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Fabrication and Evaluation of a Skutterudite-Based Thermoelectric Module for High-Temperature Applications

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We report a straightforward methodology for the fabrication of high-temperature thermoelectric (TE) modules using commercially available solder alloys and metal barriers. This methodology employs standard and accessible facilities that are simple to implement in any laboratory. A TE module formed by nine n-type $Yb_xCo_4Sb_{12}$ and p-type $Ce_xFe_3CoSb_{12}$ state-of-the-art skutterudite material couples was fabricated. The physical properties of the synthesized skutterudites were determined, and the module power output, internal resistance, and thermocycling stability were evaluated in air. At a temperature difference of 365 K, the module provides more than 1.5 W cm^{-3} volume power density. However, thermocycling showed an increase of the internal module resistance and degradation in performance with the number of cycles when the device is operated at a hot-side temperature higher than 573 K. This may be attributed to oxidation of the skutterudite thermoelements.

Key words: Thermoelectric generator, skutterudites, manufacturing, electrical contact resistivity, power output, device fabrication

INTRODUCTION

Thermoelectric (TE) devices have been identified as promising converters for energy harvesting due to their capability to convert heat into electricity.[1] A great amount of heat is released from high-temperature systems such as internal combustion engines, industrial furnaces, and incinerators. A wide range of materials able to work at different temperature ranges have recently appeared showing relatively high efficiencies, given by the TE figure of merit ZT.[2] However, there are significant parasitic losses when the materials are integrated to form a complete device, which lower the device efficiency below the value predicted on the basis of the ZT of the component materials. Reduction of these losses is a key challenge to achieve efficient TE devices.[3] This requires the creation of contacts with suitable electrical and thermal properties. When the device operates under a large temperature difference, thermomechanical stress, diffusion, and chemical reaction of materials at the interfaces become important concerns. As TE materials have high electrical conductivities, very low electrical contact resistances are required between the materials and the electrodes, which themselves should have high electrical and thermal conductivities relative to the TE materials. Additionally, the electrodes often include diffusion barriers to prevent diffusion of certain electrode materials into the thermoelements and vice versa, because such diffusion can produce poisoning and degradation of the materials. Moreover, commercial solder alloys with melting points in the 573 K to 973 K range are difficult to find,[4] and sometimes suitable joining materials have to be synthesized[5,6] or the contacts prepared by using additional techniques such as spark plasma sintering[7,8] or hot-pressing,[9] complicating the manufacturing processes. Herein, we present a simple methodology for the fabrication of high-temperature TE devices. It is based on the use

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