All-silicon thermoelectric micro/nanogenerator including a heat exchanger for harvesting applications

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HIGHLIGHTS

- Energy harvesting is in high demand for powering IoT devices.
- Managing the thermal match is crucial to enhance the performance of a thermogenerator.
- Different Si-based materials are successfully integrated into thermal generators.
- The heat exchanger integration route is compatible with microdevice fragility.
- Power density attained is in accordance with the requirements of IoT microsensors.

GRAPHICAL ABSTRACT

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ABSTRACT

This paper describes a specific route for the complete integration of a novel planar thermoelectric microgenerator (μTEG) that can operate under environmental conditions using commercial miniaturized heat exchangers. The proposed heat exchanger integration process is compatible with the fragility of planar micromachined silicon structures. The main structure of the μTEG is built around a micromachined silicon platform defined by silicon microfabrication technologies. Different silicon-based materials, such as bottom-up grown silicon and silicon-germanium nanowire arrays as well as top-down fabricated silicon microbeams are used as thermoelectric materials. μTEGs with those materials are characterized both before and after heat exchanger integration. The presence of the heat exchanger increases the μTEG performance significantly and power densities around 40 μW cm⁻² are obtained when placed on a heat source at 100 °C and exposed under natural convection to a surrounding ambient at room temperature.

1. Introduction

Energy harvesting or scavenging is in high demand for powering Internet of Things devices. Due to their potential to convert heat into electricity, thermoelectric microgenerators (μTEGs) are considered as candidates for power sources in wireless sensor networks when a waste
heat source is available. Their reduced size and zero maintenance cost, which make them applicable in different environments, and their potential compatibility with IC technology (especially when implemented in silicon) provide additional advantages for several applications. Therefore, many studies have employed different strategies to develop thermoelectric microgenerators [1–10].

One important limitation of μTEGs is the quite moderate temperature differences available across the thermoelectric material when placed on a waste heat source due to the small size of the device. The thermal resistance between the small surface and the ambient is so high that under natural convection the device is only capturing a minimum fraction of the total temperature difference existing between the heat source and the ambient. Most of the studies reporting on μTEGs specify their performance applying a temperature difference (ΔT) to the system by means of artificially dissipating a power on it, or actively pinning the temperature at both ends of the device. However, in a real application case, which is harvesting the energy from the waste heat source, ΔT should develop naturally across the μTEG itself under the operating ambient conditions. To achieve a high ΔT across the integrated thermoelectric material (i.e. to maximize the power output), optimizing the thermal link of the generator to heat sink and heat source is necessary. In order to do so, integration of a heat exchanger on the cold side of a μTEG is crucial. However, its implementation on such micron-featured devices is also challenging especially for planar μTEGs, which usually include a fragile suspended platform.

This study reports on the integration of a heat exchanger on novel all-silicon (Si) based μTEGs with the aim of maximizing the power output [11]. Some researchers have conducted measurements/simulations of vertical μTEGs including a heat spreader or a heat sink [12–14]. In the case of planar devices, Yuan et al. [15] recently reported on a high thermal resistance μTEG using the Si substrate as heat concentrator and evaporator on both hot and cold ends of the device. Yang et al. [16] presented a planar μTEG in which the bulk Si substrate was cooled by a heat sink and a fan. In that work, the suspended platform was heated by a distant heat source to prevent breaking. Researchers from Singapore [4] embedded thermocouples between vacuum cavities partially coated with a metal layer to produce a heat sink. To the best of the authors’ knowledge, this is the first attempt to define a specific route for the integration of the heat sink on a planar μTEG using commercial heat exchangers in a way that is compatible with the fragility of planar μTEGs. Moreover, the performance of our all-Si based μTEG is reported under operating conditions both with and without the heat exchanger. Although, scientifically speaking a heat sink is a thermal energy reservoir able to receive large amounts of heat without changing its temperature, and a heat exchanger is a physical passive element enhancing the heat transfer between a hot body and a surrounding fluid, both terms will be used indistinctly for the latter throughout the text.

2. Materials and methods

2.1. Thermoelectric microgenerator (μTEG)

The μTEG reported in this study has a planar architecture and is fabricated by top-down MEMS micromachining techniques on silicon on insulator (SOI) substrates. Standard photolithography, thin film processing, anisotropic wet etching and deep reactive ion etching were applied for fabrication. Architecture details of the μTEG are given in Refs. [17,18]. Briefly, it is composed of a suspended rectangular Si platform (SOI device layer) surrounded by a bulk Si rim (SOI device layer + Si handle wafer). Bottom-up grown Si or silicon-germanium (SiGe) nanowires (NWs) or top-down Si microbeams are monolithically integrated between those two areas (Fig. 1a). When placed on top of a waste heat source, the bulk Si rim acts as the hot side and the suspended platform acts as the cold side.

The thermoelectric material is placed bridging the predefined trench walls on three sides of the rectangular platform. In devices with NWs, they are grown by means of a post-process Chemical Vapor Deposition - Vapor Liquid Solid (CVD-VLS) method [19]. In devices with Si microbeams, they are defined simultaneously with the rest of the microplatform. The fourth side of the platform is occupied by a perforated low thermal conductivity silicon nitride (Si3N4) membrane that is released by a wet anisotropic etch (the same process used to define the < 111 > trench walls). It should be noted that our planar μTEG is fabricated employing a uni-leg architecture, in which the thermocouple is formed by a semiconductor thermoelectric material (p-type Si or SiGe NW arrays or Si microbeams) and a tungsten metal layer. This metal layer also bridges platform and rim (i.e. cold and hot sides) with the support of the Si3N4 membrane.

Fig. 1b shows a standard chip (7 × 7 mm²) which features four single devices, each device represents a single thermocouple as the one previously described. For the μTEGs with NWs, those four devices exhibit different number of trenches between the suspended platform and the bulk Si rim. This is a way to increase the effective length of the NWs by using a single growth process. Each trench has a width of 10 μm and the number of trenches ranges between 1 and 4 (referred as T1, T2, T3 and T4 devices). An example of a three trench device with Si NWs grown (T3) is shown in Fig. 1c. On the other hand, for Si microbeams (Fig. 1d), all devices feature a single trench (10 μm) but different number of beams bridging the hot and the cold sides are considered.

2.2. Integration of a heat exchanger

The heat exchanger assembly is designed in a way that the bulk Si rim acts as the hot side and the suspended platform acts as the cold side. A reversed heat flow option (Si rim acting as the cold side and platform as the hot side) was considered but simulations showed a worst thermal performance, so only the first option will be shown hereafter.

Since the proposed μTEG has a planar layout built around a suspended platform, the heat sink cannot be placed directly on top of this fragile platform. Additional components are included in the assembly to prevent the application of an excessive vertical force on the suspended platform. Thermal conductivities and dimensions of these components are critical to optimize the heat flow. In order to be able to measure the performance of the μTEG once assembled, a printed circuit board (PCB) is designed onto which the μTEG is wire bonded. The PCB is made of FR-4, which is composed of woven fiberglass cloth with an epoxy resin binder that is flame resistant. Fig. 2 shows the PCB layout and a picture of a wired bonded chip.

Fig. 3 shows the components of the heat exchanger assembly together with the cross-sectional schematic of the assembly. The first component of the assembly is a 500 μm thick copper (Cu) plate (4 × 2 cm²) with 4 holes (1.5 mm in diameter) for the alignment with the other components using pins. On that Cu plate, a smaller square Cu piece (6 × 6 mm², 1 mm thick) is attached using a solvent based alcoholic paste. Alignment of this Cu piece is done by using the perforated PCB, which exhibits a matching cavity and also includes the aligning holes. These Cu components are used to transfer the heat from the hot plate to the base of the bulk Si rim of the device due to their high thermal conductivity. The PCB is fixed to these bottom Cu components with a silver-based epoxy (EPO-TEK®H70E). Then, the attached components are placed inside the oven at 120 °C for 15 min to cure the epoxy. The same curing step is repeated after each application of an epoxy. Next, the μTEG is placed inside the cavity of the PCB and fixed. It is important to note that the chip should be in contact with the Cu piece to enable heat flow, therefore, an epoxy is used to ensure the thermal contact between the hot plate/Cu plates and the chip. Next, the contact pads of the μTEG chip are wire bonded to the Cu traces on the PCB.

The heat sink adapter is the component used to direct the heat flow from the suspended microplatform to the heat sink. The preparation sequence of the heat sink adapter component is given in Fig. 4a. It includes a brass piece (8 × 8 mm², 500 μm thick) with four holes (each
with \(~510\,\mu\text{m} \) diameter) in which Cu wires were inserted. Diameter of the wires is compatible with the size of the suspended microplatforms they will contact after assembly. The height of the protruding wires is set to \(375\,\mu\text{m} \) leaving a \(125\,\mu\text{m} \) gap between the wire tip and the suspended microplatform when in place (see next paragraph). The tips of the wires are dipped in thermal paste, which according to our tests has a thickness between \(150\) and \(250\,\mu\text{m} \). The soft thermal paste fills the gap between the copper wires and the suspended platforms and ensures the thermal contact. Since the wire diameter is about half of the size of the suspended microplatform, we have enough clearance till the NWs area, since smearing paste over them will produce a thermal shortcut. Optical microscopy images of one of the wires dipped into thermal paste and of the test device after removing the heat sink can be seen in Fig. 4b and c, respectively. Even though squeezing-out is observed, the microplatform area is large enough to keep the droplet within its borders.

To prevent the heat sink from exerting an excessive force on the suspended microplatform through the heat sink adapter, a spacer is placed between the heat sink and the \(\mu\text{TEG} \). Since the heat should preferably flow from the suspended microplatform to the heat sink through the heat sink adapter, any direct heat flow between the bulk Si rim and heat sink through the spacer (and thus skipping the NWs) is to be minimized. For that purpose, polymethyl methacrylate (PMMA) is chosen as spacer material due to its low thermal conductivity of around \(0.19-0.22\ \text{Wm}^{-1}\text{K}^{-1} \) [20,21] and its easy and cheap fabrication. A laser cutter (Epilog Mini 24) is used to cut a \(500\,\mu\text{m} \) thick PMMA piece into an S-shaped spacer, which is also aligned with the rest of the components using the alignment pins. The thickness of the spacer is chosen so that it is larger than the Cu wires length (\(375\,\mu\text{m} \)) and smaller than the sum of the wires length and the minimum thickness of thermal paste (\(375\,\mu\text{m} + 150\,\mu\text{m} \)). An additional square shaped PMMA piece is cut to be used for the alignment of the heat sink adapter on top of the spacer.

The last step of the heat exchanger integration is to place the commercial aluminium (Al) heat sink on top of the heat sink adapter using a thermal paste to secure them in place with an appropriate thermal contact. The small heat sink has the dimensions of \(8 \times 8 \times 6\ \text{mm}^3 \). Fig. 4d shows the final assembly. With the ancillary pieces described and the integration proposed, the heat exchanger is in thermal contact with the fragile suspended platforms by means of a gentle physical attachment through the thermal paste at the tip of the wires, which also absorbs most of the vertical clearance uncertainties. We are currently working on reducing the manual steps and improving the integration scheme, but the described approach enables a first assessment of the performance improvement brought by the heat exchanger to a \(\mu\text{TEG} \) working in real operation conditions.

### 3. Results and discussions

#### 3.1. Simulations

During the planning of the route for the integration of a heat exchanger, simulations have been conducted to anticipate how thermal
and electrical performances may evolve. COMSOL Multiphysics 5.2 finite element analysis software is used for the simulations. The final physical model is sketched in real dimensions using CAD software and imported into COMSOL software.

The relevant parameters of thermoelectric active materials used for the simulations are listed in Table 1. Properties of the other materials (Cu, Si₃N₄, SiO₂, FR-4 and PMMA) are added from the material library of the simulation software. Simulating thousands of NWs bridging the trenches requires too many meshing elements and memory. To prevent this, an equivalent material is considered between the trenches, the properties of which are introduced by combining air and NW properties. The electrical and thermal conductivities of the equivalent material are chosen considering that the NWs occupy nearly 5% of the total trench area. Since these simulations aim at investigating the possible improvement brought by the heat exchanger, only the Si NWs based µTEGs case study will be shown in the next paragraphs. An electrical resistivity and a Seebeck coefficient akin to bulk highly doped Si have been chosen. A lower than bulk thermal conductivity, but yet a modest reduction, has been considered to account for nanostructuring effects when NWs are involved [22]. Those choices are compatible with the current understanding of electric and thermal properties of Si NWs of 100 nm of diameter.

In the first place, the temperature distribution in the overall system has been simulated using the heat transfer module of the simulation software. The boundary conditions applied to the model are setting the bottom surface to different temperatures, and to apply natural convection conditions on the exposed surfaces with an ambient temperature of 293 K. Two different cases have been simulated in the same conditions: with and without heat sink. For the device without heat sink, convective heat flux is applied on the bare suspended microplatform. For the assembly with heat exchanger, convective heat flux is applied on both vertical walls and horizontal plates of the heat sink fins.

Fig. 5 shows the temperature distribution maps when the hot plate temperature is set to 373 K. It can be seen that without heat exchanger a maximum ΔT of only 0.5 K drops across the NWs, out of a total of 80 K of temperature difference available between the hot plate and ambient. On the other hand, for the heat exchanger case, ΔT across the NWs increases to 23 K in the same conditions. In all the cases, the maximum ΔT is observed for T4 devices due to the larger effective length of NWs in such devices.

Next, the heat transfer module interface is coupled to the electric one to be able to extract the thermoelectric power from the µTEGs. Seebeck voltage (Voc) is determined from the obtained temperature distribution through the Seebeck coefficient given in Table 1. To extract the thermoelectric power, a sweep of the voltage is applied on the internal collector of each device (from 0 V to Voc), and the current values for each applied voltage are evaluated at the external collectors. Power density is then calculated directly from those values taking into account a device area of 2 mm². Current-voltage and power density curves for the cases with and without heat sink are shown in Fig. 5.

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**Fig. 3.** a) Schematic of the heat exchanger integration route including the components. b) Cross-sectional schematic of the assembly indicating the different components involved.
As anticipated, a higher thermal resistance is provided by longer NWs and hence larger ΔTs and higher Seebeck voltages are obtained for rising number of trenches. This is also the case when the heat sink is present although the relative impact of this parameter in Voc is much smaller. Considering thermopower, however, the higher NWs electric resistance may counterbalance the positive effect of more trenches in terms of Voc. When a heat sink is used, most of the attained ΔT is due to its presence rather than to the thermal properties of the Si NWs. This is why the detrimental electrical effect of having longer NWs is more determinant and is made evident for lower number of trenches when the heat exchanger is integrated. Indeed, for the particular case considered using the material properties shown in Table 1, the maximum power is obtained for the μTEG with 3 trenches (T3) when heat exchanger is present whereas the device with four trenches (T4) still yields the highest power output without the heat exchanger.

### 3.2. Measurements

For the measurements of μTEGs with and without heat exchanger, an additional control PCB is designed to connect the PCB of the assembly to the measurement units. Harvesting measurements are applied by placing the assembled devices on a Linkam HMS 350 V heating stage at different temperatures. Since the assembly includes a PMMA spacer layer, the temperature of the measurements is limited to 100 °C. No forced convection is applied, all the measurements have been made in a natural convection environment.

#### 3.2.1. Si NWs

Fig. 6 shows the Seebeck voltage vs hot plate temperature curves for the Si NWs-based μTEGs with different number of trenches. Three different cases are presented: without heat sink, with heat sink and with heat sink + pressing. For the case of heat sink + pressing, an additional force is applied on top of the assembly. This extra force helps to decrease the thickness of the thermal paste reducing the thermal resistance and consistently leads to a better performance in all cases. This application does not harm the suspended platform since the Cu wires cannot reach physically the suspended platform because of the PMMA spacer in between. As expected the devices without heat exchanger performed poorer (less voltage), and the more trenches the higher the voltage. For the two cases with heat sink (heat sink and heat sink + pressing), a tremendous improvement is observed, resulting from the much reduced thermal resistance between the cold side of the device and the ambient, thus leading to larger ΔTs across the NWs and to higher voltages. Ideally for devices with heat exchanger, the highest Seebeck voltage should also be obtained for T4 devices with the highest thermal resistance of the NWs, however, no clear trend of improvement

| Parameter                      | Si NW | Si bulk | W    | TIMa | air       |
|--------------------------------|-------|---------|------|------|-----------|
| Electrical Conductivity, σ (S m⁻¹) | 12000 | 12000   | 7 × 10⁶ | –    | –         |
| Thermal Conductivity, K (W m⁻¹K⁻¹) | 25    | 150     | 174  | 5    | 25 × 10⁻³ |
| Seebeck Coefficient, S (V K⁻¹) | 250 × 10⁻⁶ | 210 × 10⁻⁶ | 510⁻⁶ | –    | –         |

* Thermal interface material.
is observed from T1 to T4. In this case, different factors affecting the thermal contact between the components of the assembly may be the cause. One of the most significant of these effects is the thickness of the thermal paste. Since the thermal paste is applied manually on the Cu wires, the thickness of the thermal paste changes in the range of 150–250 μm, and hence may be not connecting equally on all four devices. This ends up in different thermal resistances at the interface and different ΔTs between the two ends of the NWs. As can be seen from Fig. 6d, T4 ended up in lower voltages compared to others. After the measurement, the setup was disassembled and the microplatform area covered by the thermal paste was indeed observed to be smaller for T4.

The maximum power densities obtained are 21.5 μW cm⁻² (T1), 14.3 μW cm⁻² (T2), 41.6 μW cm⁻² (T3) and 7.3 μW cm⁻² (T4) at 100 °C hot plate temperature. Comparing with the power densities without heat exchanger that are 0.061 μW cm⁻² (T1), 0.048 μW cm⁻² (T2), 0.087 μW cm⁻² (T3) and 0.063 μW cm⁻² (T4), a tremendous increase in power density is observed after the integration. The low performance of T4 device is not only due to its higher electric resistance, but also to aforementioned thermal contacting problems. The few selected parameters shown in Table 2 summarise the effect of the heat sink integration on the Seebeck voltage and the power density for Si NWs-based μTEGs. Results from devices with one (T1) and three trenches (T3) are presented for a hot plate temperature of 100 °C.

3.2.2. SiGe NWs

The abovementioned approach is repeated using the SiGe NWs based μTEGs. As can be seen from Fig. 7, significantly higher Seebeck voltages are observed in all cases for this material when compared to Si NWs. This is due to its lower intrinsic thermal conductivity that results in the higher thermal resistance of the NWs. In the case without heat exchanger, the obtained voltages scale with the number of devices as expected. This is also true for the cases with heat sink integration to the extent of the data available (plots are given for the devices T1 to T3, since T4 was not functional before the integration of the assembly). Adding the heat sink improved significantly the performance of the μTEG, which improved further for the case of heat sink + pressing, both in voltage and power. Power curves of the T3 device with different heat sink cases are presented in Fig. 7d, and a maximum power output close to 1 μW is obtained when the hotplate temperature is kept at 100 °C.

In more detail, power densities obtained from the SiGe NWs-based devices are increased from 0.032 μW cm⁻² to 1.4 μW cm⁻² (T1), 0.05 μW cm⁻² to 4.5 μW cm⁻² (T2) and 0.073 μW cm⁻² to 18.9 μW cm⁻² (T3) with the integration of a heat exchanger at the hot plate temperature is set to 100 °C.
3.2.3. Si microbeams

As pointed out before, μTEGs with microbeams were also designed and fabricated to be able to compare the performances of micron-sized Si beams with the Si NWs. Different number of beams were fabricated inside the (single) trench area yielding different area occupancies, which in the end determine the thermoelectric power output. From the volumetric shape of the beams defined after the anisotropic KOH etch process, final trench occupancies for Si beams can be calculated. Power curves obtained from the device with an area occupation of 11% are given in Fig. 8. A dramatic three orders of magnitude increase is obtained in the power output (from $\sim 650$ pW to $\sim 690$ nW for a single thermocouple, i.e. from $32.5$ nW cm$^{-2}$ to $34.5$ μW cm$^{-2}$) by integrating a heat sink on the μTEG and applying a small amount of force on top of the assembly. In this way, it is shown that even with high thermal conductivity Si microbeams, it is possible to obtain high power densities by optimizing its area occupation and electrical properties when a heat sink, securing a good enough $\Delta T$, is present. An additional design including lower area occupancies of Si microbeams is under development.

4. Conclusions

In this study, harvesting measurements have been performed on Si NWs, SiGe NWs and Si microbeams based μTEGs including an integrated heat exchanger. The integration route for the heat exchanger took into account the fragility of the suspended platforms of the μTEGs. The assembly was first simulated and then tested. A significant improvement has been observed for all tested μTEGs based on the different thermoelectric materials considered. The power densities with the heat exchanger integrated assembly are 50–1000 times larger than for

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Table 2

|                | T1                  | T3                  |
|----------------|---------------------|---------------------|
| Voc (mV)       | PD (μW·cm$^{-2}$)   | Voc (mV)            | PD (μW·cm$^{-2}$) |
| Without heat sink | 0.12               | 0.061               | 0.16               | 0.087               |
| With heat sink  | 1.46               | 10.4                | 2.07               | 12.5                |
| With heat sink + pressing | 2.27               | 21.5                | 4.21               | 41.6                |

Fig. 6. Seebeck voltage vs hot plate temperature for Si NWs-based μTEGs with different number of trenches (T1-T4). Figures are presented for three different cases: without heat sink, with heat sink and with heat sink + pressing.
Fig. 7. a)-c) Seebeck voltage vs hot plate temperature graphs for SiGe NWs based μTEGs with different number of trenches (T1-T3). Figures are presented for three different cases: without heat sink, with heat sink and with heat sink + pressing. d) Power curves obtained from T3 for those three different cases at a hot plate temperature of 100 °C.

Fig. 8. I-V and power curves obtained from the Si microbeams based μTEG with the beam area occupation of 11%. Measurements are conducted a) without and b) with heat exchanger at a hot plate temperature of 100 °C and a three-order magnitude increase is obtained.
similar devices without heat exchanger at the same hot plate temperature. Maximum power densities obtained from heat exchanger integrated µTEGs based on NWs are 41.2 μW cm⁻² (Si NWs) and 45.2 μW cm⁻² (SiGe NWs) at a hot plate temperature of 100 °C. From µTEGs based on micro sized Si beams we observed 34.5 μW cm⁻² for the same hot plate temperature of 100 °C. The measured power densities from all three types of devices are adequate for supplying energy to low power wireless sensor nodes. After experimental confirmation of the tremendous improvement brought by the placement of a heat exchanger, efforts are now focused on the optimization of the integration route and selecting high temperature stable components to be able to make measurements, and to operate, at higher temperatures.

CRediT authorship contribution statement

Iinci Donmez Noyan: Conceptualization, Formal analysis, Investigation, Methodology, Software, Writing - original draft.
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Marc Salleras: Conceptualization, Formal analysis, Investigation, Methodology, Software, Supervision, Validation.
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Albert Tarancón: Conceptualization, Formal analysis.
Luis Fonseca: Conceptualization, Formal analysis, Funding acquisition, Methodology, Project administration, Resources, Supervision, Validation, Writing - review & editing.

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