Miniaturized Radioisotope Solid State Power Sources

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Abstract. Electrical power requirements for the next generation of deep space missions cover a wide range from the kilowatt to the milliwatt. Several of these missions call for the development of compact, low weight, long life, rugged power sources capable of delivering a few milliwatts up to a couple of watts while operating in harsh environments. Advanced solid state thermoelectric microdevices combined with radioisotope heat sources and energy storage devices such as capacitors are ideally suited for these applications. By making use of macroscopic film technology, microgenerators operating across relatively small temperature differences can be conceptualized for a variety of high heat flux or low heat flux heat source configurations. Moreover, by shrinking the size of the thermoelements and increasing their number to several thousands in a single structure, these devices can generate high voltages even at low power outputs that are more compatible with electronic components. Because the miniaturization of state-of-the-art thermoelectric module technology based on Bi₂Te₃ alloys is limited due to mechanical and manufacturing constraints, we are developing novel microdevices using integrated-circuit type fabrication processes, electrochemical deposition techniques and high thermal conductivity substrate materials. One power source concept is based on several thermoelectric microgenerator modules that are tightly integrated with a 1.1W Radioisotope Heater Unit. Such a system could deliver up to 50mW of electrical power in a small lightweight package of approximately 50 to 60g and 30cm³. An even higher degree of miniaturization and high specific power values (mW/mm³) can be obtained when considering the potential use of radioisotope materials for an alpha-voltaic or a hybrid thermoelectric/alpha-voltaic power source. Some of the technical challenges associated with these concepts are discussed in this paper.

INTRODUCTION

Deep space missions have a strong need for compact, high power density, reliable and long life electrical power generation and storage under extreme temperature conditions. Conventional power generating devices become inefficient at very low temperatures (temperatures lower than 200K encountered during Mars missions for example) and rechargeable energy storage devices cannot be operated thereby limiting mission duration. At elevated temperatures (for example temperatures of 600K and higher for solar probe or Venus lander) thin film interdiffusion destroy electronic devices used for generating and storing power. Solar power generation strongly depends upon the light intensity, which falls rapidly in deep interplanetary missions (beyond 5 a.u.) or in planetary missions in the sun shadow. Moreover, it has been observed during the Mars pathfinder mission that significant performance degradation occurred when solar cells get covered with dust particles. Radioisotope thermoelectric generators (RTGs) have been successfully used for a number of deep space missions RTGs. However, their energy conversion efficiency and specific power characteristics are quite low, and this technology has been limited to relatively large systems (more than 100W).

The National Aeronautics and Space Administration (NASA) and the Jet Propulsion Laboratory (JPL) have been planning the use of much smaller spacecrafts that will incorporate a variety of microdevices and miniature vehicles such as microdetectors, microsensors and microrovers. Except for electrochemical batteries and solar cells, there are currently no available miniaturized power sources. Novel technologies that will function reliably over a long duration mission (ten years and over), in harsh environments (temperature, pressure, and atmosphere) must be developed to enable the success of future space missions. It is also expected that such micro power sources could have a wide range of terrestrial applications, in particular when the limited lifetime and environmental limitations of batteries are key factors. The first step towards such compact power sources consists of a miniaturized, versatile

milliwatt power source (MWPS) device that is currently being developed. The original MWPS concept was advanced by engineers at JPL (Chmielewski and Ewell, 1994) and consists of a 1.1 W Radioisotope Heating Unit (RHU) to provide the high temperature source for a thermoelectric converter which generates sufficient electrical power (~40 mW) to trickle-charge a rechargeable battery pack. The battery power can then be used in low duty cycle, low power applications. The MWPS approach is based on several technologies developed earlier by space power programs. The RHU was developed by the Department of Energy for the Galileo and Ulysses missions, the thermoelectric converter, or thermopile, is a combination of the early RTG technology, pace maker technology with new packaging techniques, and a lithium-ion rechargeable space battery being developed at JPL, under a separate ongoing program. The prototype MHPS, illustrated in Figure 1, is 67 mm in diameter, 81 mm long, and weighs about 0.41 kg.



FIGURE 1. Milliwatt Power Source (MHPS) concept incorporating a radioisotope heater unit (RHU), a thermoelectric converter (TEC) and rechargeable batteries (not shown).

In collaboration with industry, JPL is now pursuing the fabrication and testing of MHPS. This effort is described in more details elsewhere (Bass, 1998; Borshchevsky et al., 1997). However, the MHPS will still be too bulky for several next generation miniature "sciencecrafts" and a couple of new approaches to ultra-compact solid state power sources is presented here.

THERMOELECTRIC GENERATORS

Solid state thermoelectric generators covering a wide range of power outputs, from nanowatts to kilowatts, have demonstrated attractive characteristics such as long life, the absence of moving parts or emissions, low maintenance and high reliability. In spite of a large number of potential civilian and military applications, their use has been severely limited due to their relatively low energy conversion efficiency and high development costs. As a consequence more efficient advanced radioisotope power systems (ARPS) are now being developed, based on competing thermal-to-electric technologies. One approach to higher performance thermoelectric devices and systems is the discovery and infusion of novel thermoelectric materials more efficient above room temperature than the current state-of-the-art Bi_2Te_3 , PbTe or SiGe alloys. Recent results in several laboratories have successfully identified superior materials in several temperature ranges (Fleurial et al, 1996; Sales et al., 1996; Caillat et al., 1997). There is currently an effort to introduce some of these new compounds into simple unicouple configurations to demonstrate increased conversion efficiency of up to 20% (Caillat et al, 1999).

A second approach is to significantly improve the design, specific power (watts per unit area or volume) and lower the costs of generator devices even when using state-of-the-art thermoelectric materials. This is of great interest when considering large-scale applications using waste heat recovery schemes, or low power devices integrated with electronics and optoelectronics components. For both aerospace and terrestrial applications, there is a growing need for developing miniaturized on-chip low power batteries with long life, high voltage, resistance to extreme temperatures and low environmental impact characteristics (Rowe, 1995; Fleurial et al., 1997). Figures 2.a and 2.b illustrate the potential of miniaturized thermoelectric power sources based on state-of-the-art Bi₂Te₃ alloys. The calculations show that in spite of the low conversion efficiency, high specific power values (in W/cm³) can be achieved even for relatively small temperature differentials, in the 20 to 200K. This is due to the fact that thermoelectric devices are scalable, provided that the aspect ratio of the thermoelements (or legs) is kept constant and that electrical and thermal resistance loss can be kept low. Thus miniaturized thermoelectric devices with leg size in the tens of microns range (instead of tens of millimeters) are very attractive.

However, current thermoelectric module technology is ill suited to such development due to mechanical and manufacturing constraints for thermoelement dimensions (100-200µm thick minimum) and number (100-200 legs maximum). In addition to the widespread use of semi-manual assembly techniques that results in high costs for more compact configurations, these devices have typically undesirable high current and low voltage characteristics. Much smaller devices capable of high voltage (up to 5V) power output in the nW to tens of μ W range have also been developed: monolithic structures and more recently thin film devices. Most of the monolithic module configurations have been used in nuclear battery type devices, operating across large temperature differences (100-200K), with a small amount of radioisotope material (usually PuO_2) as the heat source (Rowe, 1995; Bass, 1998). The specific power density of the monolithic thermopiles is typically measured in tens of mW/cm³, but falls to about $60 \text{ }\mu\text{W/cm}^3$ when taking into account the complete power source package. Thin film devices producing 20 mW at 4V under load with a temperature difference of 20K have been recently described (Stordeur and Stark, 1997). The 0.22 cm^3 device is comprised of 2250 thermocouples deposited on Kapton thin foils packed together and was fabricated using integrated circuit-type techniques. However, in spite of this remarkable achievement that could allow for batch fabrication of these devices, the specific power density still remains quite low, close to 90 μ W/cm³ (heat source not included). This is mainly due to the fact that the length of the thermoelectric legs is supported by the Kapton substrate, thus introducing a very significant thermal shunt and dramatically degrading conversion efficiency.



FIGURE 2. (a) Improvement of thermoelectric generator performance when using more efficient materials. The temperature differences reported here are relevant to state-of-the-art Bi_2Te_3 alloys. (b) Calculated increase in specific power (per unit volume) with increasing leg miniaturization (constant cross section to length leg aspect ratio) and increasing temperature differences of operation. Data are shown for both low (alumina, blue dots) and high (diamond, red lines) thermal conductivity substrates.

MINIATURIZED THERMOELECTRIC GENERATOR DEVICES

To circumvent key shortcomings described in the preceding section, the Jet Propulsion Laboratory (JPL) is pursuing the development of vertically integrated thermoelectric microdevices that can be fabricated using a combination of thick film electrochemical (ECD) and integrated circuit (IC) processing techniques (Fleurial et al., 1998). Indeed, current prototype devices leave much room for performance improvement, as illustrated in Figure 2b. Even for relatively small temperature differences, such as 10 to 20K, high specific power outputs in the 1 to 10 W/cm³ are potentially achievable provided that the legs be no thicker than 50 to100 μ m.

Microdevice configuration

The term "vertically integrated" here refers to the conventional thermoelectric module configuration shown in Figure 3. This design eliminates the large heat losses observed in planar thin film thermoelectric devices where the

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legs are deposited onto a supporting substrate. However, planar configurations do offer a very convenient way of fabricating electrical interconnects between the thin film legs by using traditional masking techniques. Thermal resistances due to heat transfer through the metallizations and substrates, as well as electrical resistances due to the interconnects between n-type and p-type thermoelectric legs, rapidly become important issues when increasing device miniaturization. High thermal conductivity substrates, thin metallizations and intimate contact with the heat source and heat sink media are key to minimizing thermal issues when the microgenerators operate in particular under low temperature differences and high heat flux conditions. Since high voltage power outputs are highly desirable from a power conditioning aspect, this means that the microdevices will typically possess several thousands of very short thermocouples. Electrical contact resistances can thus easily become a very large fraction of the total internal device resistance. However, low values are routinely obtained in the electronic semiconductor industry and similar processing techniques have been developed here. Finally thermally stable diffusion barriers are needed to maintain the integrity of the multilayered stack of substrates, metallic interconnects and thermocouples. The effectiveness of amorphous transition metal nitride diffusion barriers for metallizations on diamond, AlN and thermally oxidized silicon substrates has been recently demonstrated (Kacsich et al., 1998).



FIGURE 3. Schematic representation of a vertically integrated thick film thermoelectric generator using thin high thermal conductivity substrates.

Microdevice fabrication

Hot side temperatures for microdevice applications that we are currently considering are 200 to 500K. $Bi_{2-x}Sb_xTe_{3-y}Se_y$ alloys are the state-of-the-art materials best suited to these temperatures of operation. Since the thickness of the legs selected in our various device concepts ranges from 10 to 60µm, we have actively pursued the development of an electrochemical thick film deposition process. ECD constitutes an inexpensive way to synthesize semiconducting films (Pandey et al., 1996) and, depending on the current density used in deposition, the deposition potential or solution concentration may possibly be used to induce off-stoichiometric films, thus providing p- or n-type doping through stoichiometric deviation. The electrodeposition of thermoelectric materials has not been widely investigated (Muraki and Rowe, 1991; Takahashi et al., 1994) and new experimental methods have been developed to obtain p-type and n-type $Bi_{2-x}Sb_xTe_{3-y}Se_y$ compositions which are optimal for thermoelectric power generation in the temperature range of interest. An additional advantage of ECD is that some of the interconnect layers necessary to the fabrication of these devices, such as Cu for the electrical path or Ni for the Cu diffusion barrier can also be deposited by using different aqueous solutions. More details have been reported elsewhere (Fleurial et al., 1998).

Building on the availability of new thick photoresist commercial products, we have developed templates suitable to the electrochemical deposition of legs as thick as 70µm and as small as 6µm in diameter. Actually, it has been determined that to be able to tightly control the geometry of the legs and prevent "mushrooming" growth, electrodeposition must be conducted in equally thick photoresist templates. The thick positive photoresist template is patterned with deep square or round shaped holes that must be pre-aligned on top of metallic interconnects.

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Figures 4.a and 4.b illustrate the result of IC-type processing. More processing steps are required to successively deposit n- and p-type legs on top of the bottom substrate interconnects, and then ensure proper joining to a top substrate with similarly patterned interconnects. Based on commercial electrolytes, we have used ECD techniques to deposit high quality Cu, Ni and Pb-Sn solder layers as well. The Pb-Sn layer can be used to form solder bumps on top of the legs, as done for flip-chip bonding (Annala et al., 1997). These processing steps are illustrated in Figure 5. The combination of ECD and IC-type techniques offers a degree of flexibility in designing and fabricating microdevices. Typically, a single photolithography mask can combine all of the necessary patterns to completely fabricate one generator configuration.



(a)

(b)

FIGURE 4. (a) Thick positive photoresist template on top of a metallized Si/SiO_2 substrate. Deep cylindrical holes where the thermoelectric leg will be deposited can be seen. (b) Cu metallization on top of a Si/SiO_2 substrate where interconnects have been patterned for subsequent deposition of the thick photoresist template and thermoelectric legs. The fully metallized square pads are for providing electrical contact tests.



FIGURE 5. (a) Bi_2Te_3 legs electrodeposited on top of Cu interconnect metallizations (using a Si/SiO₂ substrate) and (b) $Ni/Bi_2Te_3/Ni/Pb$ -Sn legs electrodeposited on top of Cu interconnects.

Advanced Milliwatt Power Source

This concept proposes to integrate highly miniaturized thin film thermoelectric converters with a single RHU into a lightweight, rugged, long life, compact and modular solid-state power source capable of delivering tens of milliwatts. A schematic of the complete advanced milliwatt power source (AMWPS) is shown in Figure 6.a. Figure 6.b illustrates how the spherical power source could be integrated with next generation microrovers designed for planetary and asteroid missions.



FIGURE 6. (a) Center cut view of an advanced milliwatt power source (AMWPS) concept based on a single RHU thermal source and six thermoelectric thin film microgenerators (TEMGs). The schematic shows both the external shell and internal jacket for the RHU to allow for the bonding of planar thermoelectric microdevices. (b) Illustration of an AMWPS device integrated with a next generation microrover.

The RHU unit is a cylinder 32mm high and 26mm in diameter and weighs about 40g. The palletized fuel is surrounded by a platinum-alloy capsule, pyrolitic graphite thermal insulation and a graphite ablation shell. Based on the 1.1W RHU thermal output and thermoelectric microgenerators fabricated with thick films of Bi_2Te_3 alloys and operating across a 250K to 300K temperature difference, the novel power sources could produce up to 50mW. The total weight of the MWPS device is predicted to be close to 60g, compared to the 350-400g weight of the MWPS based on bulk thermopiles. Energy storage devices such as capacitors could also be used in combination with this power output to deliver much higher energy levels at brief time intervals (for example 0.5W at 10% duty cycle). Technical issues associated with this concept include in particular the development of an external jacket for the RHU to allow for the bonding of planar thermoelectric microdevices and that is compatible with RHU atmospheric re-entry requirements. In addition thermal and mechanical studies must be conducted to minimize heat losses and maximize mechanical ruggedness.

ALPHA-VOLTAICS

Another attractive solid state power generator device investigated at JPL is based on the direct conversion of the kinetic energy of alpha particles into electricity (Patel, 1999). This device is expected to exhibit a high conversion efficiency (over 14%) and to function continuously over a long period of time in the temperature range of 20 to 800K without any recharging needs or the presence of any sunlight thanks to a unique long life design. The use of alpha particle kinetic energy for conversion into electricity using an existing SiC photodiode was reported earlier (Rybicki et al., 1996). This particular device was found to have degraded in a rapid manner because it was not specifically designed to avoid the crystal damage from implanted alpha particles. Results on similar devices using beta and gamma rays were also published (Olsen, 1974), but these power sources exhibited extremely low conversion efficiency (0.1 to 4%) and required substantial shielding to reduce the dose from its radiation to adjacent electronics. Such devices also had a relatively short lifetime (2-6 years) and delivered very low power levels (a few milliwatts).

The device design pursued by JPL is aiming at minimizing lattice damage from alpha particles in the active semiconductor p-n junction, as illustrated in Figure 7. The key design feature of the technology lies in the determination of the diode dimensions so that alpha particles with energy of 5.8 to 6.1 MeV do not stop in the active device volume but in the inactive substrate layers. Alpha particles cause severe lattice damage when they stop since they lose a large fraction of their kinetic energy just before stopping in materials. Nevertheless, some lattice damage is expected in the active p-n junction, but it has been observed that such damage is continuously annealed during the ionization process in semiconductors. Semiconductors such as GaAs or SiC that are stable at high temperatures will be used in the fabrication of the alpha particle-based power source. Curium-244, a nearly pure alpha particle source with negligible soft gamma emission and an 18-year half-life has been initially selected as the

radioisotope material. Initial studies indicate that optimally configured miniaturized alpha voltaic power sources could offer high specific power values close to a milliwatt per cubic millimeter (mW/mm³).



FIGURE 7. Schematic representation of an alpha-voltaic power generator device. The high-energy alpha particles emitted by the radioisotope thin film power source cross the thin depleted GaAs p-n junction, creating electron-hole pairs that are collected at the metallic contacts. Most of the lattice damage from the alpha particle kinetic energy occurs in the inactive substrate layers.

CONCLUSION

Some new NASA missions, such as asteroid rovers, Europa cryobots (link modules), or the Mars Network, are designed for deep space, cold planetary environments and call for milliwatt to watt power source requirements. Current electrical power source and storage technology is either too bulky, limited to a narrow temperature range of operation, or dependent on sunlight intensity. Radioisotope solid state energy conversion devices offer attractive possibilities for configuring rugged, lightweight, long life, modular power sources that could potentially achieve high specific power values in the mW/mm³ range. Thermoelectric microgenerators based on state-of-the-art materials could be combined with RHUs, or even other more compact advanced fuel packages, to deliver tens of milliwatts. However, that technology to date is limited to bulky configurations based on monolithic thermopiles or to inefficient planar thin film devices. To fabricate high performance microdevices in a "classic" vertically integrated module configuration, a combination of electrochemical deposition techniques and integrated circuit technology is now under development. Thick photoresist templates up to 70 µm have been successfully developed and patterned using conventional UV photolithography, resulting in the reproducible fabrication of highly packed arrays of thousands of legs as small as 6µm in diameter. We are now focusing on the fabrication of operational prototype devices with high specific power, high voltage characteristics that in particular could operate in a 200 to 500K temperature range. Direct conversion of the kinetic energy of alpha particles into electricity is also a very attractive technology. JPL is currently focusing on the study of simple p-n junction devices based on wide band gap materials such as GaAs that combine high conversion efficiency and long lifetime. The key challenge is designing the alpha-voltaic device to minimize lattice damage from the alpha particles. Because of their complementary energy conversion processes, it might even be possible to fabricate hybrid radioisotope alpha-voltaic/thermoelectric microgenerators, provided than the alpha-voltaic device can operate at elevated temperatures. If such technologies can be successfully developed, it is likely that they will be introduced in many terrestrial applications

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