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Thermoelectric Properties and Power Generation of p–$Ca_3Co_4O_9$ and n–$Sr_{0.87}La_{0.13}TiO_3$ Thermoelectric Modules

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Abstract. The $Ca_3Co_4O_9$ (CCO) and $Sr_{0.87}La_{0.13}TiO_3$ (SLTO) are good property of oxide thermoelectric (TE) materials. They synthesized by solid state reaction (SSR) method to study thermoelectric properties and fabrication of thermoelectric module. It was found that, synthesis of CCO shows that Seebeck coefficient, electrical resistivity, thermal conductivity and ZT values are 130 μV K^{-1}, 8.31 mΩ cm, 0.82 W m^{-1} K^{-1} and 0.08, respectively at 473 K. The Seebeck coefficient, electrical resistivity, thermal conductivity and ZT values of SLTO are –359 μV K^{-1}, 2.9 mΩ m, 18.09 W m^{-1} K^{-1} and 1.13×10^{-3}, respectively at473 K. TE modules of CCO and SLTO were fabricated by ultra sonic soldering method. The power generation of TE modules were measured with temperature difference (ΔT) of 10–180 K. The 1 pair and 2 pairs TE modules for a maximum power generation of matching load are 19 kΩ and 30 kΩ, respectively. The maximum output power of 2 pairs TE module is larger than 1 pair TE module about two times.

Introduction

Thermoelectric materials can convert thermal to electricity and vice versa which no moving part, no CO_2 and CO. Oxide TE materials have been high performance at high temperature and non toxic. The efficiency of TE materials was confirmed by ZT value ($ZT = S^2T/\rho\kappa$; S is Seebeck coefficient, T is temperature, ρ is electrical resistivity and κ is thermal conductivity). However, power generation of thermoelectric module was also important and has been interested in the present [1]. In this work, prepared CCO and SLTO by SSR method to study TE properties, fabricated TE module for power generation measurement.

Methodology

The CCO and SLTO were prepared by solid state reaction method. Preparation of CCO was started form $CaCO_3$ (99% purity, Quality Reagent Chemical Co. Ltd, New Zealand) and Co_3O_4 (<10 μm, Sigma Aldrich, St. Louis, MO, USA) mixed by ball–mill at atmosphere for 2 h. The mixed powders were calcined at 1073 K for 10 h. After calcine, The powders were pressed into pellet by hydrolic single axis at 686 MPa holding for 5 min. The pellet was sintered in atmosphere at 1123 K for 12 h. The SLTO was synthesised form $SrCO_3$ (98%, Sigma–Aldrich, St. Louis, MO, USA), La_2O_3 (99.90%, Sigma–Aldrich, St. Louis, MO, USA) and TiO_2 (99%, Ajax Finechem Pty. Ltd, New Zealand) powders which 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 in atmosphere at 1773 K for 12 h. The CCO and SLTO pellet were cut into dimension $2.1\times2.1\times10\ mm^3$ by low speed saw (ISO Met Low Speed Saw; Buehler Co. Ltd, USA).

Thermoelectric module fabrication was using ultrasonic soldering method (USS-9210 Ultrasonic Soldering System, Switzerland). Commercial solder alloys (Alloy CS#297) was soldered on top and bottom of CCO and SLTO materials to connect with Cu electrode. The ceramic substrate was used with size of $25\times25\times1\ mm^3$ to absorb and reject heat of thermoelectric module. The measurement power was generated by thermoelectric module.

The crystal structure and crystallite size were characterized by X–ray diffraction (XRD; Shimadzu 6100, Japan) method using CuKα radiation at 40 kV, 30 mA with a scanning speed of 5°/min at 2θ steps of 0.02°. The morphology was observed by scanning electron microscope (SEM; JEOL JSM–5401, Germany). Seebeck coefficient, electrical resistivity and thermal conductivity were measured by steady state method at temperature ranges of 323–473 K. Power generation of thermoelectric module was tested by TE performance system in differential temperature range of 0–200 K (control cold side for < 323 K).

Results and Discussion

The X–ray diffraction patterns at room temperature of CCO are shown in Fig. 1 (a). The diffraction peaks were indexed by comparing with ICDD PDF card number 00–021–0139 and consistent as well. The XRD result of CCO was indicated monoclinic structure. Figure 1 (b) shown diffraction peaks of SLTO with ICDD PDF card number 00–035–0734. The diffraction peaks shows single phase and cubic perovskite structure.

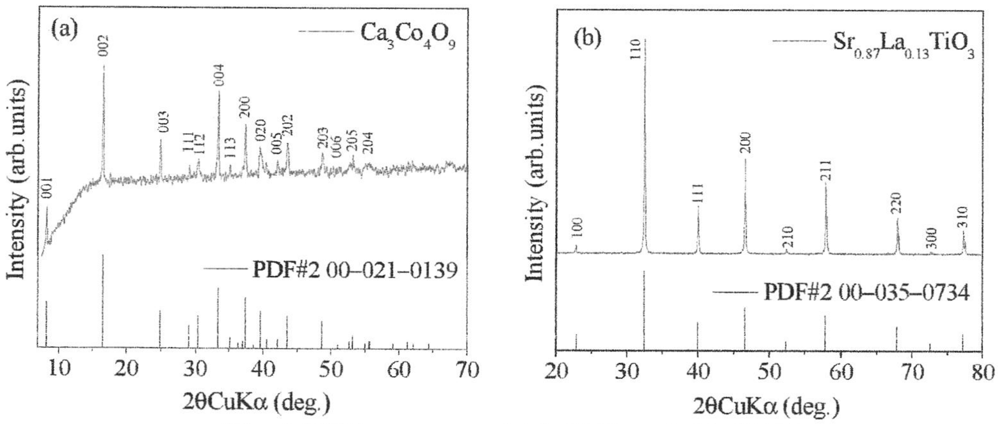

Fig. 1 XRD patterns of (a) CCO and (b) SLTO

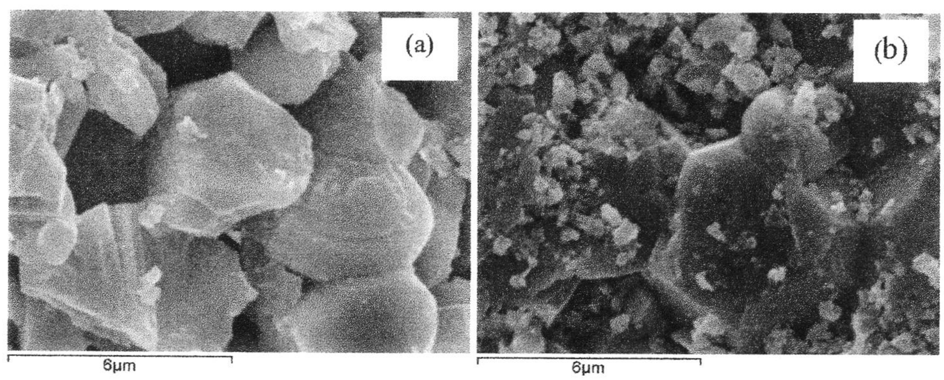

Fig. 2 SEM images of (a) CCO and (b) SLTO

The microstructure of CCO and SLTO are showed in Fig. 2 (a) and (b). The SEM image of CCO show invariably grains boundaries connection and affects to low electrical resistivity. The different crystal sizes were observed in SLTO which obtain high electrical resistivity. The average grain size of the sintered sample was found to be 4.05 μm and 5.50 μm for CCO and SLTO, respectively.

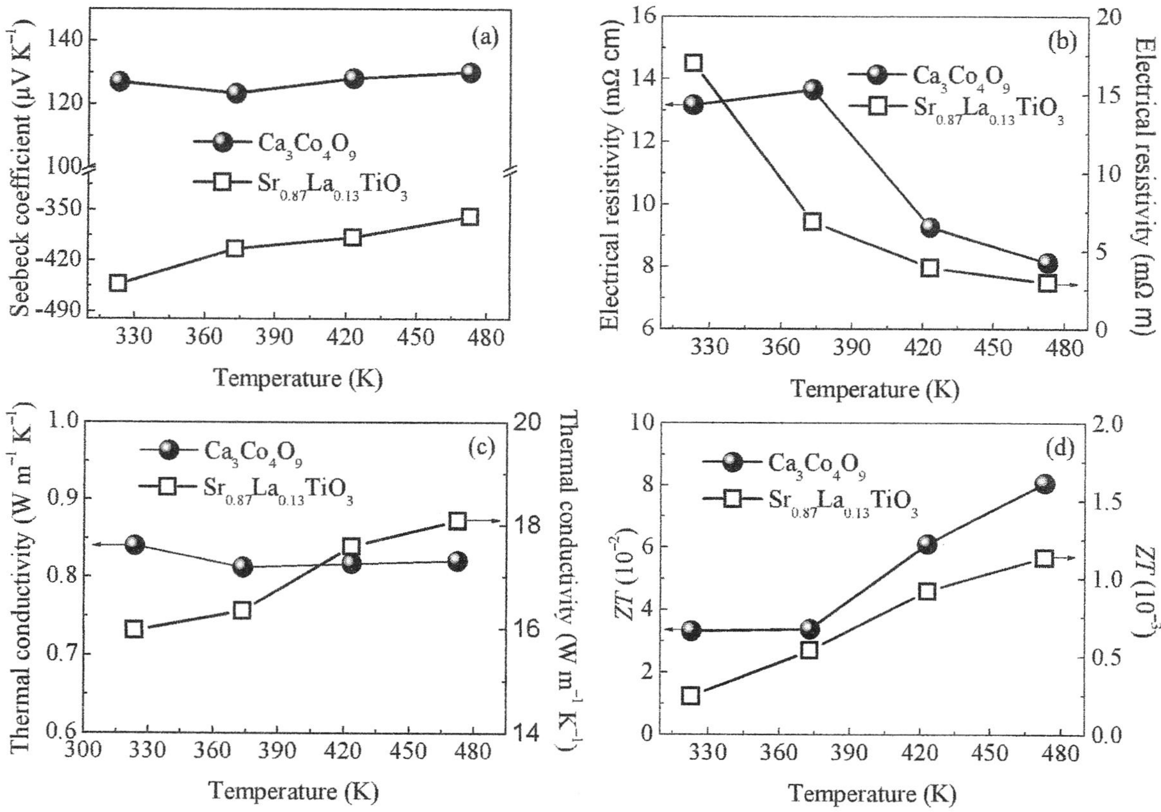

Fig. 3 (a) Seebeck coefficient (b) Electrical resistivity (c) Thermal conductivity and (d) Dimension less figure of merit depend on temperature of *p*–type $Ca_3Co_4O_9$ and *n*–type $Sr_{0.87}La_{0.13}TiO_3$

Thermoelectric properties of CCO and SLTO are showed in Fig. 3. The Seebeck coefficient of CCO and SLTO were obtianed positive and negative, indicating *p*–type and *n*–type TE materials, respectively. Signify the *p*–CCO and the *n*–SLTO have large holes carrier density and large electron carrier density, respectively. The electrical resistivity of CCO and SLTO at low temperature range of 323–473 K was decreased with increasing temperature, which shows semiconducting–like behavior [2–4]. The electrical resistivity value of SLTO was obtained larger than CCO of 100 times which could be explained by the carrier concentration is lower than [5–7]. Thermal conductivity of CCO was small change, while SLTO increases with temperature increasing. Indicating that phonon–phonon of SLTO scattering is dependent on temperature. The highest *ZT* value of CCO and SLTO are 8.07×10^{-2} and 0.11×10^{-2}, respectively. Now, we know the *p*–type $Ca_3Co_4O_9$ and *n*–type $Sr_{0.87}La_{0.13}TiO_3$ TE properties and then we interest fabrication of TE module using both TE materials to generate electric power.

The TE module was fabricated 1 and 2 pairs of *p* and *n* legs to generate voltage and current into power. The open circuit voltage of 1 and 2 pairs thermoelectric module were compared as a function of temperature and shows the maximum value at ΔT=180 K of 186.9 mV and 398 mV, respectively. The voltage and output power depen on current of 1 and 2 pairs thermoelectric module as shown in Fig. 4 (b). The maximum power of 1 and 2 pair TE module was observed at R_{load}= 19 kΩ and 30 kΩ about 0.45 μW and 0.227 μW, respectively. The output power of 1 and 2 pairs thermoelectric modules show increases exponential value and a maximum value at ΔT =180 K of 0.67 and 1.54 μW, respectively as shown in Fig. 4 (c).

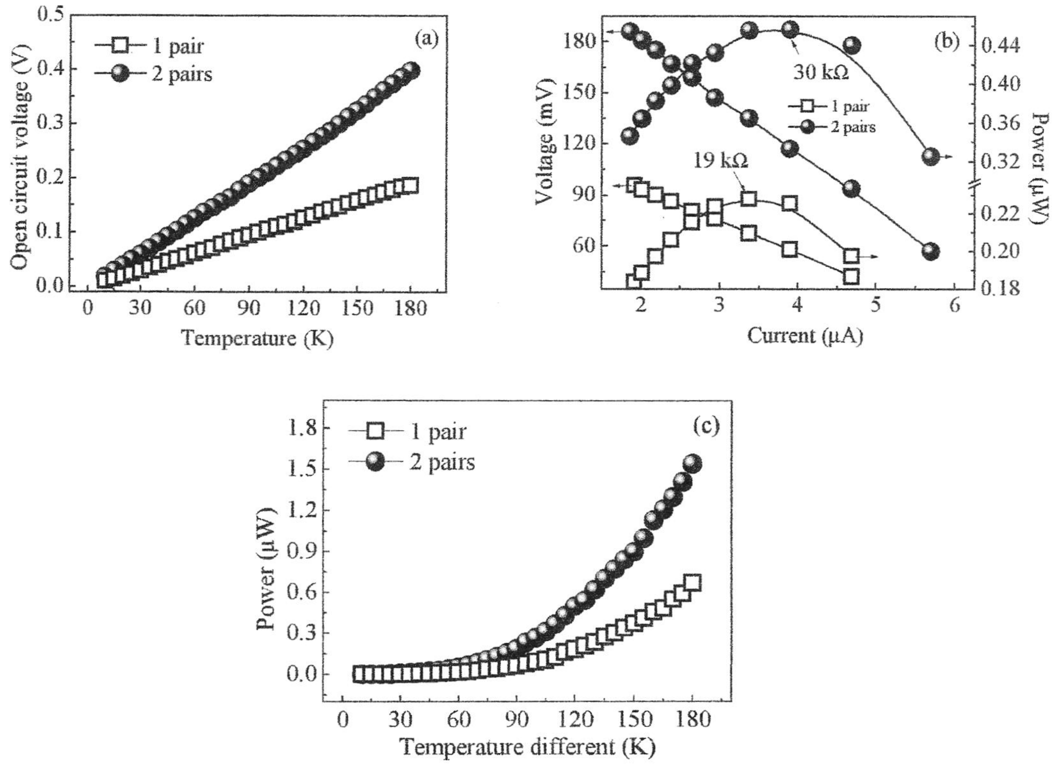

Fig. 4 (a) The open circuit voltage depend on temperature (b) The voltage and output power depend on current (c) The output power depend on temperature difference

Conclusion

Thermoelectric modules were fabricated form *p*–type $Ca_3Co_4O_9$ and *n*–type $Sr_{0.87}La_{0.13}TiO_3$ by ultrasonic soldering. We can be generated power depend on temperature difference and number of couple pairs. The ultrasonic soldering method was suggested for good connecting TE materials with electrode and best conduction.

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Mechanical Engineering				[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	Q4	Q4	Q4	[illegible]
Mechanics of Materials	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	Q4	Q4	Q4	Q4	Q4

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SJR	0,257	0,279	0,251	0,200	0,205	0,205	0,212	0,216	0,184	0,184	0,201	0,173	0,169	0,167	0,188	0,207
Total Documents	501	786	1.130	891	890	2.082	2.003	2.713	3.959	2.130	1.462	2.334	3.606	3.173	2.380	3.945
Total Docs. (3years)	1.252	1.587	1.628	2.417	2.807	2.911	3.863	4.975	6.798	8.675	8.802	7.551	5.926	7.402	9.113	9.159
Total References	6.407	8.171	9.666	8.828	8.639	17.430	18.841	21.308	32.673	19.997	14.461	24.218	33.185	32.130	27.531	40.734
Total Cites (3years)	266	354	391	456	544	773	931	1.339	1.577	1.918	1.860	1.786	1.461	1.582	1.954	2.037
Self Cites (3years)	11	17	12	37	65	153	192	257	372	221	164	140	127	160	207	311
Citable Docs. (3years)	1.243	1.580	1.620	2.403	2.782	2.881	3.827	4.940	6.757	8.624	8.749	7.508	5.863	7.299	8.962	8.996
Cites / Doc. (4years)	0,21	0,24	0,25	0,21	0,21	0,25	0,24	0,26	0,24	0,23	0,21	0,23	0,23	0,22	0,22	0,21
Cites / Doc. (3years)	0,21	0,22	0,24	0,19	0,20	0,27	0,24	0,27	0,23	0,22	0,21	0,24	0,25	0,22	0,22	0,23
Cites / Doc. (2years)	0,20	0,23	0,22	0,17	0,20	0,25	0,24	0,26	0,22	0,22	0,20	0,26	0,26	0,20	0,22	0,26
References / Doc.	12,79	10,40	8,55	9,91	9,71	8,37	9,41	7,85	8,25	9,39	9,89	10,38	9,20	10,13	11,57	10,33
Cited Docs.	197	244	265	338	410	559	667	922	1.122	1.393	1.350	1.230	1.017	1.106	1.388	1.401
Uncited Docs.	1.055	1.343	1.363	2.079	2.397	2.352	3.196	4.053	5.676	7.282	7.452	6.321	4.909	6.296	7.725	7.758
% International Collaboration	10,38	9,92	6,19	2,36	11,46	14,65	12,58	11,76	10,76	9,86	9,71	9,17	6,82	8,13	10,59	8,90