ENHANCEMENT OF THERMOELECTRIC PROPERTIES OF $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

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Abstract

The n–$Sr_{1-x}La_xTiO_3$ (x = 0, 0.08 and 0.13) materials were synthesized by solid state reaction method to study thermoelectric properties. The n–$Sr_{1-x}La_xTiO_3$ was synthesized form $SrCO_3$, TiO_2 and La_2O_3. The powders were mixed by ball–milling for 2 h, calcined at 1123 K for 12 h, pressed into pellet at 4.9 MPa, and sintered at 1773 K for 12 h. It results show that the crystal structure is a single perovskite phase and agrees with PDF#2 00–035–0734. The calculated lattice parameter values of x = 0, 0.08 and 0.13 are 0.3905, 0.3906 and 0.3902 nm, respectively. All the pellets of n–$Sr_{1-x}La_xTiO_3$ have density more than 95%. The La substitution at x = 0.13 gave the best thermoelectric properties and appropriated thermoelectric module fabrication.

Keywords: Strontium titanate, solid state reaction, synthesis, substitution, thermoelectric material

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Introduction

Thermoelectric material can change thermal to electricity. Now, high efficiency of thermoelectric material was synthesized form semiconductor of Te–Sb–Ge base due to high dimensionless figure of merit (ZT; $ZT=S^2T/\sigma\kappa$; S is Seebeck coefficient, T is temperature, σ is electrical resistivity and κ is thermal conductivity) (Park, 2013). However, these materials have high toxic and low stability (Wang *et al.*, 2008). Oxide material has stability and nontoxic, although the ZT of this material is lower than Te–Sb–Ge base. Such problem can be solved by dope with higher valence ions (Fergus, 2012). In this work was synthesized $SrTiO_3$ and substitute La in Sr site for enhancement thermoelectric property.

Methods

The n–$Sr_{1-x}La_xTiO_3$ (x = 0, 0.08 and 0.13) were synthesized form $SrCO_3$ (98%, Sigma–Aldrich, St. Louis, MO, USA), TiO_2 (99%, Ajax Finechem Pty. Ltd, New Zealand) and La_2O_3 (99.90%, Sigma–Aldrich, St. Louis, MO, USA). The powders were mixed by ball–milling for 2 h. The mixed powders were calcined at 1123 K for 12 h and pressed into pellet at 156 MPa. The pellets were sintered at 1773 K for 12 h in atmosphere. The crystal structure was characterized by X–ray diffraction (XRD; Shimadzu 6100, Japan) method using CuKα radiation at 40 kV, 30 m A with a scanning speed of 5°/min at 2θ steps of 0.02°. The crystallite size was observed by transmission electron microscope (TEM; JEM–2010, JEOL, Germany). Hardness and density of samples were measured by the Micro Hardness Tester (HMV–2T SHIMADZU, Japan) and the Density Kit (MS–DNY–54, Mettler Toledo, USA) Seebeck coefficient, electrical resistivity and thermal conductivity were measured by steady state method at temperature ranges of 323–473 K.

Results and Discussion

The X–ray diffraction pattern of sintered $Sr_{1-x}La_xTiO_3$ matched with ICDD PDF card number 00–035–0734 ($SrTiO_3$) indicating the single phase for cubic structure as shown in Figure 1 The lattice parameter values of x = 0, 0.08 and 0.13 are 3.905, 3.906, and 3.902, respectively. The enlarge (110) pattern showed in Figure 2 and FWHM decreased with x value that indicate the crystal size grow up. The Figure 3(a-c) show the transmission electron microscope (TEM) images of $SrTiO_3$, $Sr_{0.92}La_{0.08}TiO_3$ and $Sr_{0.87}La_{0.13}TiO_3$, respectively. The average crystal size of the sintered samples found to be 0.18, 0.24 µm and 0.35 µm for 0, 0.08 and 0.13 of x value, respectively that agree with result from

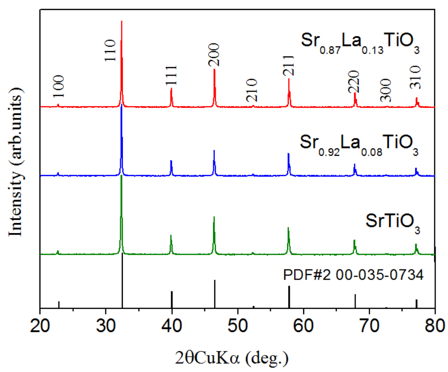

Figure 1. The X–ray diffraction pattern of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

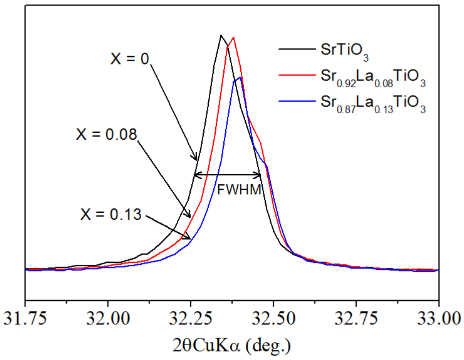

Figure 2. The enlarged for (110) peaks of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

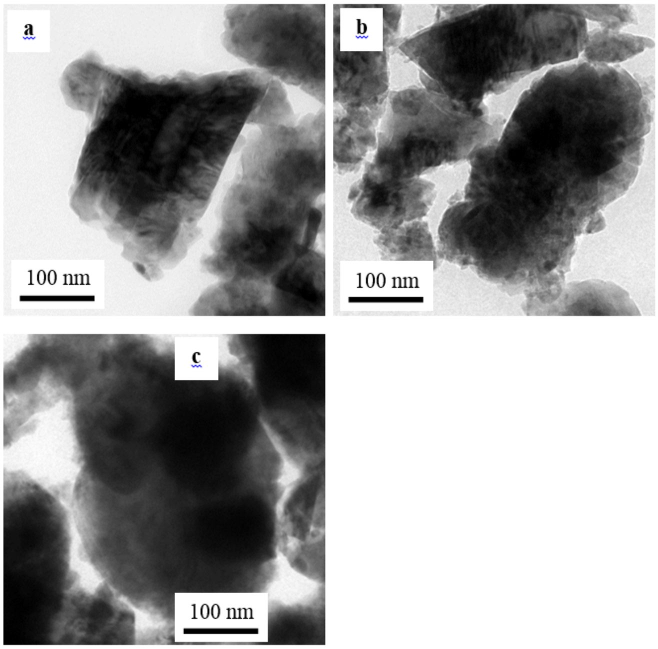

Figure 3. The TEM images of (a) $SrTiO_3$, (b) $Sr_{0.92}La_{0.08}TiO_3$ and (c) $Sr_{0.87}La_{0.13}TiO_3$

calculation by Scherrer equation in order about 0.28, 0.36, and 0.51 µm. The density and hardness of x = 0, 0.08 and 0.13 show in the Table 1. All samples have relative density up to 95% and density increase with x value which affect to the electrical resistivity decrease. All the samples show high hardness closer to the quartz. Seebeck coefficient, electrical resistivity, thermal conductivity and dimensionless figure of merit of samples compare with literature data (Shang *et al.*, 2010; Wang *et al.*, 2010) show in Figures 4, 5, 6, and 7, respectively. The Seebeck coefficient of all samples show negative, indicating

Table 1. Density and hardness of sintered samples

Materials	Density ($g\ cm^{-2}$)	Relative density (%)	Hardness ($N\ m^{-2}$)
$SrTiO_3$	4.85 ± 0.11	95.02	490.21
$Sr_{0.92}La_{0.08}TiO_3$	5.06 ± 0.04	96.74	534.33
$Sr_{0.87}La_{0.13}TiO_3$	5.09 ± 0.19	95.85	543.66

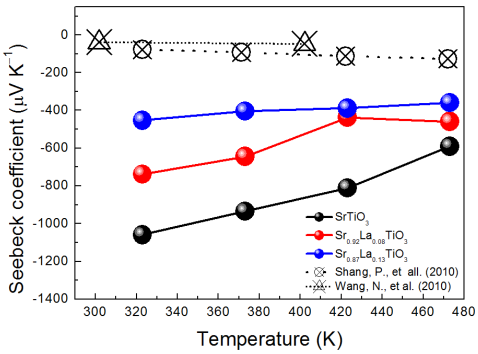

Figure 4. Temperature dependence of Seebeck coefficient of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

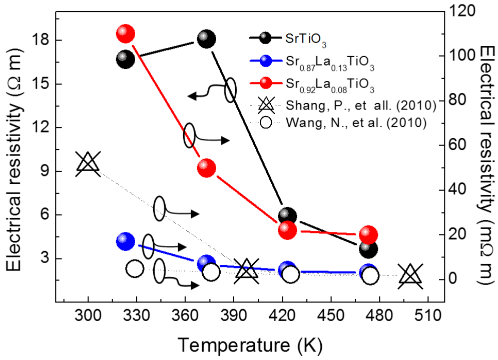

Figure 5. Temperature dependence of electrical resistivity of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

n–type TE material and show higher than literature data, due to larger the carrier concentration. The electrical resistivity of almost samples increased while temperature increasing. The La substitute has affected to electrical resistivity decrease. The thermal conductivity of all samples higher than single crystal of $SrTiO_3$ (Wang, 2011) and depend on temperature according to the behavior of the phonon. The $Sr_{0.87}La_{0.13}TiO_3$ shows highest ZT value of 1.13×10^{-3} at 473 K.

Conclusions

In this work was substituted La in Sr site of $SrTiO_3$ for enhancement thermoelectric properties. The crystal size of the samples increased with x value which has affect to good electrical property. The substitution showed high Seebeck coefficient and reduced electrical resistivity. The reduction of thermal conductivity was found in only $Sr_{0.92}La_{0.08}TiO_3$. The sample for x = 0.13

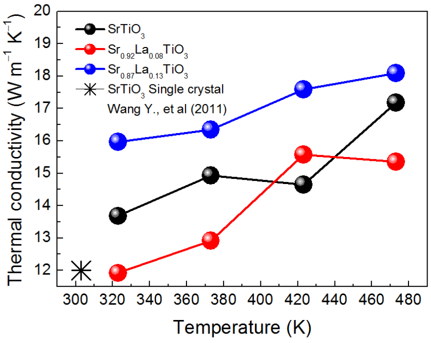

Figure 6. Temperature dependence of thermal conductivity of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

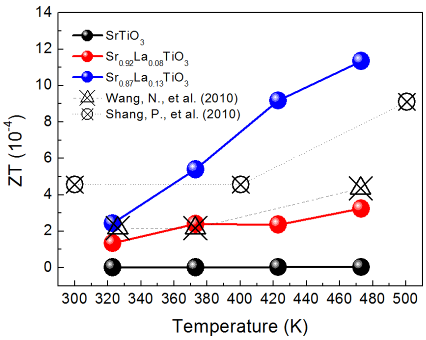

Figure 7. Temperature dependence of dimensionless figure of merit of $Sr_{1-x}La_xTiO_3$ (x = 0, 0.08, 0.13)

showed the maximum ZT of 1.13×10^{-3} at 473 K. This is enhancement thermoelectric property of $SrTiO_3$.

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