Co-sintering of an all-oxide thermoelectric stacking device by SPS

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Highlights
- All-oxide TE-devices are stable at high temperatures in air
- SPS enable co-sintering of oxides with different sintering properties
- A co-sintered single TE-pair produced 23 mW cm⁻²

1. Introduction
Due to high stability at elevated temperatures in air combined with environmental friendliness and low cost, oxides are potential candidates as components in thermoelectric (TE) devices for conversion of waste heat into electricity at high temperatures. Beside the advantages, oxides have challenges due to efficiency and processing. The efficiency of a single TE material is described by $zT$ or the-figure-of-merit, which includes Seebeck coefficient, electrical and thermal conductivity. An all-oxide thermoelectric device (Fig. 1) is composed of a $n$- and a $p$-type conductor electrically connected at the high temperature end and separated by an electrical insulator. Development of this novel device design requires improved efficiency of the oxide materials through designed microstructure followed by an advanced ceramics processing approach.

2. Methods
For our all-oxide device, we have selected Ca$_{0.932}$MnO$_3$ (CMO), Ca$_3$Co$_4$O$_9$ (CCO) and LaAlO$_3$ (LAO) as $n$-type, $p$-type and insulator materials respectively, based on $zT$ and coefficients of thermal expansions. Each of these three oxides were synthetized by spray pyrolysis to obtain fine nano-sized powder precursor with good sinterability. The insulating material (LAO) were tape casted and embedded in a SPS-graphite crucible with CMO and CCO powders on both sides. The whole assembly was co-sintered by SPS for 5 minutes at 880 °C and 75 MPa pressure. Power output and efficiency of the device were recorded from 700 to 800 °C in air.

3. Results and discussion
After co-sintering SEM-analysis revealed a thin and crack free reaction layer at the p-n junction ensuring good contact between the active components. The device performed satisfactory during testing of electrical parameters and efficiency and a maximum power output of 23 mW cm⁻² was recorded. CMO, CCO and LAO represent oxide compositions with large variations in sintering properties and special measures are necessary to successfully co-sinter all three components. The presentation will further discuss in more detail the challenges related to using SPS as tool for producing small all-oxide TE-devices.

4. Conclusions
A single pair all-oxide TE-device was successfully fabricated by co-sintering using SPS. The device was fully functional and produced a maximum power output of 23 mW cm⁻².
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References
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Keywords
Processing, oxides, thermoelectric device; co-sintering