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Thermoelectrics

Make contact with silver

Contact design is a major challenge in the development of thermoelectric devices. New research shows that silver nanoparticles soldered at low temperature can sustain high service temperatures, improving the stability of devices operating across a wide range of temperatures and exploiting the maximum working temperature of thermoelectric materials.

Johannes de Boor

Progress in the development of thermoelectric (TE) materials has been impressive in recent decades but is yet to be transferred into performance improvement at the device level. Contact development – the design of an interface that joins the TE material and the metal interconnects (or bridges) – is a key bottleneck [1]. Contacts should fulfil various requirements: low electrical and thermal resistances, inhibition of undesired chemical reactions and interdiffusion of elements between the TE material and other components of the device, and mechanical integrity – all these requirements need to be met under large and often varying temperature gradients. To comply with them, a multilayer design is often used for the contact layer: this usually consists of a diffusion barrier layer, which is deposited on top of the TE material, and a solder layer, which is connected to the bridge (Figure 1a).

The choice of the solder material, in particular, is not straightforward: the solder layer should remain stable at the (maximum) working temperature of the TE material and the temperature at which the layer can be soldered to the device stack should be below the working temperature of the TE material so that the latter does not degrade during the soldering process. The combination of these two requirements cannot be fulfilled by traditional soldering or brazing materials (materials soldered at high temperatures).

Writing in Nature Energy [2], Jun Mao, Mingyu Li, Zhifeng Ren, Qian Zhang and team at the Harbin Institute of Technology, Chinese Academy of Sciences and University of Houston use silver nanoparticles as a soldering material, which allows low temperatures to be used for soldering (573 K) while retaining stability at working temperatures over 1000 K. The researchers demonstrate high conversion efficiencies and thermal stability in three TE generators made from very different TE materials that operate across a wide range of temperatures, demonstrating the general applicability of using Ag nanoparticles as solder layer.

By systematically analysing literature, the researchers emphasize the incompatibility between the requirements of working and soldering temperatures for TE materials and traditional solders: the temperatures TE materials can withstand during the soldering step are usually just above their maximum service temperature (Figure 1b). The soldering temperature of traditional solder materials, instead, must be significantly above their working temperature to avoid creep failure.

Thus, a TE device employing solder materials whose processing temperature is at or below the working temperature of the TE material will be restricted in the maximum temperature at which it can operate without risking long term failure. As such, a fraction of the temperature difference that the TE material could in principle harvest is not exploited (referred to as “wasted ΔT” in Figure 1b), making the TE device less efficient. On the other hand, using a solder with a higher working temperature is in principle possible but requires going above the working temperature of the TE material during the soldering step (“overheating”). TE materials require an optimized carrier concentration, balancing various material properties through the control of the concentration of extrinsic (doping) or intrinsic point defects. Heating the material above its working temperature may change the previously optimized carrier concentration, e.g. by sublimation and hence causes significant material degradation even for short overheating times. [3, 4] Overheating can also compromise the mechanical stability of the TE material.

The key idea from Zhang and team is to decouple the soldering and the working temperature of the solder material to make it compatible with the TE material. Owing to their high surface to volume ratio and hence high surface energy, Ag nanoparticles can be bonded to other metals at temperatures which are far below the working temperature of bulk silver –this is what makes Ag nanoparticles different from traditional bulk solders. The researchers show that Ag nanoparticles can be used like solder materials with a “soldering” temperature of 573 K and withstand device operating temperatures above 1000 K without showing signs of thermally-induced degradation.

The usage of Ag nanoparticles as solder materials is not entirely new [5], but has not been optimized for TE devices before. Zhang and team identify processing conditions for the Ag nanoparticles that yield very good mechanical properties. They also demonstrate that Ag nanoparticle solders are applicable to TE materials that operate at different temperatures, from 300 K to 1000 K, i.e. addressing a wide majority of potential applications of TE devices. They prepare low- (Bi_2Te_3-based), medium- (PbTe-based), and high-temperature (half-Heusler-based) TE generators, all showing relatively high and stable performance and no degradation at the interfaces. In particular, the PbTe prototype with around 11% conversion efficiency is among the best ever reported single-stage TE generators.

So, is the job done? Even though the results are certainly encouraging, as Zhang and team observe low electrical contact resistances for their devices, they also show that roughly 20-30% of power and efficiency are lost due to temperature drops at the interfaces, indicating the need to optimize the interface design with respect to the thermal properties.

Furthermore, Ag nanoparticles solders might not be a definitive or universal solution. First of all, as also shown in the inset of Figure 1, the contact design consists typically of at least two different materials, the solder and the diffusion barrier. The work of Zhang and team thus only provides a solution for a part of the problem while relying on existing diffusion barriers for the device making. The use of known diffusion barriers was feasible as the researchers employ traditional and well-studied TE materials for which barrier materials are known. However, the identification of a diffusion barrier is a material-specific problem that is yet to be solved for many TE materials and is usually addressed by time-consuming experimental work. However, with the expanding computational capabilities diffusion barrier pre-selection strategies are being established. These are based on the prediction of newly formed phases during contacting, energetical barriers for interdiffusion or the potential impact of point defects formed due to interdiffusion [6-8]. The use of Ag nanoparticles as solder materials might set further requirements on the diffusion barrier: for instance, there might be the need for preventing Ag diffusion into the TE material.

The work of Zhang and team presents an important step that could accelerate the development of TE devices and hence their widespread application. If a similarly reliable approach for the diffusion barriers can be found, the future of TE device manufacturing would look brighter, indeed.

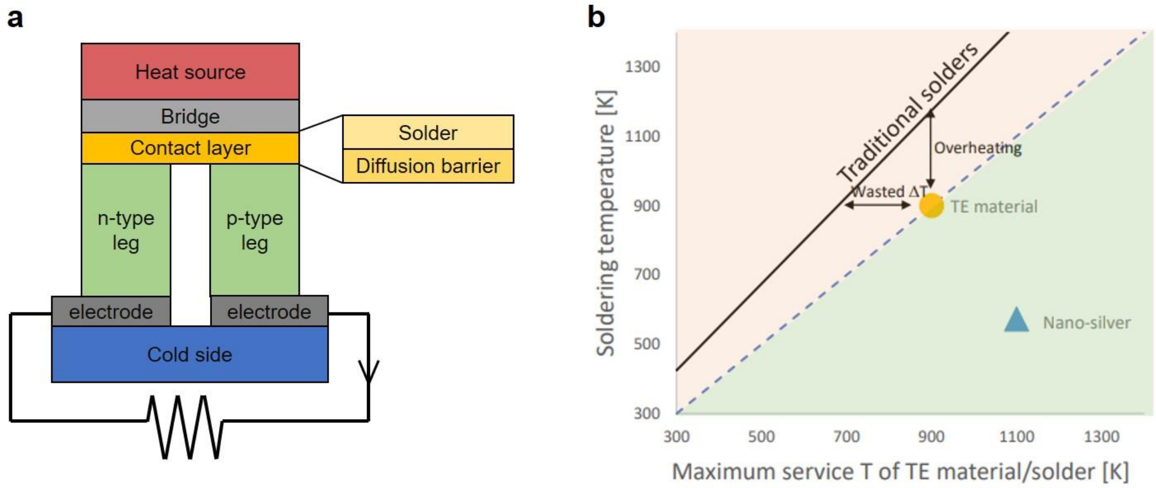

Figure 1. Working and soldering temperature for thermoelectric material and solder. a) The structure of a TE generator with zoom-in on the multilayer design of the contact layer. **b)** Traditional solders have a maximum working temperature below their soldering temperature (black line). The dashed line indicates the maximum temperature TE materials with different working temperatures can withstand during the soldering step: solder materials with suitable soldering and working temperatures should appear in the green area. An hypothetical TE materials with a maximum working temperature of 900 K is shown with an orange dot to illustrate the impact of using traditional solders: either the TE material is heated above its working temperature in the process (overheating) or a lower temperature difference than that the TE material can harvest is exploited (wasted ΔT) to guarantee long term stability of the devices. Silver nanoparticles, however, have a working temperature far above their “soldering” temperature and therefore allow to exploit the full potential of most TE materials. Panel b is adapted from ref. [2].

Competing Interest

The author declares no competing interests.

Affiliation

Johannes de Boor, Johannes.deboor@dlr.de

German Aerospace Center (DLR), Institute of Materials Research, Cologne, Germany

University of Duisburg-Essen, Institute of Technology for Nanostructures (NST) and CENIDE, D-47057 Duisburg, Germany

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