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CEPTED MANUSCRIPT

# Optimization on the Figure-of-Merit of *P*-type Ba<sub>8</sub>Ga<sub>16</sub>Ge<sub>30</sub> Type-I Clathrate Grown via the Bridgman Method by Fine Tuning Ga/Ge Ratio

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#### Abstract

The thermoelectric properties of polycrystalline *p*-type Ba<sub>8</sub>Ga<sub>16</sub>Ge<sub>30</sub> type-I clathrates were explored by fine tuning Ga/Ge ratios of clathrates and compared with the carrier properties. The clathrates with five different Ga/Ge ratios were grown from melt by means of the vertical Bridgman method with the raw materials prepared by vacuum arc melting. The carrier concentration of the clathrates decreases with increasing the Ga/Ge ratio from 0.567 to 0.586. The temperature dependence of electrical conductivity shows that these clathrates are heavily doped semiconductors. The Seebeck coefficient increases with the Ga/Ge ratio at room temperature, while the sample with the Ga/Ge ratio of 0.579 has the highest Seebeck coefficient at 550°C. These tendencies can be explained in dependence of carrier concentration. The clathrate with a Ga/Ge ratio of 0.572 has the optimal dimensionless figure-of-merit of 1.0 at 500°C, while its power factor and thermal conductivity are  $2.11 \times 10^{-3}$  W/mK<sup>2</sup> and 1.55 W/mK respectively. However, the clathrate of with Ga/Ge ratios between 0.567 and 0.579 possesses *zT* values close to 1.0. This indicates a Ga/Ge ratio range for steady *zT* values.

Keywords: thermoelectric materials, figure-of-merit, clathrate, Ba<sub>8</sub>Ga<sub>16</sub>Ge<sub>30</sub>, Bridgman method, carrier concentration.

#### 1. Introduction

Thermoelectric generation recovers the thermal energy released from furnaces, engines or boilers into electrical work and provides a possible way in using energy more efficiently. More than 75% of total thermal energy is emitted as the waste heat from a vehicle engine at up to a peak temperature about 500°C [1]. A thermoelectric device can quietly operate in a limited space and therefore is ideal for automobile applications. The efficiency of a thermoelectric device depends on the temperature differences applied on it and the figure-of-merit of its material. The higher is the figure-of-merit, the higher is its efficiency. The figure-of-merit, normally symbolled as *z*, is defined as  $s^2\sigma/\kappa$  where *s*,  $\sigma$  and  $\kappa$  are respectively the Seebeck coefficient, electrical conductivity and

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