Occupancy	Wyckoff position
Li1	0.375, 0, 0.25	0.8(6)	0.58(4)	24d
Li2	0.686(1), 0.581(2), 0.098(2)	1.7(6)	0.37(1)	96h
La	0.125, 0, 0.25	0.55(7)	1	24c
Zr	0, 0, 0	0.3(1)	0.7	16a
Ta	0, 0, 0	0.3(1)	0.3	16a
O	-0.0303(2), 0.0532(2), 0.1483(2)	1.22(7)	1	96h
LBO/LLZTO with a space group of <i>Ia-3d</i> for 10 h, a = 12.9325(5) Å, wRp = 3.64%				
Atom	Position	Uiso*100	Occupancy	Wyckoff position
Li1	0.375, 0, 0.25	1.3(6)	0.59(3)	24d
Li2	0.687(2), 0.578(2), 0.095(1)	1.7(5)	0.38(1)	96h
La	0.125, 0, 0.25	0.38(8)	1	24c
Zr	0, 0, 0	0.1(1)	0.7	16a
Ta	0, 0, 0	0.1(1)	0.3	16a
O	-0.0306(2), 0.0547(3), 0.1477(3)	1.21(7)	1	96h
LBO/LLZTO with a space group of <i>Ia-3d</i> for 20 h, a = 12.9305(5) Å, wRp = 3.42%				
Atom	Position	Uiso*100	Occupancy	Wyckoff position
Li1	0.375, 0, 0.25	5(1)	0.66(6)	24d
Li2	0.686(2), 0.580(2), 0.102(2)	1.0(6)	0.36(1)	96h
La	0.125, 0, 0.25	0.74(8)	1	24c
Zr	0, 0, 0	0.6(1)	0.7	16a
Ta	0, 0, 0	0.6(1)	0.3	16a
O	-0.0305(2), 0.0545(3), 0.1475(3)	1.16(7)	1	96h