Structural And Property Studies of The Thermoelectric Thin Film Materials $Bi_{1-x}Te_x/Sb_{1-y}Te_y$

Christian-Albrechts-University of Kiel

Institute of Inorganic Chemistry

X. Liu, W. Bensch

Introduction

Due to the potential in improving the sustainability of electric energy through recycling waste heat, thermoelectric materials have drawn remarkable interest from solid state scientists. Conventionally thermoelectric applications were developed to utilize this phenomenon for commercial refrigeration and power generation from homogeneous bulk materials [1]. Numerous attempts have welled up [2] in order to reproduce the pioneering high figure of merit ZT obtained by Venkatasubramanian *et*. *al*. in 2001 in Bi_2Te_3 and Sb_2Te_3 materials [3]. In the current work, we report the studies of Bi:Te, Sb:Te and Bi:Sb:Te samples prepared using a molecular beam epitaxy (MBE) system that are off-stoichiometric from 2:3, in order to examine the influence of non-stoichiometry on the thermoelectric properties of thin film samples.

Synthesis

Table 1 Bi : Te (BT), Sb : Te (ST) and Bi : Sb : Te (BST) thin film samples prepared via multi-layer approach (light blue boxes) and co-deposition approach (darker blue boxes).

Sample ID	Te %	PF (μW/ (K²cm))	
		As-deposited	Annealed
BT - 1	19.8	4.2	-
BT - 2	21.6	1.0	1.2
BT - 3	22.6	-	0.5
BT - 4	56.1	0.7	2.4
BT - 5	57.9	1.1	-
BT - 6	58.4	1.2	3.5
BT - 7	62.4	1.1	4.4
BT - 8	68.6	1.1	5.3
BT - 9	68.7	1.2	2.6

	Bi : Sb : Te	PF	
		As-deposited	Annealed
BST	5.0 : 29.5 : 65.5	2.6	24.9

Sample ID	Te %	PF (μW/ (K²cm))	
		As-deposited	Annealed
ST 1	39.6	0.6	-
ST 2	40.5	0.7	3.5
ST 3	40.8	2.2	-
ST 4	41.3	2.0	-
ST 5	41.8	-	20.0
ST 6	42.3	0.5	3.3
ST 7	43.7	4.7	11.6
ST 8	44.2	0.5	6.5
ST 9	45.1	7.7	-
ST 10	50.8	5.8	7.9
ST 11	60.4	4.4	25.1
ST 12	61.4	4.5	20.8

In-situ XRD characterisation

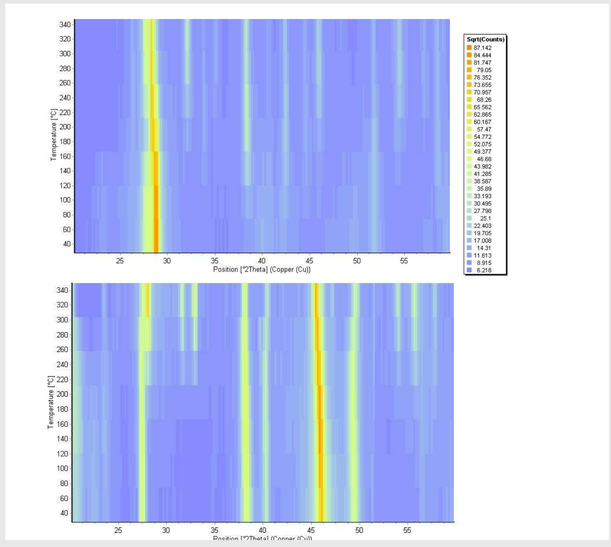

Fig. 1 In-situ XRD patterns on as-deposited samples of ST-5 (top), and BT-3 (bottom)

Physical properties and structural analysis

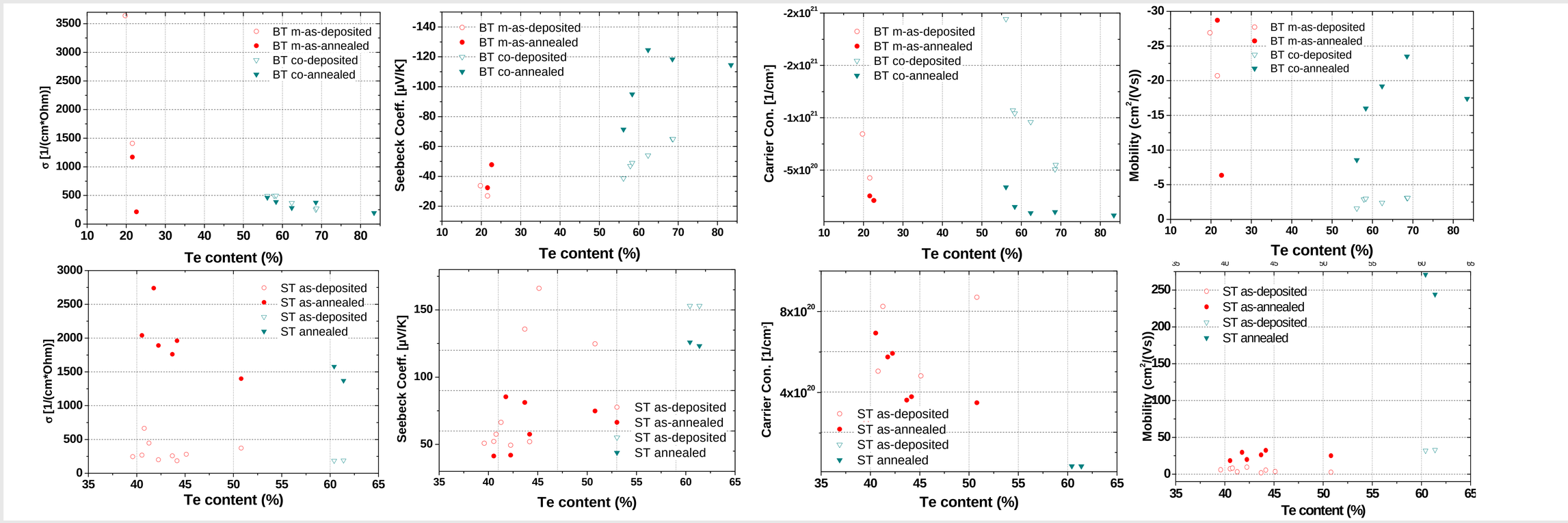

Fig. 2 Plots of conductivities, Seebeck coefficients, carrier concentrations and cobilities (from left to right) as function of Te content for ST (top) and BT samples (bottom).

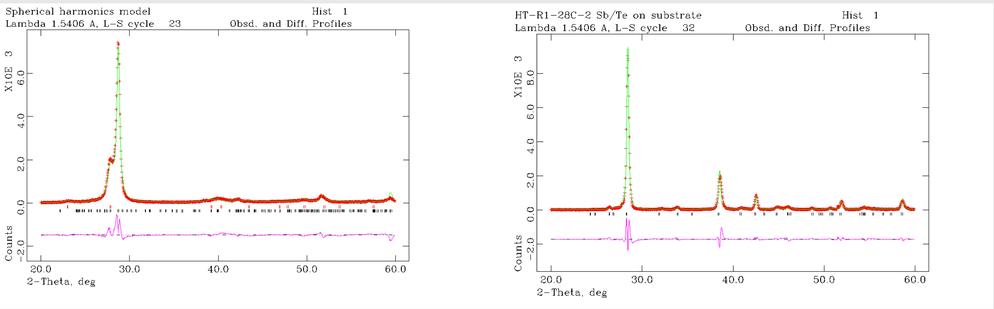

Fig. 3 Fitted XRD patterns for ST-5 as deposited (left) and annealed (right)

Table 2 Refined parameters for ST-5 as deposited (left) and annealed (right)

Phase	I ($Sb_{16}Te_3$)	II (Te)
Crystal system	Rhombohedral	Hexagonal
Space group	*R* -3 *m*	*P*31 2 1
Unit cell parameters	*a* = *b* = 4.2817(-) Å and *c* = 108.7017(-) Å	*a* = *b* = 4.468 (3) Å and *c* = 5.712 (8) Å
Z	3	3
Density	6.734 g cm^{-3}	6.429 g cm^{-3}
Phase fraction	0.612 (3)	0.388 (12)
Texture index	2.0212	2.1654
R_{wp} = 0.2186, R_p = 0.1597 and χ^2 = 11.72 for 18 variables		

Phase	Sb_2Te_3
Crystal system	Rhombohedral
Space group	*R* -3 *m*
Unit cell parameters	*a* = *b* = 4.2602 (8) Å and *c* = 30.330 (6) Å
Z	3
Density	6.544 g cm^{-3}
Texture index	1.1711
R_{wp} = 0.2658, R_p = 0.1968 and χ^2 = 13.24 for 17 variables	

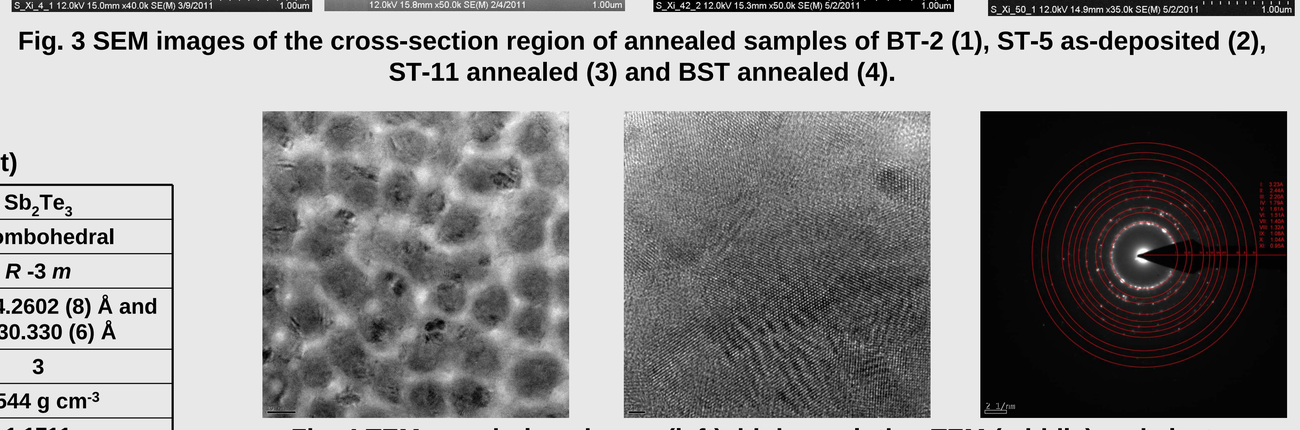

Fig. 3 SEM images of the cross-section region of annealed samples of BT-2 (1), ST-5 as-deposited (2), ST-11 annealed (3) and BST annealed (4).

Fig. 4 TEM morphology image (left), high resolution TEM (middle) and electron diffraction pattern of ST-11 (right).

Acknowledgement:

Thanks to the collaborating partners in the project: M. Winkler, J. D. König, Dr. H Böttner (Freiburg) and Dr. U. Schürmann, Prof. L. Kienle (Kiel). Many thanks to Deutsche Forschungsgemeinschaft (DFG) for financing the program SPP 1386 (Nanothermoelectrics).

Contact:

Prof. Dr. Wolfgang Bensch
Institute of Inorganic Chemistry, Max-Eyth-Str. 2, 24118 Kiel
Tel.: +49 (0)431 880-2419
wbensch@ac.uni-kiel.de

Reference:

[1] P. Vaqueiro, A.V. Powell, J. Mater. Chem., **20**, (2010), 9577.
[2] M. Winkler, Jan D. König, S. Buller, U. Schürmann, L. Kienle, W. Bensch, H. Böttner, Proc. 8th Europ. Conf. Thermoelectrics, Italy, (2010).
[3] R. Venkatasubramanian, E. Siivola, T. Colpitts, B. O'Quinn, Nature, **413**, (2001), 597.