

# Improved Thermoelectric Properties of Se-Doped *n*-Type PbTe<sub>1-x</sub>Se<sub>x</sub> (0 ≤ *x* ≤ 1)

RANITA BASU,<sup>1,2</sup> S. BHATTACHARYA,<sup>1</sup> RANU BHATT,<sup>1</sup> AJAY SINGH,<sup>1</sup>  
D.K. ASWAL,<sup>1</sup> and S.K. GUPTA<sup>1</sup>

1.—Technical Physics Division, Bhabha Atomic Research Centre, Mumbai 400085, India.  
2.—e-mail: ranitapaul@gmail.com

Enhancement of the thermoelectric figure of merit is of prime importance for any thermoelectric material. Lead telluride has received attention as a potential thermoelectric material. In this work, the effect of Se substitution has been systematically investigated in PbTe<sub>1-x</sub>Se<sub>x</sub>. The thermoelectric properties of synthesized alloys were measured in the temperature range of 300 K to 873 K. For the particular composition of *x* = 0.5,  $\alpha$  was highest at ~292  $\mu$ V/K, while *k* was lowest at ~0.75 W/m-K, resulting in the highest dimensionless figure of merit of  $ZT \approx 0.95$  at 600 K. The increase in thermopower for *x* = 0.5 can be attributed to the high distortion in the crystal lattice which leads to the formation of defect states. These defect states scatter the majority charge carriers, leading to high thermopower and high electrical resistivity. The dramatic reduction of the thermal conductivity for *x* = 0.5 can be attributed to phonon scattering by defect states.

**Key words:** Thermoelectric materials, defects, thermal conductivity

## INTRODUCTION

Solid-state thermoelectric energy conversion technology has prime importance, particularly for conversion of waste heat into electricity.<sup>1-6</sup> The efficiency of thermoelectric conversion is defined by the dimensionless figure of merit  $ZT \approx \alpha^2 T / \rho \kappa$ , where  $\alpha$  is the Seebeck coefficient,  $\rho$  is the electrical resistivity,  $\kappa$  is the thermal conductivity, and *T* is the average temperature of operation. To achieve a high  $ZT$  value, a material should exhibit high  $\alpha$  and low values of  $\rho$  as well as  $\kappa$ . Thus, a common strategy to improve  $ZT$  is to reduce  $\kappa$ , which has two contributions, i.e., electronic ( $\kappa_e$ ) and lattice ( $\kappa_l$ ). Tremendous efforts are being made to reduce  $\kappa_l$  in PbTe chalcogenides.<sup>7,8</sup> A significant enhancement of  $ZT$  (~1.7) in this material has already been demonstrated by introducing various nanoinclusions,<sup>9</sup> alloying with other thermoelectric materials such as PbS<sup>10-12</sup> or Bi<sub>2</sub>Te<sub>3</sub>,<sup>13</sup> and optimization of doping.<sup>14,15</sup> Recently, Pei et al. reported that simultaneous doping of PbTe (*p*-type) with Na and Se results in enhancement of

its thermoelectric properties.<sup>16</sup> Pei et al. have shown that, in *p*-type PbTe<sub>1-x</sub>Se<sub>x</sub>, convergence of many valleys at the desired temperature can be produced, which greatly enhances the thermoelectric performance if properly doped. A  $ZT$  value of 1.8 was achieved for *p*-type PbTe<sub>1-x</sub>Se<sub>x</sub>.<sup>16</sup> In this paper we report the synthesis and thermoelectric properties of *n*-type PbTe<sub>1-x</sub>Se<sub>x</sub>, because a successful strategy to increase the figure of merit is to modify an already promising compound by introduction of point defects through the synthesis of isostructural solid solutions.<sup>17</sup> Such solid solutions induce an environment of atomic mass fluctuation throughout the crystal lattice, i.e., disorder formation, which leads to strong phonon scattering and thus significantly decreased thermal conductivity and enhanced  $ZT$ .<sup>17</sup> Moreover, a practical thermoelectric device requires both *n*- and *p*-legs with similar mechanical and thermoelectric properties. In the present work, we report a systematic study on the high-temperature thermoelectric properties of *n*-type PbTe<sub>1-x</sub>Se<sub>x</sub> (0 ≤ *x* ≤ 1). We demonstrate that, for *x* = 0.5,  $ZT$  shows its highest value of ~0.95 at 600 K, because of the incorporation of high density of defect states.<sup>18</sup>

(Received July 9, 2012; accepted May 22, 2013;  
published online June 12, 2013)