

Synthesis of Bi_2Te_3 and $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$ by Spark Plasma Sintering for Thermoelectric Material Applications

Meeting at UC Davis, July 29, 2008

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Activity one: Meeting at UCD for project planning, 6/16/08

Minutes of TE Project (6/20/08): Yang/ Zhehui/Zhou Meeting Objective:

- (1) To determine material science subjects and scientific information that are relevant to the SNL technical objective and therefore, make modification of milestone accordingly
- (2) Role and responsibility between UC Davis and SNL, CA.

Following were the subjects of interest and meeting discussions:

- (1) Study of thermal evolution event (DSC, DTA or SPS) UCD/SNL
Ambient to melting temperature Room temperature—melting point scan to characterize recover, recrystallization and grain grow of the Bi-Te thermal legs.
- (2) Effect of Process induced Porosity on the electrical conductivity, thermal conductivity and/or Seebeck coefficient and mechanical properties.-SNL/UCD
 - Thermal conductivity κ
 - Electrical conductivity σ
 - Others, such as power factor $\alpha^2\sigma T$ and Figure of Merit $ZT = \frac{\alpha^2\sigma T}{(\kappa_e + \kappa_{ph})}$
- (3) Chemical Effect by adding Sb into the Bi₂Te₃ parent compound similar to the material used in the commercial thermal legs. -(Bi₈Sb₃₂)Te₆₀
 - Comparison of pure Bi₂Te₃ v.s. (Bi, Sb)2 Te₃
- (4) Thermal stability and material compatibility with material used for interconnect assemble, solder in particular. (SNL/UCD)
- (5) Experimental parametrics

FY09, selecting a single powder size (micrometer regime, to be determined according to sifting yield)

Compound formula	Bi ₂ Te ₃	(Bi,Sb) ₂ Te ₃ Bi ₂ Te ₃ +Sb ₂ Te ₃ , Composition ratio mix to be determined by Sandia
Synthesis by SPS (UCD)	DSC (UCD) Porosity change (UCD) by varying P and T ↓ TE properties (κ , σ , etc.) (Sandia)	DSC (UCD) Porosity change (UCD) ↓ TE properties (κ , σ , etc.) (Sandia)
Microstructure (SNL)	Grain size Conventional SEM, TEM (SNL/UCD) EBSD (Sandia) HRTEM (depending É)	Grain size Conventional SEM, TEM (SNL/UCD) EBSD (Sandia) HRTEM (depending É)
Optimization	Porosity Grain size	Porosity Grain size
	6-12 disks (UCD), 1.3-1.5 mm thick	6-12 disks (UCD)
Soldering	Solder (Sandia) Thermal stability(SNL)	Solder (Sandia) Thermal stability(SNL)

Completed



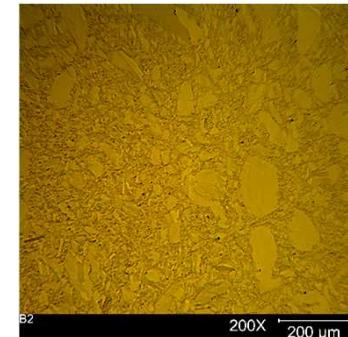
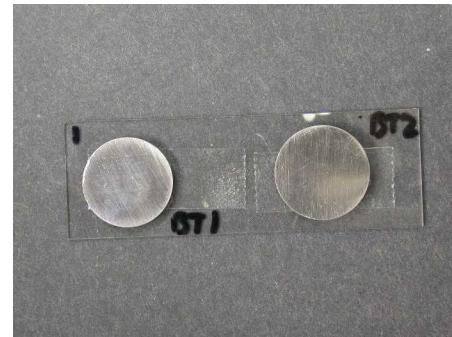
SPS specimens from UCD

1st shipment

BT1-440°C-80Mpa-1/2"x2mm thick?

BT2-440°C-50Mpa-1/2"x2mm thick?

100% dense



2nd shipment

BT3-400°C-50MPA-

BT4-400°C-80MPA

Porosity??



SNL cold pressed/sintered disks of $(\text{Bi0.2Sb0.8})_2\text{Te}_3$
for SPS at UCD



Activity Three: Calculation of the $(Bi_{0.2} Sb_{0.8})_2 Te_3$ compound mix

reaction: $Bi_2Te_3 + 4(Sb_2Te_3) = (Bi_{0.2}Sb_{0.8})_2Te_3$			
		atomic weight	
	Bi	208.98	
	Te	127.6	
	Sb	121.76	
		molecular weights	
2* Bi +3* Te_3	Bi_2Te_3	800.76	
2* Sb +3* Te_3	Sb_2Te_3	626.32	
4* Sb_2Te_3	$4Sb_2Te_3$	2505.28	
	$(Bi_{0.2}Sb_{0.8})_2Te_3$	3306.04	
		Target	10 grams
	Bi_2Te_3		2.422112255 grams
	Sb_2Te_3		7.577887745 grams

Completed



Specimen geometry Cold pressed/sintered disks



Cold pressed



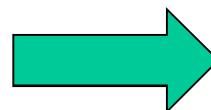
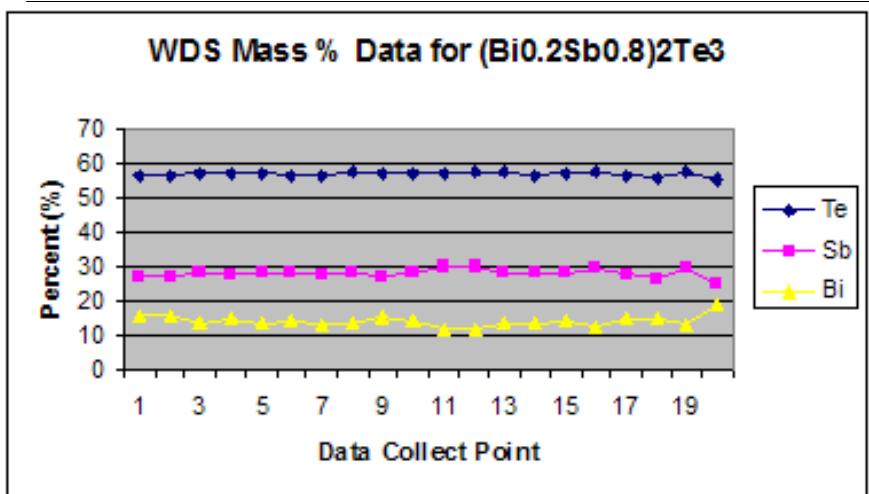
Sintered



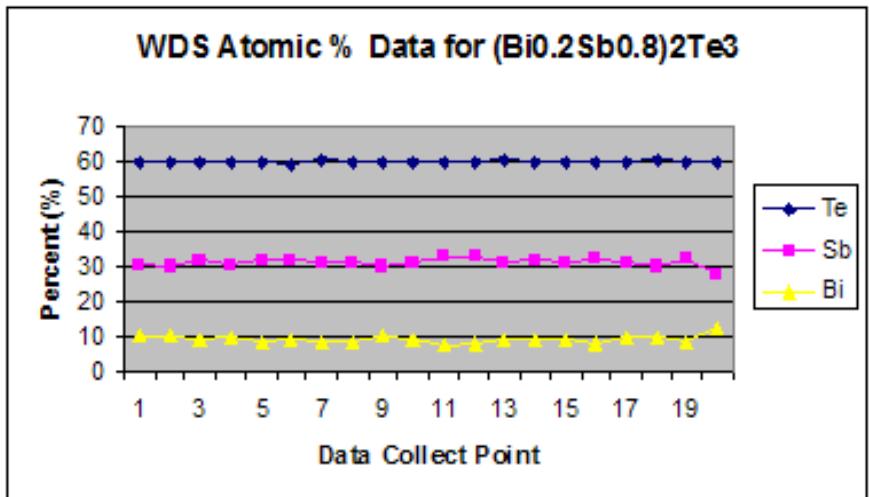
Sintering Process

- Make quartz tube to hold specimen during sintering
 - 13mm ID, 15.8mm OD tubing ; 2-ft stock
- Vacuum seal $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$ in tube
 - About 7×10^{-8} torr
- Hydrogen fill tube
 - Pump in ~ 100 torr of H after flushing system
- Melt quartz, seal tube for sintering
- Sinter at 550°C for 48 hrs ($\sim 90\%$ of T_m)
 - $T_m = 614^\circ\text{C}$ (887K)
- Test chemical composition by XRD, WDS
- Send to UC Davis for SPS processing

Chemical composition for SNL $(\text{Bi}_{0.2}\text{Sb}_{0.8})_2\text{Te}_3$ Sintered at 550°C for 48 hrs



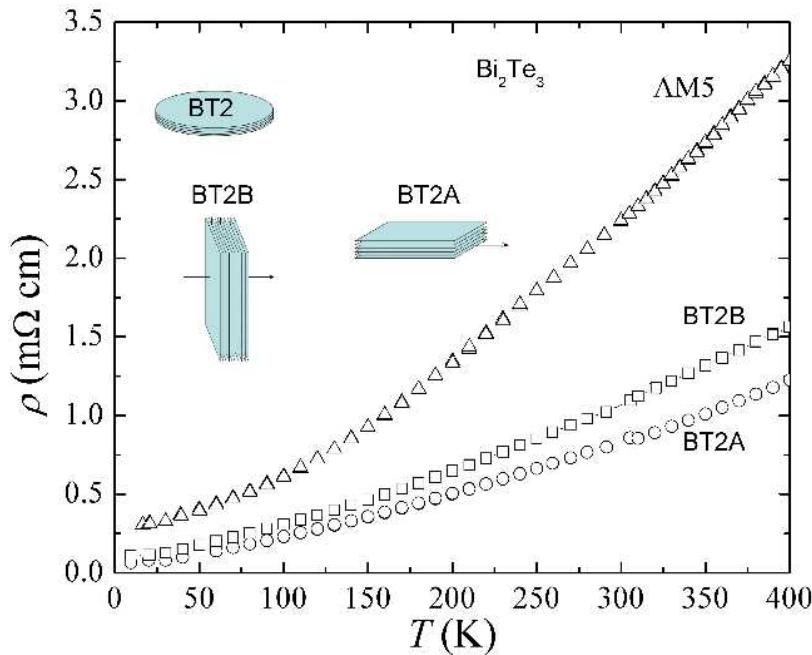
Te	56.79
Sb	28.05
Bi	14.11



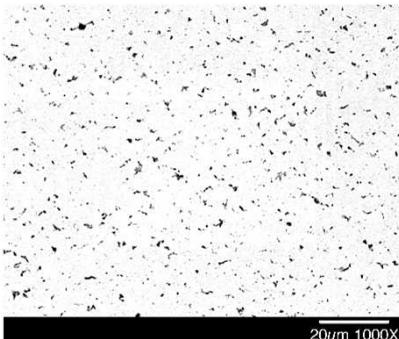
Te	59.91
Sb	31.00
Bi	9.09



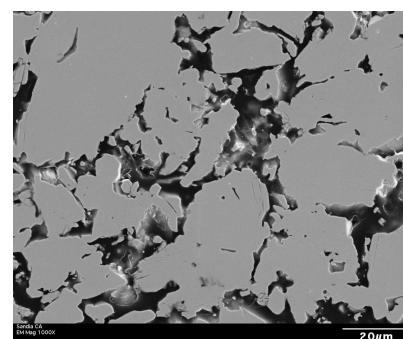
What do we know now???



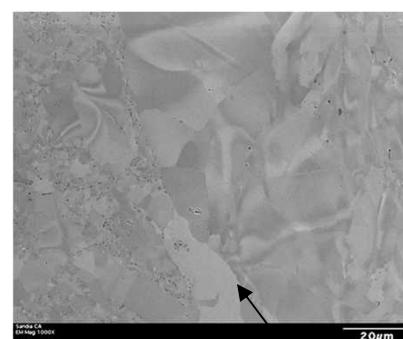
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- Others, such as power factor $\alpha^2 \sigma T$ and Figure of Merit $ZT = \frac{\alpha^2 \sigma T}{(\kappa_e + \kappa_{ph})}$



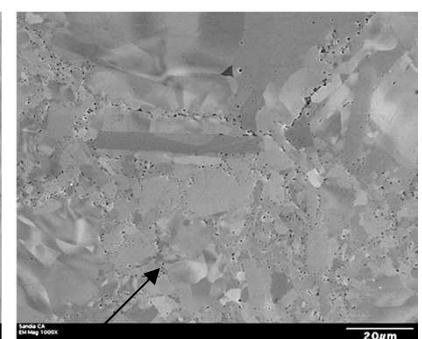
????



Cold press



BT1



SPS



Thermoelectric (TE) Property Measurements

- Will include graphs



New porosity



Activity Four: Set up experimental procedure to produce (Bi0.2,Sb0.8)Te3 powder mixture

For both the Bi₂Te₃ and Sb₂Te₃, I used the following settings:

200 rpm

20min on

30min pause

20min reverse

One repeat

This would get most of it broken up. If need be, I would hand grind a bit more.

I milled the two different materials separately, then weighted out the precise amounts, mixed them together by hand, and weighted out 3 grams for each pellet. I pressed each to about 4,000# under rough vacuum.

If you ever need to mill pure Bi, I used the following procedure:

300 rpm

30min on

30min pause

30min reverse

Three repeats

50 grams (Bi_{0.2}Sb_{0.8})₂Te₃ powder mix was prepared and ready to be sintered and compressed into pellets for the future microstructure and TE performance evaluations.



Activity five:

Investigate the TE performance of Bi₂Te₃ parent compound produced by SPS

- Prepare conference paper for MRS in spring, 2009
 - Completed microstructure and texture analyses
 - Completed electrical and thermal conductivity measurement of the two existing pellets
 - study porosity effect by preparing the pellet with high porosity using SPS at UCD (in progress)
- Determine the optimum porosity by varying temperature and pressure for