Manufacturing & Prototyping

d Metallization for Yb₁₄MnSb₁₁-Based Thermoelectric Materials

Process enables device fabrication using advanced, high-temperature thermoelectric material.

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Thermoelectric materials provide a means for converting heat into electrical power using a fully solid-state device. Power-generating devices (which include individual couples as well as multicouple modules) require the use of ntype and p-type thermoelectric materials, typically comprising highly doped narrow band-gap semiconductors which are connected to a heat collector and electrodes.

To achieve greater device efficiency and greater specific power will require using new thermoelectric materials, in more complex combinations. One such material is the p-type compound semiconductor Yb₁₄MnSb₁₁ (YMS), which has been demonstrated to have one of the highest ZT values at 1,000 °C, the desired operational temperature of many space-based radioisotope thermoelectric generators (RTGs).

Despite the favorable attributes of the bulk YMS material, it must ultimately be incorporated into a power-generating device using a suitable joining technology. Typically, processes such as diffusion bonding and/or brazing are used to join thermoelectric materials to the heat collector and electrodes, with the goal of providing a stable, ohmic contact with high thermal conductivity at the required operating temperature.

Since YMS is an inorganic compound featuring chemical bonds with a mixture of covalent and ionic character, simple metallurgical diffusion bonding is difficult to implement. Furthermore, the Sb within YMS readily reacts with most metals to form antimonide compounds with a wide range of stoichiometries. Although choosing metals that react to form high-melting-point antimonides could be employed to form a stable reaction bond, it is difficult to limit the reactivity of Sb in YMS such that the electrode is not completely consumed at an operating temperature of 1,000 °C. Previous attempts to form suitable metallization layers resulted in poor bonding, complete consumption of the metallization layer or fracture within the YMS thermoelement (or leg).



Metallized Yb₁₄MnSb₁₁ Leg, terminated on both ends with Mo layers.

An approach to forming a stable metallization layer to consolidated YMS parts or legs has been developed, however, using thin molybdenum foils. The foil is diffusion bonded to the YMS part at temperatures above 900 °C, under the application of adequate pressure for several hours. Use of a thin foil eliminates the fracture typically observed within YMS parts when thicker foils are used (induced by thermo-mechanical stresses) as seen in the figure. The metal can be bonded prior to dicing of the legs from a hot-pressed/consolidated YMS form, or it can be bonded to a pre-cut leg of the correct geometry. The contact is thermally stable with respect to YMS, exhibiting no de-bonding or increase in contact resistance after a 1,000 °C vacuum heat treatment for 1,500 hours. This metallization layer may then be bonded or brazed to other components, such as heat collectors or current-carrying electrodes. To enable successful bonding of the molybdenum metallization requires the preparation of YMS legs of sufficient strength, which is facilitated by the hot pressing of YMS powders above 900 °C (using proper cooling rates to minimize residual stress formation and avoid fracture of the parts).

This metallization layer may then be bonded or brazed to other components, such as heat collectors or current-carrying electrodes. Furthermore, it can be implemented at both the hot and cold sides of the leg. It can also be applied as a metallization layer to other similar compositions of thermoelectric materials. The specific nature of the interaction between Mo and YMS is still under investigation; however, it is clear the molybdenum reacts sufficiently to form an adequate bond, without the extensive reaction observed with similar metals such as nickel, niobium, or titanium. The process may be similar to that involved in the formation of other metal/ceramic bonds, wherein the metal has limited solubility in the ceramic material (for example, in diffusion bonded metal/alumina joints).

This work was done by Samad Firdosy, Billy Chun-Yip Li, Vilupanur Ravi, Jeffrey Sakamoto, Thierry Caillat, Richard C. Ewell, and Erik J. Brandon of Caltech for NASA's Jet Propulsion Laboratory. Further information is contained in a TSP (see page 1). NPO-46670