Journal of ELECTRONIC MATERIALS, Vol. 42, No. 8, 2013
DOI: 10.1007/s11664-013-2631-y

Thermoelectric Properties of $Y_{1-x}Ag_xBaCo_4O_{7+\delta}$ Ceramics

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The thermoelectric properties of the Ag-doped ceramics $Y_{1-x}Ag_xBaCo_4O_{7+\delta}$ (x = 0.0, 0.05, 0.1, 0.15, and 0.2) were investigated from 373 K to 973 K. The results show that the doping of Ag can reduce the electrical resistivity. The Seebeck coefficients of the samples decrease when the Ag doping amount is small, but increase when the Ag doping amount is large. The activation energy of the electrical conductivity was calculated using Arrhenius plots, and it was found that the activation energy descends with increase of the Ag doping amount. According to the power factors, the optimum Ag doping amount is x = 0.15, which results in a higher power factor of 81 μW m^{-1} K^{-2} at 973 K, 72.7% higher than for the sample without Ag doping.

Key words: Thermoelectric materials, element doping, $Y_{1-x}Ag_xBaCo_4O_{7+\delta}$

INTRODUCTION

Recently, thermoelectric materials have attracted much attention because they can convert waste heat into useful electricity without any moving parts and can be used as cooling devices without harmful liquids. Most of the research is focused on thermoelectric alloys, such as Bi_2Te_3-, PbTe-, and Co_4Sb_{12}-based alloys, because such alloys have higher figure of merit ZT values.[1] The figure of merit is defined as $ZT = \sigma S^2 T/\kappa$, where σ is the electrical conductivity, S is the Seebeck coefficient, κ is the thermal conductivity, and T is the absolute temperature. However, these alloys have lower melting points, are easily decomposed, and are not suitable for use in oxidizing surroundings. Thus, another important research area is oxide thermoelectric materials, such as layered cobaltites $Bi_2Sr_2Co_2O_y$, $NaCo_2O_4$, and $Ca_3Co_4O_9$.[2,3]

Recently, another type of layered cobalt oxide $RBaCo_4O_{7+\delta}$ (R = Y, Dy–Lu, and In) synthesized by Valldor et al.[4,5] has attracted much attention because of its favorable physical and chemical properties.[6–11] For use as thermoelectric materials, Caignaert et al.[9] and Maignan et al.[12] reported the electrical resistivity and Seebeck coefficients of $YbBaCo_4O_{7+\delta}$, $Yb_{0.5}Y_{0.5}BaCo_4O_{7+\delta}$, and $YBaCo_4O_{7+\delta}$ at low temperature, and found a precise temperature adjustment of the resistivity transition between ~60 K (R = Lu) and ~310 K (R = Y). Our earlier investigation on the influence of R^{3+} ion sizes on the high-temperature transport properties of $RBaCo_4O_{7+\delta}$ (R = Dy, Ho, Y, Er) revealed that the $YBaCo_4O_{7+\delta}$ materials have the highest power factors.[13] Therefore, $YBaCo_4O_{7+\delta}$ was selected for further improvement of its thermoelectric properties by Zn substitution for Co.[14] However, the result was contrary to our expectation, due to decrease of the carrier mobility. Because $YBaCo_4O_{7+\delta}$ is a p-type semiconductor, to increase its electrical conductivity, low-valence ions should be selected for doping. Therefore, in this paper, we report the influence of partial substitution of Ag^{1+} for Y^{3+} on the thermoelectric properties of $YBaCo_4O_{7+\delta}$ ceramics.

EXPERIMENTAL PROCEDURES

$Y_{1-x}Ag_xBaCo_4O_{7+\delta}$ (x = 0.0, 0.05, 0.1, 0.15, and 0.2) samples were prepared using the solid-state reaction method. Stoichiometric amounts of Y_2O_3, Ag_2O, $BaCO_3$, and Co_3O_4 raw materials were mixed thoroughly in an agate mortar. The mixed powder was heated slowly up to 1000°C in a box furnace and

(Received March 12, 2013; accepted May 2, 2013;
published online June 7, 2013)