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**Electronic Supplementary Information**

# Inducing oxide ion vacancies in $Sr_2CoSbO_6$ as a strategy to decouple power factor and thermal conductivity

Manickam Ramesh and Krishnendu Biswas*

Chemistry Division, School of Advanced Sciences, Vellore Institute of Technology, Chennai – 600127, India.

*e-mail: krishnendu.biswas@vit.ac.in

## Experimental details

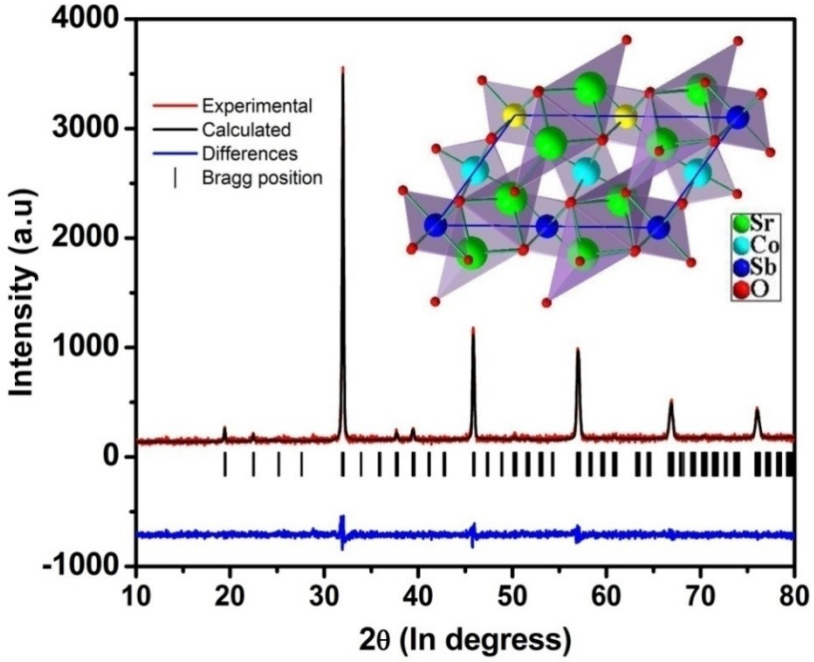
### Materials and methods

The synthesis of all the compounds was carried out using the reported literature method [18]. Strontium Carbonate ($SrCO_3$, LOBA Chemie., 98.0% purity), Cobalt Oxide ($Co_3O_4$., SRL, 99.0% purity), Antimony Trioxide ($Sb_2O_3$., LOBA Chemie, 98.5% purity), and Titanium dioxide ($TiO_2$, LOBA Chemie, 99.5% purity) precursors were used as received for the synthesis. These precursor materials were weighed according to the stoichiometry of $Sr_2CoSb_{1-x}Ti_xO_6$ (x = 0, 0.1, 0.3, and 0.5) and ground well for 30 minutes. The powders were then transferred into alumina crucibles and heated at 400° C for 12 hours, 700° C for 12 hours and 1150° C for 24 hours with intermittent grinding. To avoid the volatilization of $Sb_2O_3$, slow heating was carried out for allowing considerable time for diffusion. The synthesized powder was then densified by making round pellets by a uniaxial hydraulic press under a pressure of 10 tons and then heating at 1150° C for 24hours. The coin-shaped samples having a density of 75 - 79 % were used for thermal conductivity and Hall Effect measurements. The same samples were then polished into rectangular bars with a dimension of 12 x 4 x 1.5 mm and used for Seebeck coefficient and electrical conductivity measurements.

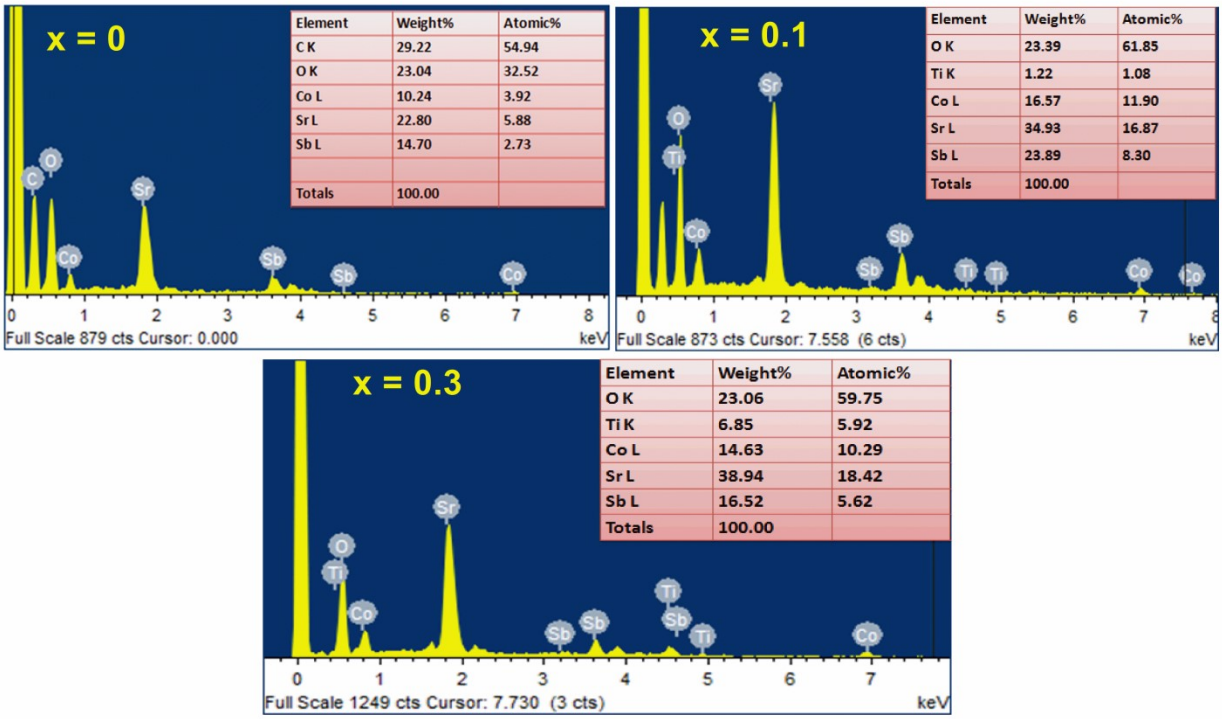
### Characterization techniques

The phase purity and crystal structure of all the powders were recorded by X-ray powder diffraction (XRD) using Rigaku SmartLab Ultima IV diffractometer with Cu $K_\alpha$ ($\lambda$ = 1.5406 Å) radiation in the range of 20-80° at a scan rate of 5°/min. The SEM and EDAX were carried out with JDX-3350, JEOL, Japan for morphology and elemental analysis. X-ray photoelectron spectroscopy (XPS) was carried out using ULVAC-PHI (PHI5000 Version Pro III) to confirm the oxidation states of the elements. The oxygen vacancies were confirmed by TGA analysis using TGA-DTA (Seiko Thermo Analyzer) scanning from room temperature to 800° C at a heating rate of 10° C/min. The graphite spray-coated pellets were subjected to thermal diffusivity and specific heat capacity using NETZSCH LFA 467 HT Hyper flash instrument, Germany, with continuous Ar flow. The total thermal conductivity was calculated by $\kappa_{total} = DC_p\rho$, where D is the density, $C_p$ is the specific heat capacity and $\rho$ is the thermal diffusivity of the materials. The room temperature hall measurements for all the pellets were determined simultaneously by the four-probe method done with ECOPIA (AHTSSYS) HMS – 7000. The temperature dependent thermoelectric properties like electrical resistivity ($\rho$), Seebeck coefficient (S), and power factor ($S^2\sigma$) were simultaneously measured using the ZEM-3 ULVAC-RIKO instrument with He atmosphere.

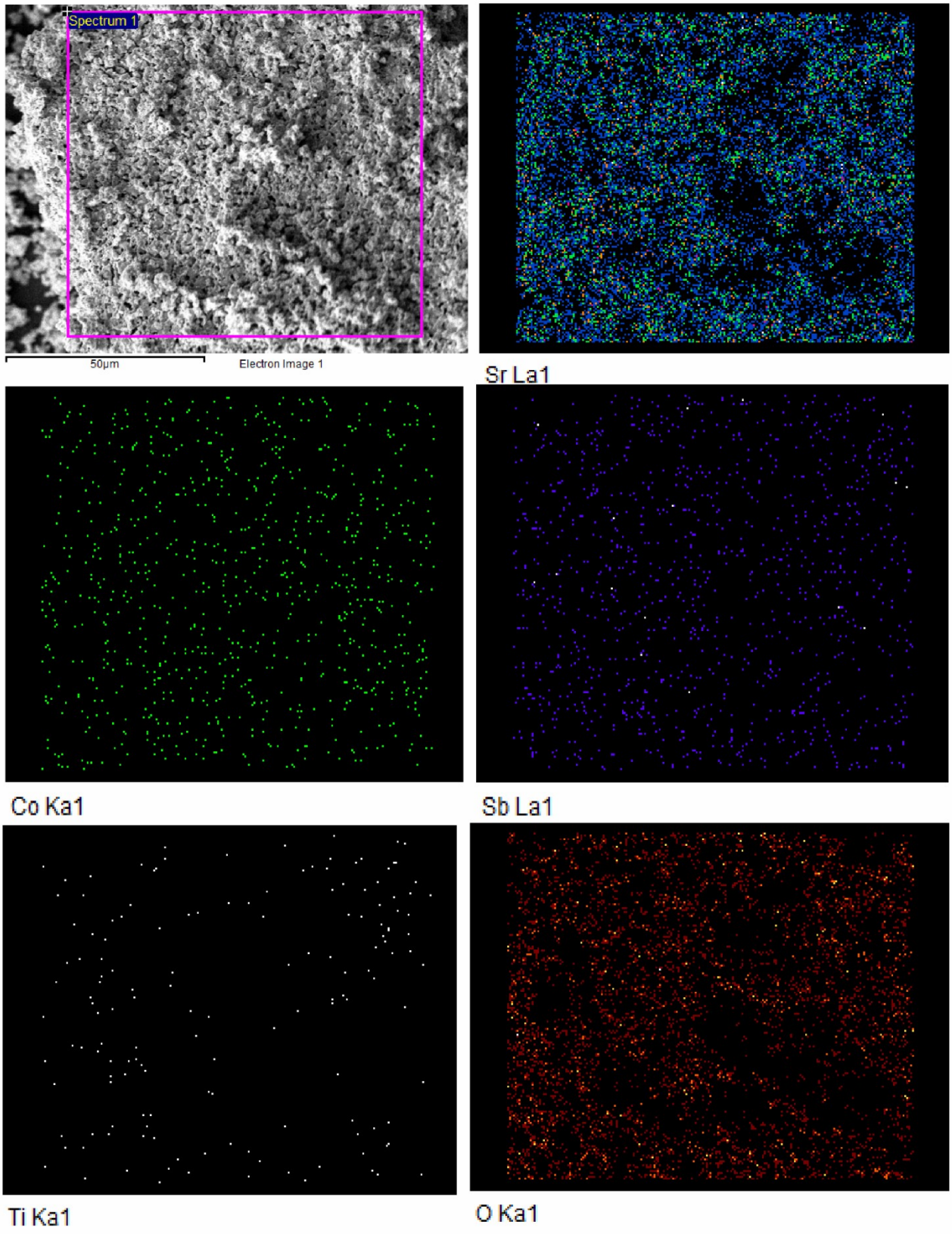
## Figures




ESI-S1 Rietveld analysis result and crystal structure of $Sr_2CoSbO_6$




ESI-S2 EDAX results of $Sr_2CoSb_{1-x}Ti_xO_6$ ($0 \le x \le 0.3$).




ESI-S3 Elemental mapping results (a) Electron image, (b) Sr, (c) Co, (d) Sb, (e) Ti and (f) O of $Sr_2CoSb_{1-x}Ti_xO_6$ (x = 0.3).

**Iodometric titration method:**

The iodometric titration was carried out to determine the oxygen content of all the compositions of synthesized $Sr_2CoSb_{1-x}Ti_xO_{6-\delta}$ ($0 \leq x \leq 0.3$) powders by the following procedure. The 50 mg of powder was weighed and dissolved in 100 ml of 2.5 M HCl solution containing an excess of KI (2 g) powder. The $I_2$ liberated in the chemical reaction was then titrated with a standard thiosulfate $Na_2S_2O_3$ solution (0.01 M) using ~3 mL of starch solution as an indicator. The end-point was detected visually as the blue colour of the starch complex disappeared and the solution turned colourless due to the formation of CuI. The concentration of the 0.01 M $Na_2S_2O_3$ solution was standardized against the 0.01 M $K_2Cr_2O_7$ solution [1]. Thus, the oxygen content in the samples was examined based on the valance of cobalt could be calculated based on the amount of $S_2O_3^{2-}$ consumed and the amount of oxide powder applied. The iodometric titration results of the $Sr_2CoSb_{1-x}Ti_xO_{6-\delta}$ ($0 \leq x \leq 0.3$) oxides at room temperature, which were following the decreasing trend with increasing x = 0 to x = 0.3 concentrations as listed in the Table. 1.

| S. No. | DPs samples | Oxygen content |
|---|---|---|
| 1 | $Sr_2CoSbO_{6-\delta}$ | 5.7990 |
| 2 | $Sr_2CoSb_{0.9}Ti_{0.1}O_{6-\delta}$ | 5.8021 |
| 3 | $Sr_2CoSb_{0.7}Ti_{0.3}O_{6-\delta}$ | 5.8053 |

**Reference**

[1] Conder, K., E. Pomjakushina, A. Soldatov, and E. Mitberg. Mater. Res. Bull. 2 (2005) 257-263.