The effect of synthesis technique on the microstructure of high performance PbSe thermoelectric materials

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Abstract: During the past decade, the scientific community has witnessed a tremendous increase of research activity in the field of high efficiency thermoelectric materials (TE). PbTe is the most widely studied lead chalcogenide with high efficiency in the intermediate temperature range (600-800K). The sister material PbSe has received comparatively little attention, due to a lower figure-of-merit in this range. However, PbSe has several advantages since Se is 50 times more abundant, much less expensive and it has lower thermal conductivity than Te. Furthermore PbSe melts at a relatively higher temperature than PbTe and thus can perform better on higher temperatures (900K).

Various synthesis methods are used to fabricate PbSe such as, vapour-liquid-solid (VLS), Bridgman, microwave irradiation for PbSe nanoparticles, sublimation and from the melt. In any of these cases, a microscopically homogeneous material has to be assumed in order to acquire a comprehensive understanding between the measured thermoelectric properties (Seebeck coefficient or free carrier concentration) and the microscopic intrinsic features of each material. However, the general assumption about electronically homogeneous materials was disproved and the limitations of conventional Hall and Seebeck effect measurements which provide only one bulk average value were illustrated in previous work.

In this work, we present the effect of the synthesis technique on the microstructure of high performance PbSe. Direct melt-cooling of Pb, Se, Na and Spark Plasma Sintering (SPS) were applied in order to prepare p-type $Pb_{1-x}Na_xSe$ ($0 \leq x \leq 0.04$). The chemical composition of all samples and the morphology was determined by SEM/EDS analyses using a Jeol 840A scanning microscope. Fourier transform infrared spectroscopy (FTIR) measurements were performed in the range of 700-4000 cm^{-1} using a microscope with 100 μm iris (PerkinElmer spectrometer with i-series microscope), which enables FTIR mapping of the samples (Figure 1). The Plasmon frequency derived by the μ-FTIR measurements and a Drude model analysis, was used as a probe for mapping local in-homogeneities in the microstructure induced by different dopant content diluted in the matrix.

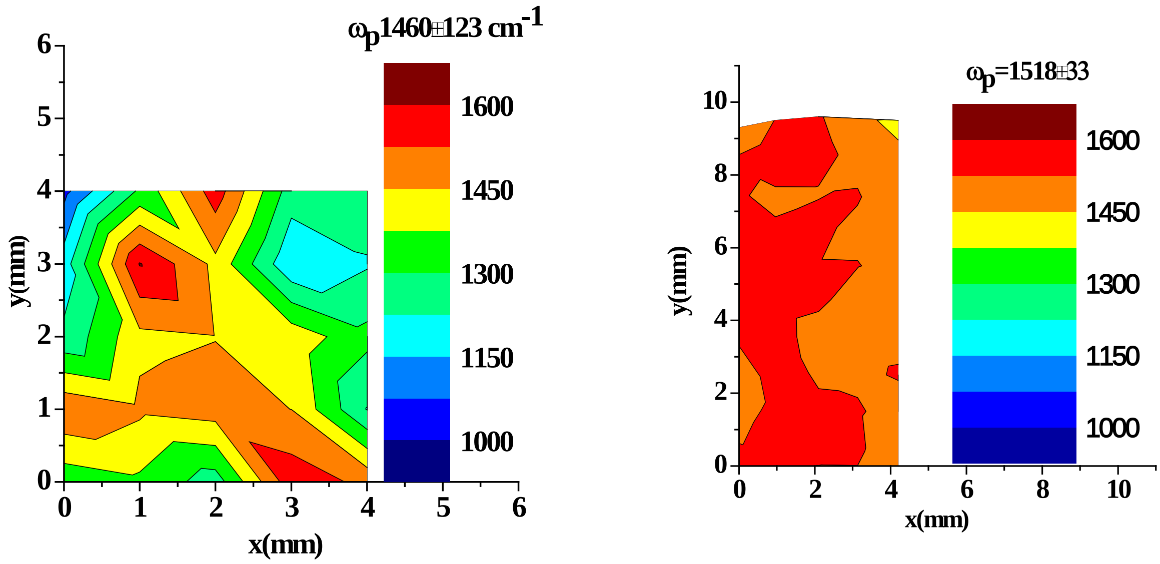

Fig.1 contour plots of plasmon frequency for two samples, by the melt (left figure) and SPS (right figure) with the same nominal Na content (1% Na). Sodium diluted in the matrix of SPS samples more homogeneously and in higher percentage, giving rise to higher plasmon frequency values.