Nanostructured thermoelectric alloys obtained by mechanical alloying followed by hot extrusion or by microwave sintering

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Abstract

The introduction of multiple interfaces in bulk materials is expected to enhance their thermoelectric (TE) properties by lowering their thermal conductivity [1]. Bismuth telluride and lead telluride based alloys and composites nanostructured powders can be obtained by mechanical alloying (see Fig. 1) [2-4]. They are then processed into polycrystalline solids by hot extrusion. We have determined ranges of processing parameters in order to obtain mechanically strong bulk alloys, adequate for module fabrication processes [5], and whose TE performance has been proven in high power density modules (see for example Fig. 2)[2,6]. We have also succeeded in compacting lead telluride powders and producing bulk alloys by microwave sintering (see Fig.3). We analyze the structural and TE properties of different p-type bismuth antimony telluride composites produced so as to maintain the same Bi/Sb ratio (0.2/0.8) which we compare with a conventional $(Bi_{0.2}\ Sb_{0.8})_2Te_3$ homogeneous alloy. The minimum grain size of these composites, estimated at 180 nm, show a 50 % reduction compared to the homogeneous alloy. Hall Effect measurements suggest that the hole mobility values are limited by two dominant scattering mechanisms: ionized impurity and acoustic phonon scattering. The TE properties were evaluated via the Harman method from 300K up to 440K. No degradation of the power factor of the composites has been observed and peak dimensionless figure of merit (*ZT*) values range from 0.86 to 1.04. The thermal conductivity of the composites show a slight increase instead of the reduction expected due to the smaller grains and thus enhanced phonon scattering. Two concurrent factors can explain this increase: i) composites may not yet contain a significant number of grains with size sufficiently small to increase phonon scattering, and ii) each of the combined components of the composites corresponds to a composition with thermal conductivity higher than the minimum value corresponding to the homogeneous alloy. The introduction of multiple interfaces has to be implemented so as to override such limitations.

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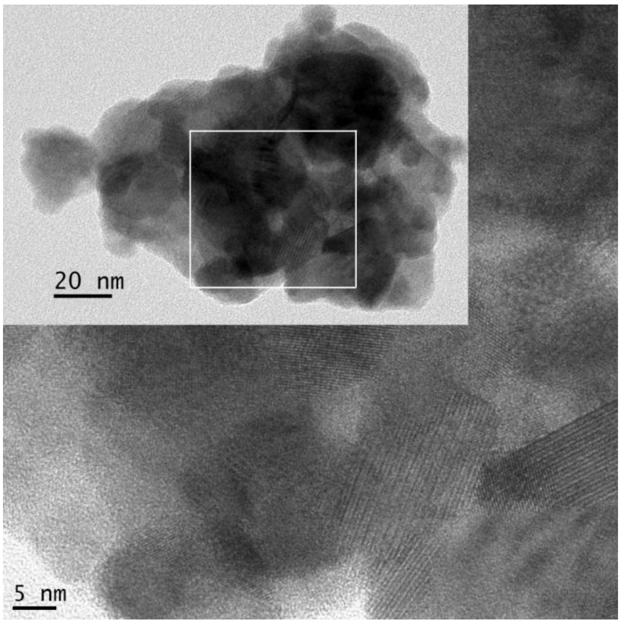

Fig. 1 (left) : High-resolution (HR) transmission electron microscope (TEM) images of *n*-type bismuth telluride based alloy *powders* after 2 hours of mechanical alloying in an attritor (from ref. 3). Nanograins with similar sizes of about 20-30 nm can be clearly identified on all images taken with different resolutions. The inset shows a bright field medium resolution TEM image illustrating the location of the observed nanograins inside the mother agglomerate.

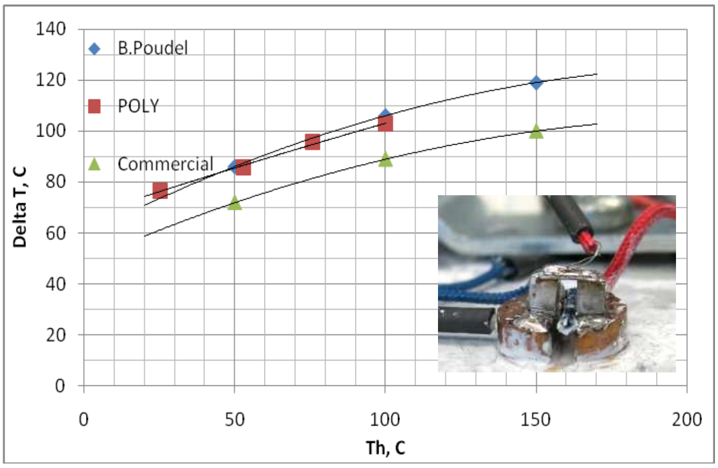

Fig. 2 (left): Variation of the maximum temperature difference generated by a thermocouple with p- and n- legs of dimensions: 1.8 x1.8 mm^2 (section) and 2.4 mm (length), as a function of the hot side temperature. The experimental setup is presented in the inset. The room temperature point for the curve labelled POLY corresponds to a 23 legs (0.6 x 0.6 mm^2 and 1 mm long) module produced by the Thermion Company from alloys extruded at the École Polytechnique de Montréal.

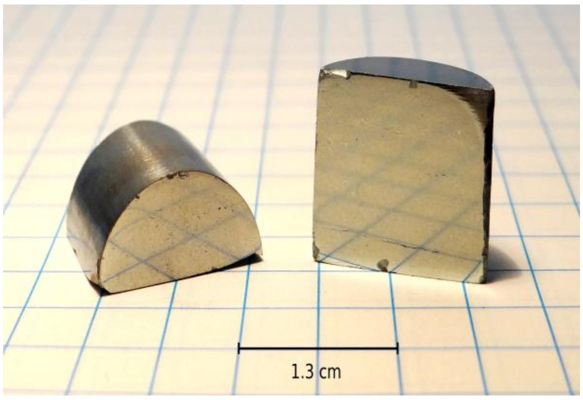

Fig. 3 (left) : Bulk PbTe produced by microwave sintering. The PbTe powder is homogenized by milling during 3 hours. It is then compacted for one minute under 1 metric ton pressure, and micro-wave sintered at 540 °C.