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Crystallographic, Thermoelectric, and Mechanical Properties of Polycrystalline $Ba_8Al_xSi_{46-x}$ Clathrates

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The Al content dependence of crystallographic, thermoelectric, and mechanical properties is reported for polycrystalline $Ba_8Al_xSi_{46-x}$ (nominal x = 15 to 17) clathrates prepared by combining arc melting and spark plasma sintering methods. The elastic constants and the coefficient of thermal expansion (CTE), which are also important properties for designing thermoelectric devices, are presented. Powder x-ray diffraction, scanning electron microscopy, and energy-dispersive x-ray spectroscopy (EDX) indicate that the type I clathrate is the major phase of the samples but impurity phases (mainly $BaAl_2Si_2$, Si, and Al) are included in the samples with high Al contents. The actual Al content x determined by EDX ranges from approximately 14 to 15. The absolute value of the Seebeck coefficient increases and the electrical conductivity decreases as the Al content increases. The changes in Seebeck coefficient and electrical conductivity are explained in terms of the dependence of the carrier concentration on the Al content. The elastic constants and the CTE of the samples depend weakly on the Al content. Some of the properties are compared with reported data of single crystals of $Ba_8Al_{16}Ge_{30}$, $Ba_8Ga_{16}Ge_{30}$, $Sr_8Ga_{16}Ge_{30}$, silicon, and germanium as standard references. The effective mass, Hall carrier mobility, and lattice thermal conductivity, which govern the transport properties, are determined to be $\sim 2.4m_0$, ~ 7 cm^2 V^{-1} s^{-1}, and ~ 1.3 W m^{-1} K^{-1}, respectively, for actual Al content x of about 14.77. The thermoelectric figure of merit ZT is estimated to be about 0.35 at 900 K for actual Al content x of about 14.77.

Key words: Clathrate, $Ba_8Al_xSi_{46-x}$, thermoelectric properties, coefficient of thermal expansion, elastic modulus, elastic constant

INTRODUCTION

There has been growing interest in the search for materials with high thermoelectric energy conversion efficiency.[1] If the efficiency is sufficiently high, solid-state devices using thermoelectric materials can generate electricity directly from the enormous amount of unused waste heat as well as natural heat resources such as solar heat and geothermal sources. The thermoelectric efficiency of a material is evaluated by the dimensionless thermoelectric figure of merit $ZT = S^2\sigma T/\kappa$, where S is the Seebeck coefficient, σ is the electrical conductivity, κ is the thermal conductivity, and T is the absolute temperature.[2] To maximize the ZT of a material, a large absolute value of the Seebeck coefficient, a high electrical conductivity, and a low thermal conductivity are required. To achieve the maximum ZT in a material, it is essential to study comprehensively the transport properties, especially the material parameters such as the density-of-states effective mass m^*, the carrier mobility μ, and the lattice thermal conductivity κ_L, which are closely related to ZT as $ZT \propto m^{*3/2}\mu T/\kappa_L$.[2]

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