Development of Segmented Thermoelectric Multicouple Converter Technology

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Abstract—The Jet Propulsion Laboratory (JPL), Pratt & Whitney Rocketdyne, and Teledyne Energy Systems, Inc., have teamed together under JPL leadership to develop the next generation of advanced thermoelectric space reactor power conversion systems. The program goals are to develop the technologies needed to achieve a space nuclear power system specific mass goal of less than 30 kg/kW at the 100 kW power level with a greater than 15 year lifetime. The technologies required for such a power system include liquid metal cooled reactors with outlet temperatures ranging from 1125 K up to 1325 K, segmented thermoelectric multicouple converter (STMC) arrays which can achieve greater than 8 percent system efficiency and carbon-carbon heat pipe radiator panels to reduce the radiator subsystem specific mass to a goal of 5 kg/m^2. The STMC Program's development efforts focused on a highly compact conductively coupled modular thermoelectric converter assembly (TCA) design. STMC design efforts were based on a multicouple design similar to the SP-100 Program's design but using segmented thermoelectric (TE) legs rather than the single alloy silicon-germanium legs. Efforts have addressed in parallel the selection and optimization of the most promising high temperature thermoelectric materials, the development of the various STMC components and sub-assemblies, design, analysis, fabrication and assembly of subscale STMC devices as well as scale-up plans to the 100 kW-class power level. The performance of the selected high temperature TE materials and initial thermal, electrical and mechanical test results on several STMC demonstration devices are reported.

Keywords: Enter Keywords here.
PACS: 84.60.Rb; 85.80.Fi; 89.30.Gg

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INTRODUCTION

Recently, under NASA's Project Prometheus, the Nuclear Systems Program, the Segmented Thermoelectric Multicouple Converter (STMC) Development Program, the Jet Propulsion Laboratory (JPL) has led a technical team in developing a new class of thermoelectric materials and technologies for application to high power nuclear electric propulsion (NEP) applications [Johnson 2004]. The primary focus of the STMC Program over the past year has been the development and fabrication of the lower temperature multicouple stage of the STMC converter using a conductively coupled generator design concept and skutterudite thermoelectric materials. Pratt & Whitney Rocketdyne, with Teledyne Energy Systems, Inc., has been under contract to JPL to provide systems integration support and converter fabrication expertise for the preparation of materials, components, subassemblies and fabrication of tooling for assembling and bonding the low temperature thermoelectric multicouple converter (LT-TMC). JPL's efforts have also included identification of the potential upper stage thermoelectric materials; a task supported by more than a half-dozen U.S. university research laboratories.

Prior studies have indicated that a high power, high temperature thermoelectric power converter is mass competitive with dynamic conversion technologies for such applications. More efficient thermoelectric materials are predicted to allow for lower converter operating temperatures and thus lower risks for the nuclear heat source development. However, several critical technical developments need to be completed to enable this new technology. The three highest near term priorities addressed by the STMC program were: a) Demonstration that a high performance 2x2 couple array mini-TCA based exclusively on skutterudite materials can be fabricated and operated, maintaining complete, low resistance bonds at all material interfaces and acceptably small parasitic temperature drops across non-performing layers; b) completion of a stress

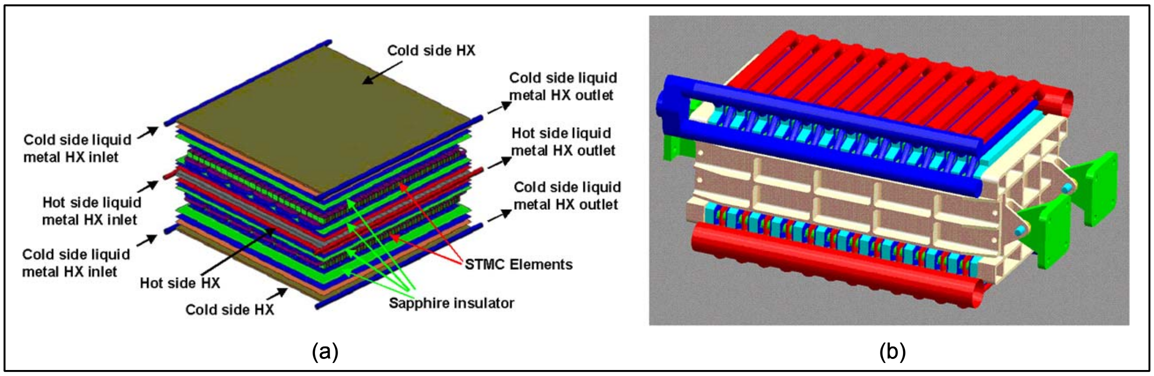

FIGURE 1 - Schematics showing the modular structure of the thermoelectric power conversion subsystem: (a) thermoelectric converter assembly (TCA) with thermoelectric couple arrays conductively coupled to a central hot liquid metal heat exchanger and two cold side liquid metal heat exchangers located on both external sides; TCAs are then stacked into a power converter assembly with manifolds for distribution of the hot and cold liquid metal into each TCA.

analysis, incorporating measured material properties, for a full power conversion assembly (PCA), consistent with unit survival and acceptable margin during both fabrication and temperature sublimation rates of the new thermoelectric material options, and the associated mitigation strategies, to confirm the viability of long term device operation.

CONCEPTUAL POWER CONVERSION SYSTEM DESIGN

In the first phase of the STMC Program, the system design efforts had been focused on radiatively-coupled power conversion subsystem design concepts. While somewhat reducing risks in converter technology development, all of these concepts appeared to be too high in mass (i.e., 4200 to 4600 kg) at the 100 kWe power level to make NEP missions a practical alternative to chemical propulsion. At the beginning of Phase II, conceptual system design efforts were focused by JPL on a close packed array thermoelectric converter design. A liquid metal heat transport loop was used to transfer thermal power from the reactor outlet directly to the hot shoes of the segmented thermoelectric couple array bonded to a thermoelectric converter assembly (TCA) heat exchanger configuration similar to the SP-100 Program's power conversion approach, as shown in Figure 1a [Mondt 1990]. A series of 12 TCAs were formed into a modular power conversion assembly or PCA (as shown in Figure 1b).

This change significantly reduced the power conversion assembly and the heat transport loop masses and their thermal losses. A secondary liquid metal heat transport loop was used to transfer the waste heat from the PCA to a radiator heat pipe panel equipped with a liquid metal manifold and heat pipes. The working fluid for the radiator heat pipes was changed from potassium to cesium to provide higher heat transport capability at lower evaporator temperatures. This allowed the radiator heat pipe design to be optimized to a lower specific mass value (~5 kg/m2) than in the previous directly coupled converter/radiator panel design. It also allowed the application of the SP-100 Advanced Radiator Program's heat pipe technology to minimize the overall radiator panels specific mass. The temperature utilization factor of the conductively coupled TCA/PCA design improved the average thermal to electric conversion efficiency from 7.7% to 8.7% while reducing the heat source supply temperature by 70 K(from 1400 K to 1330 K) and its mass by 15%. Finally, using an even lower reactor coolant outlet temperature (1230 K vs. 1330 K) to reduce reactor heat source development risks and costs, the segmented thermoelectric couple's leg lengths were re-optimized to provide 7.1% TCA conversion efficiency with a 100 K lower temperature differential across the thermoelectric stack. C-103 alloy could be used as the structural alloy for the reactor and its primary heat transport subsystems components at these temperatures. Only segmented thermoelectric technology can provide this design flexibility in heat source temperature accommodation and still provide high efficiency output from its converter. A power system specific mass of 39 to 40 kg/kWe was estimated at the 100 kWe power level for this design concept based on liquid lithium heat transfer loop technologies. The STMC power system conceptual design efforts have resulted in a workable and flexible design solution that will maximize the specific power output of a space reactor thermoelectric power conversion system for NEP-type missions.

THERMOELECTRIC CONVERTER DESIGN

The converter design effort focused on developing candidate "conductively coupled" design approaches for the STMC based on lessons learned from SP-100 developed technologies [Mondt 1994]. In particular, the STMC design early on considered how to ensure that the converter survives fabrication and assembly stresses as well as minimize stress under steady-sate operation to enable long life. Our approach was to redistribute thermally induced stresses by selecting optimal materials combinations and element geometries combined with simplifying and streamlining fabrication steps. To that effect a rapid STMC configuration evaluation tool was developed using an elastic model of the stack of material components that allows comparing trends and identifying key risk areas. Early in Phase 2, an initial evaluation of thermal/mechanical stress environment for block of TCAs installed within one PCA was performed and used as boundary conditions. Then a thermal stress and loads analysis model of the STMC TCA was prepared in order to determine the dimensional compliance requirements for the STMC stack assembly and address issues identified during the SP-100 technology development program. A secondary objective of the design was to minimize parasitic losses (ΔT across non-TE layers and fill factor thermal losses), from about 30% experimentally determined for the SP-100 multicouple to 10% or less. This parasitic loss directly impacts the overall system mass and performance by requiring lower heat rejection temperature or higher heat source operating temperature.

STMC layout configurations having the best potential for meeting manufacturability, performance, mission and life requirements were developed. Trades between a 1-D or a 2-D array of couples for the STMC layout configurations and between fused glass or aerogel for the leg-to-leg thermal and electrical packaging were performed. Initial calculations performed on both the SP-100 multicouple and STMC module using elastic models indicated that the stress levels were comparable and significantly exceeded the elastic limit, especially after the fully assembled devices were cooled down to room temperature following high temperature assembly. Subsequently, a key concept was to design the STMC stack for minimum stress under planned in-gradient operating conditions, by using materials with higher coefficient of thermal expansion (CTE) values on the cold side to match to the hot side heat exchanger material Nb-1Zr, and high voltage insulator material, sapphire. The best match was obtained by selecting stainless steel 304L for the heat exchanger and beryllia for the high voltage insulator. Half of the STMC stack is illustrated in Figure 2a, while a schematic representation of an aerogel filled STMC module is shown in Figure 2b. After accounting for the new material selection, lower temperature fabrication steps and actual grading of the materials interfaces, lower stress level, within the elastic limit, were predicted for the first time. The validity of these predictions is expected to improve upon completion of the high temperature material database and analysis of results on bond coupons and structural tests of the STMC stack.

SEGMENTED THERMOELECTRIC MULTICOUPLE DEVELOPMENT

Development of the STMC technology was initially split into two parallel efforts: a) characterization, development and selection of the best thermoelectric materials for the high temperature segments among several candidate compounds and alloys for which high ZT values have been reported by various experimental groups and b) will development of a sub-scale fully functional thermoelectric module based on the high performance skutterudite materials already available for the 975 to 750K temperature range After completion of these two activities, the development of high temperature STMC modules was to take place using the set of n-type and p-type thermoelectric materials selected earlier. Results on the first two parallel activities are reported.

Selection of High Temperature Thermoelectric Materials

The efficiency of a thermoelectric power generating device composed of an array of n-type and p-type thermoelectric material elements operating over a large temperature differential is directly linked to the average value of ZT over this temperature range, where T is the absolute temperature and Z is a figure of merit that is given by the relationship:

Z = (Seebeck coefficient)^2/(thermal conductivity x electrical resistivity) = $\alpha^2/(\lambda\rho)$.

The value of ZT for various TE materials typically varies considerably as a function of temperature, and each TE material has a temperature range over which ZT is near the maximum. The STMC projected operating range was 1275 K to 750 K, and maximizing conversion efficiency could be best achieved by constructing each thermocouple leg with TE material segments so that each TE material segment in each leg functions within a temperature range at which its ZT is near-optimum.

TE skutterudite materials that perform well up to 975 K have already been developed by JPL to TRL ~3-4 [Caillat 2005]. Other than Si-Ge alloys, TE materials that perform well in the high-temperature range from ~ 975 K to 1275 K have not been demonstrated. STMC performance goals (8-10% conversion efficiency at the converter level) required that the average ZT in the 1275 – 750 K temperature range be at least 0.85, with an ultimate goal of 1.2.

However, integrating Si-Ge alloys in a segmented configuration was predicted to lead to significant penalties in device efficiency due to poor compatibility of their thermal, electrical and mechanical properties with those of most other TE materials and in particular the low-temperature skutterudites segment [Ursell 2002]. In addition, p-type Si-Ge has much lower ZT values, about 0.6, and would significantly impact the STMC performance. Prior to the beginning of the STMC Phase 2, several families of compatible high temperature materials were identified by JPL and various other researchers. To minimize Phase 2 technical risks within the two year initial schedule, a combined effort from a multi-university and JPL team of experts (Table 1) was initiated. This multi-pronged effort focused on synthesis of TE samples while most of the

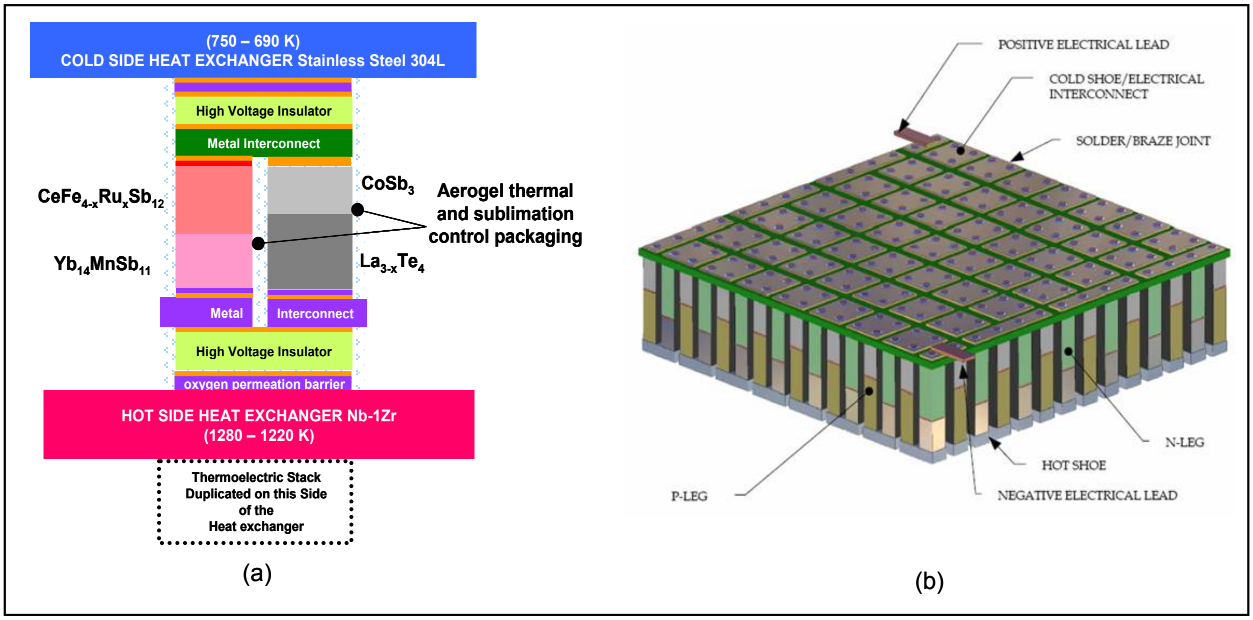

Figure 2 - Schematic illustrations of (a) the STMC stack of electrical insulators, electrical interconnects and thermoelectric material segments with the aerogel thermal packging and (b) a conceptual design for a 8x14 segmented couple module using a series-parallel laddering architecture.

Table 1 - Results obtained on TE material candidates at elevated temperatures reported by early 2004. Key material development needed and participating institutions in the STMC program are described for each material family.

Material	ZT_{max} reported	Development Needs addressed during STMC Phase 2	Participant
n-La_2Te_3	1.3 at 1275K	Reproduce synthesis, zT Determine stability	Michigan State Princeton
Low Temperature Skutterudites	0.8 to 1.2 at 950K	Optimize composition for incremental improvement to average ZT=1.2 in 975-750K	JPL, Caltech University Michigan
High Temperature Skutterudites	0.6 at 1000K	High temp measurements Optimize ZT, determine temperature stability in vacuum	JPL University Michigan
p-Chevrel Phases	0.6 to 0.8 at 1275K	Optimize composition, stability from diffusion, sublimation	Cornell JPL
p-Zintl Phases	0.6 at 675K, estimated 1.0 at 1000K	High temp measurements, thermal stability	UC Davis JPL
n-Half Heusler	1.0 to 1.4 at 1075K	High temp measurements, thermal stability	Clemson, U. Va
n-, p-Clathrates	0.9 at 875K	High temp measurements, thermal stability	U. South Fla.

high temperature TE characterization was conducted at JPL to ensure data consistency and timely comparison between material candidates.

n-Type Materials—Attractive results were obtained during the 11 month TE material screening phase on samples measured at JPL Candidate n-type materials included $Co_{1-x}Ir_xSb_3$ (skutterudites), TiNiSn-based half-heusler alloys, $Ba_8Ga_{18}(Ge,Si)_{30}$ clathrates, and rare earth chalcogenides such as $La_{3-x}Yb_yTe_4$. ZT values as high as 0.9 in the 1000-1100 K temperature range were observed for skutterudite, half-heusler and clathrate samples. Higher values appear possible but would require more research and development. In addition, establishing the long term thermal stability of these compounds at temperatures in excess of 1200 K requires more studies. Experimental results on the

$La_{3-x}Yb_yTe_4$ materials were consistent with data obtained during the late 1980's, with the high ZT values at 1275 K (> 1.2). These compositions have also demonstrated an excellent thermal stability up to temperatures in excess of 1400 K, with fairly low sublimation rates. $La_{3-x}Yb_yTe_4$ materials appear to have good mechanical and thermoelectric compatibility with the low temperature Skutterudite materials. In addition, JPL developed the ability to synthesize these materials using a mechanical alloying technique. Based on these findings, and the possibility of significant further performance improvements by optimizing doping level and composition, this material was selected for the high temperature n-type STMC segment.

p-Type Materials—Candidate p-type materials included $CeFe_{4-x}Ru_xSb_{12}$ (filled skutterudites), $Cu_2Mo_6Se_8$-based Chevrel phases, $Ba_8Ga_{18}(Ge,Si)_{30}$ clathrates, and refractory Zintl antimonide phases such as $Yb_{14}MnSb_{11}$. High ZT values reported previously on p-type clathrate compositions could not be reproduced (best value obtained was 0.3). In spite of attractive results obtained in a previous JPL effort on Chevrel phases, including excellent thermal stability up to 1500 K, optimization of the metal atom filling of the Mo_6Se_8 structure and substitution on both the Mo and Se site did not result in improving ZT values beyond 0.6 at 1275 K. The best results were by far obtained on Zintl antimonide compounds based on $Yb_{14}MnSb_{11}$ with ZT values as high as 1.1 in the 1150 to 1250 K temperature range. Zintl materials have demonstrated short term thermal stability in vacuum up to 1350 K. The initial set of materials was synthesized at the University of California at Davis using a flux technique [ref]. JPL also developed a mechanical alloying technique to prepare such compositions and allow for a more practical synthesis scale-up for device development. At the conclusion of the materials screening effort, powder metallurgy techniques have been successfully developed for all four STMC thermoelectric material segments.

The temperature dependent variation of the ZT values of the selected STMC materials are shown in Figure 3a (p-type) and 3b (n-type). The average ZT values over the projected 1275 – 750 K temperature range are also shown, with the yellow colored band corresponding to the minimum and ultimate ZT goal to meet the STMC performance requirements. Data for state-of-practice p-type and n-type Si-Ge alloys used on space radioisotope thermoelectric generators is also shown. The STMC materials would provide over 50% improvement in performance.

STMC Component and Assembly Process Development

This parallel effort focused on developing subscale STMC modules composed of 4 or 8 couples of low temperature (LT) skutterudite materials capable of operating up to 975 K. The objective was to determine the best bonding methods and process conditions for preparing multicouple layer-to-layer bonds, but also to attempt to minimize the number of high temperature assembly steps on the most fragile components, possibly the high voltage insulators and the thermoelectric elements. In addition, the fabrication approach addressed one of the key issues for high power thermoelectric converters, namely scaling up of the various fabrication and assembly processes. The main features of the flexible aerogel-filled STMC module design and interconnection schemes are shown in Figure 3. They include five discrete component sub-assemblies that are pre-fabricated using separate processes, then aligned and indexed using polymeric egg-crates for assembly that vaporize during the single diffusion bond step. The aerogel thermal packaging is cast after assembly of the module using a low temperature process. The lightweight, nanoporous aerogel is also used a barrier to potential sublimation products from the thermoelectric elements.

The low temperature thermoelectric module converter (LT-TMC) component development activity was directed at the fabrication of the five sub-assemblies: cold side and hot side high voltage insulator sub-assemblies (HVISAs), n-type and p-type sub-assemblies (TELSAs) and the cold and hot side heat exchanger sub-assemblies (HXSAs).

Thermoelectric Leg Sub-Assemblies—Past work on skutterudite materials and unicouple technology [Caillat 2005] had succeeded in developing a reproducible lab-scale material co-hot pressing process for producing 20 mm long, 12 mm diameter cylindrical samples of the thermoelectric materials terminated at both ends with a refractory conductive metal (Ti). To demonstrate the ability to produce large quantities of thermoelectric legs with a geometry compatible with the high power converter design, the STMC program built on this effort and successfully developed a similar process to produce 8 mm thick, 40 mm diameter metallized skutterudite pucks. In the course of four weeks, about 1 kg of skutterudite material was produced using a single high temperature hot-press (about 1% of what is needed for a 100 kW class converter system). After hot pressing the pucks were lapped and diced using a diamond saw. Subsequently, a high precision, high yield electrical discharge machining (EDM) process was demonstrated, with 65 legs out of 68 possible obtained for a 40 mm puck, based on the 8-couple LT-TMC design. In the course of producing several sets of n-type and p-type pucks, issues related to the uniformity of the metal layer thickness and elimination of structural defects were resolved. Six n-type and p-type sets each for the 8-couple and 4-couple LT-TMC designs were fully prepared.

High Voltage Insulators and Interconnects Sub-Assemblies—In addition to providing a good thermal and mechanical interface between the thermoelectric materials and the heat source and heat sink, as shown in Figure 4, the HVISAs combine the functions of electrical conduction between the n-type and p-type legs and electrical insulation from the heat exchangers. Design studies had identified sapphire and beryllia (BEO) as the insulator materials of

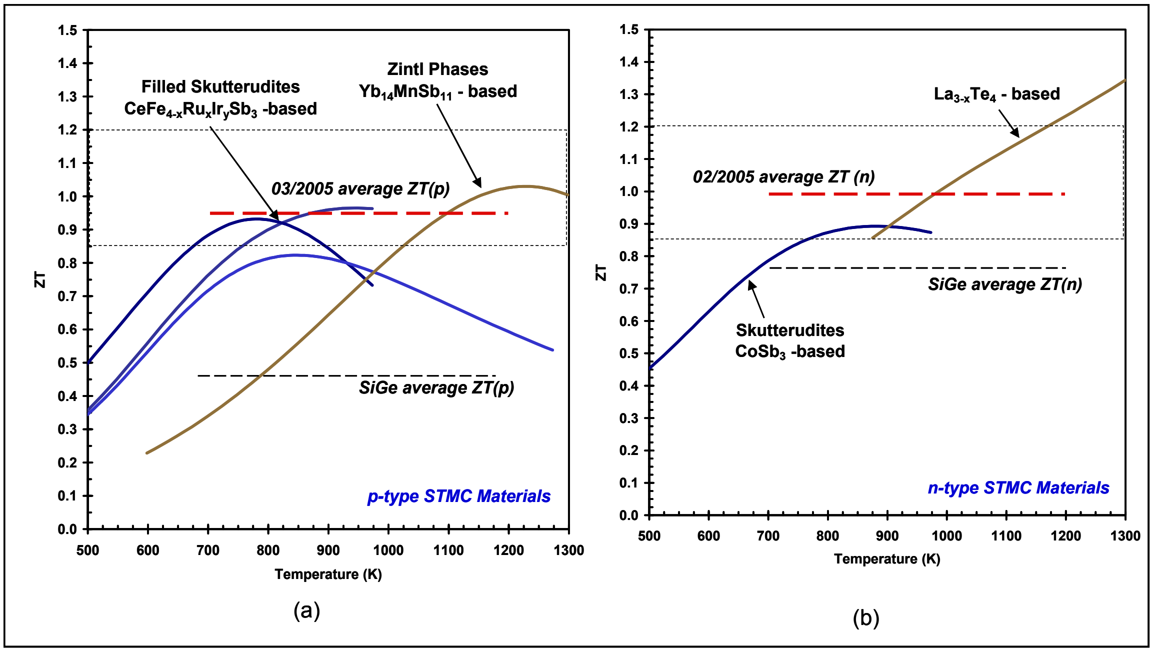

Figure 3 - ZT values as a function of temperature for (a) n-type materials and (b) p-type materials selected for development of the high temperature STMC technology. The STMC materials increase average ZT values to 1.0 in the 1275 – 750 K temperature range, 50% over state-of-practice SiGe. ZT values well above the minimum 0.85 goal have been obtained.

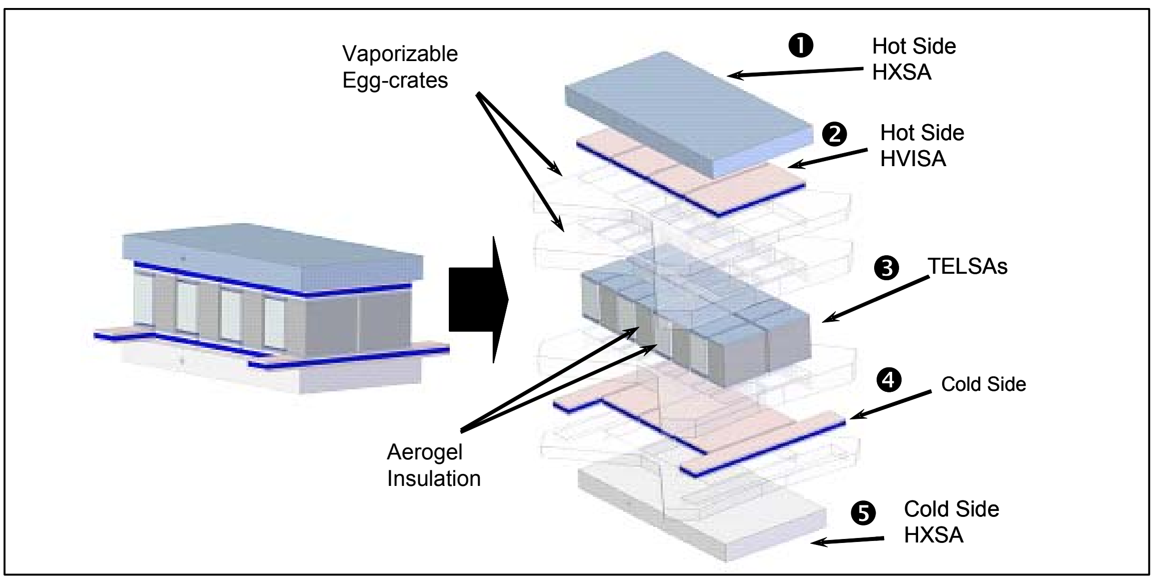

Figure 4 - 2x4 LT-TMC Schematic showing all five component sub-assemblies: heat exchanger sub-assemblies (HXSA), thermoelectric leg sub-assemblies (TELSAs) and high voltage insulator sub-assemblies (HVISAs). The vaporizable egg-crate polymeric plates used for aligning all the parts prior to assembly step are also shown. Aerogel thermal insulation packaging is cast after the assembly is completed.

choice for the hot side and cold side HVISAs respectively. A key driver for the HVISA development was the desire to have the insulators terminated with a metal compatible with a low temperature diffusion bonding process to the Ti terminated thermoelectric legs. A survey to identify commercial U.S. sources for metallization of beryllia and sapphire was conducted. Following this survey effort a number of insulator bonding and brazing trials were conducted using Mo and Nb and two vendors were each directed to apply commercial techniques for fabricating the HVISA. In parallel, custom processes were developed to widen the choice of metallization materials to include V and Pd. Also, insulator trials were conducted with BeO bonded or brazed to Ti, Pd and V. Evaluation factors included diffusion bonding temperature, braze composition and brazing temperature, thermally induced stresses due to CTE mismatches and ability to dice crack-free to the desire HVISA sizes. Batches of Pd-bonded sapphire were produced in house, while three sets of Pd-brazed BeO were procured. While long term stability needs to be demonstrated, Pd was found to b e an attractive choice for the LT-TMC fabrication by allowing for a single low pressure, low temperature diffusion bonding process to the TELSAs and HXSAs, as well as providing some mechanical compliance for the full stack of materials.

Heat Exchanger Sub-Assemblies—A thermal hydraulic study performed at the PCA and TCA levels confirmed that the multichannel hollow flat plate geometry used in the SP-100 thermoelectric converter technology development was also an adequate configuration for the cold and hot side liquid metal heat exchangers of the STMC under planned operating conditions [Fleurial 2006]. While there was no plan to develop fully functional heat exchanger component for the sub-scale STMC modules developed in this phase of the effort, it was still important to use materials and form factors representative of a full TCA. As a result, SS304L, Nb-1Zr and Ti solid rectangular plates were machined for the fabrication trials to serve as thermal and mechanical mock-ups. Coupon bonding tests with Pd foil showed that both Ti and SS304L were compatible with the low temperature and low pressure conditions desired for assembly of the full module. Nb-1Zr coupons required higher temperatures, and an additional fabrication step was used for bonding a thin Ti foil on the side to be later attached to the Pd-terminated HVISA.

STMC Module Assembly and Test

Following completion of this first round STMC component development, a total of eleven LT-TMC module fabrication trial runs were conducted at TESI, JPL, and PWR. While the ultimate objective of the trial runs was to fabricate a fully functional module and test it under nominal operating conditions, these experiments were designed to fully demonstrate the effectiveness of the vaporizable polymeric egg-crate alignment process, to assess the component dimensional tolerance requirements and to optimize the pressure and temperature conditions for this last diffusion bond step.

Assembly and Packaging—A first trial run conducted early on during the converter design process was used to validate the vaporizable egg-crate approach, successfully demonstrating the bonding of all parts while leaving no organic residues and maintaining all parts as originally positioned and indexed. The egg-crate plates were produced and patterned using a laser drilling process at an outside vendor facility. This first test was done with dummy Ti legs instead of the skutterudite legs, but it allowed eliminating alternate approaches relying on much more expensive and unwieldy fixtures such as dies and rails made out of high purity graphite. Out of the ten remaining trial runs, six runs were for 2x4 LT-TMC modules and four were for 2x2 LT-TMC modules. The first four of these runs were performed on 2x4 modules, and resulted only in partial success, mostly attributed to a combination of insufficiently meeting height tolerances for the TELSAs, inadequate high temperature bonding equipment configuration and insufficient pressure applied on egg-crated stack of components. Fully functional devices, two 2x4 and four 2x2 modules, were subsequently assembled quickly in succession. Figure 5 shows a 2x4 module after assembly and prior to high temperature testing.

After completing the assembly process, several techniques were explored to attach electrical wires for testing. While conceptually feasible, the current egg-crate layout and design of the HVISAs did not allow for in-situ bonding of the wires. Because the connections were located on the cold side of the devices (to reduce heat losses), a high temperature solder was selected for attaching thick tinned and braided copper wires to the Pd terminated HVISA pads protruding from the modules (see Figure 6a).

The use of vaporizable egg-crates enables post-assembly thermal packaging by a lightweight aerogel insulation that also serves as a sublimation barrier. Detailed studies related to the effectiveness of aerogel materials as sublimation barriers for thermoelectric elements operating at elevated temperatures are reported elsewhere [ref]. Past experiments on large thermoelectric module mockups (20x20 cm square) have demonstrated the ability of aerogel to infiltrate close-packed networks, down to microns of spacing. Close-packing actually improves the ability of aerogel to adhere intimately with the component surface, and minimize any physical shrinking of the nanoporous insulator due to long term exposure to vacuum and high temperatures. Two aerogel casting processes for thermoelectric modules have recently been developed [Sakamoto 2003; Tang 2005]. One of these processes was applied successfully to three LT-TMC modules, one 2x4 module and two 2x2 modules. A picture illustrating a fully packaged 2x2 LT-TMC module is shown on Figure 6b.

Module Tests—Two of the 2x4 LT-TMCs and three of the 2x2 LT-TMC devices have been tested. Some of the early measurements were used to ensure that the calorimetric test setup used (see Figure 7a) would be able to generate the desired heat source and heat sink temperature (975 K to 425K). In addition, measurements were made to verify that the temperature differentials across the top Mo calorimeter and across the bottom stainless steel calorimeter s were consistent with the predicted values calculated from the literature thermal conductivity values of each material. Test results on the 2x2 LT-TMC modules #2 and #4 are discussed here. Measurements on both devices near room temperature indicated that the internal device resistance was close to 5.4 mΩ, a value within 2% to that predicted by the module performance simulation (5.5 mΩ). The modules were mounted under a low compression load within the calorimeter-type high temperature fixture for measuring

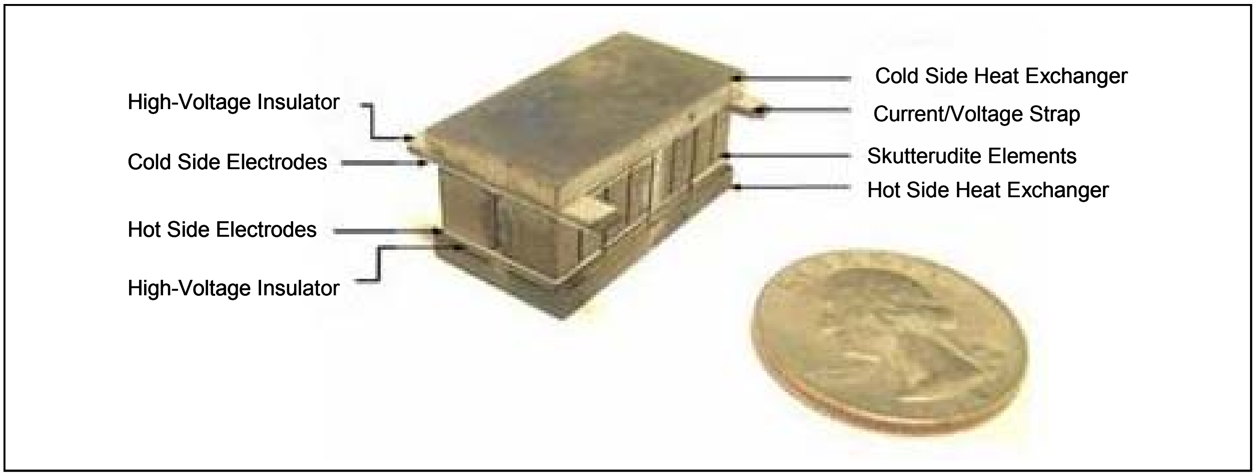

Figure 5 - 2x4 LT-TMC after fabrication. 8 couple module with 2 parallel strings of 4 couples in series each. The egg-crate plates used for alignment and indexing of the component have vaporized, leaving empty spaces between the thermoelectric legs.

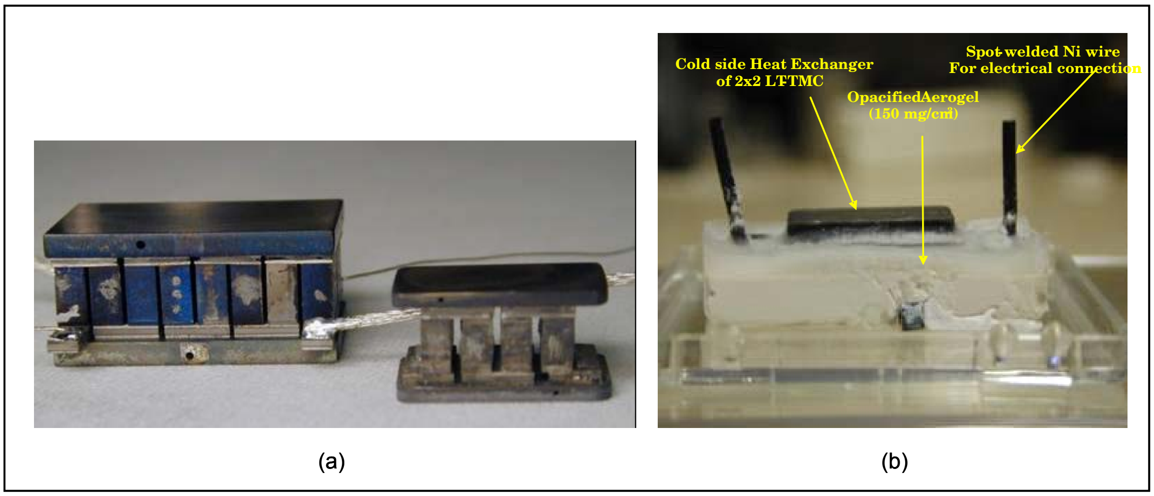

Figure 6 (a) - All-sapphire 2x2 LT-TMC module (right) and sapphire/beryllia 2x4 LT-TMC module with soldered current and voltage leads; (b) 2x2 LT-TMC module after packaging with thermal insulation (aerogel cast to fill the network of TE legs).

electrical power output, verify thermal performance across a maximum design temperature differential of 550K and mechanical integrity throughout the high temperature tests. Two measurements were made four days apart on module #2 (80 hours of operation under maximum power conditions) with the module operated across 400 K ΔT, with the hot side heat exchanger temperature above 810 K. Maximum power output was 0.695 W, with the performance remaining steady over the full 80 hours of operation with a current output of 8.5 A. This represented a current density of over 35 A/cm2 through each of the TE legs. A thermal analysis showed that about 24 W of heat was transferred through the module, corresponding to a heat flux density of 24.4 W/cm2. In addition, it was estimated that the parasitic ΔT losses (across the non-power producing layers of the material stack) were less than 10%, even under such high heat fluxes. Test data on the 2x2 LT-TMC #4 module with an improved electrical connection setup are shown in Figure 7b. While the full ΔT was not achieved across the thermoelectric legs, the experimental performance was within 5% of the predicted values, as shown in the summary Table 2. Maximum power output was 1.02 W and the internal resistance was 5.429 mΩ, within 2% of predicted performance. While a complete set of post test analyses could not be completed, including destructive metallography and microscopy of the interfaces, some important findings were made: a) the last fabrication trial runs using increased pressure loading during the diffusion bonding process significantly improved the mechanical integrity of the devices – 2x2 module #4 was left on extended test for over 1500 h and was mechanically intact after removal from the test station; b) the module components did survive the few thermal cycles they were subjected to, between room

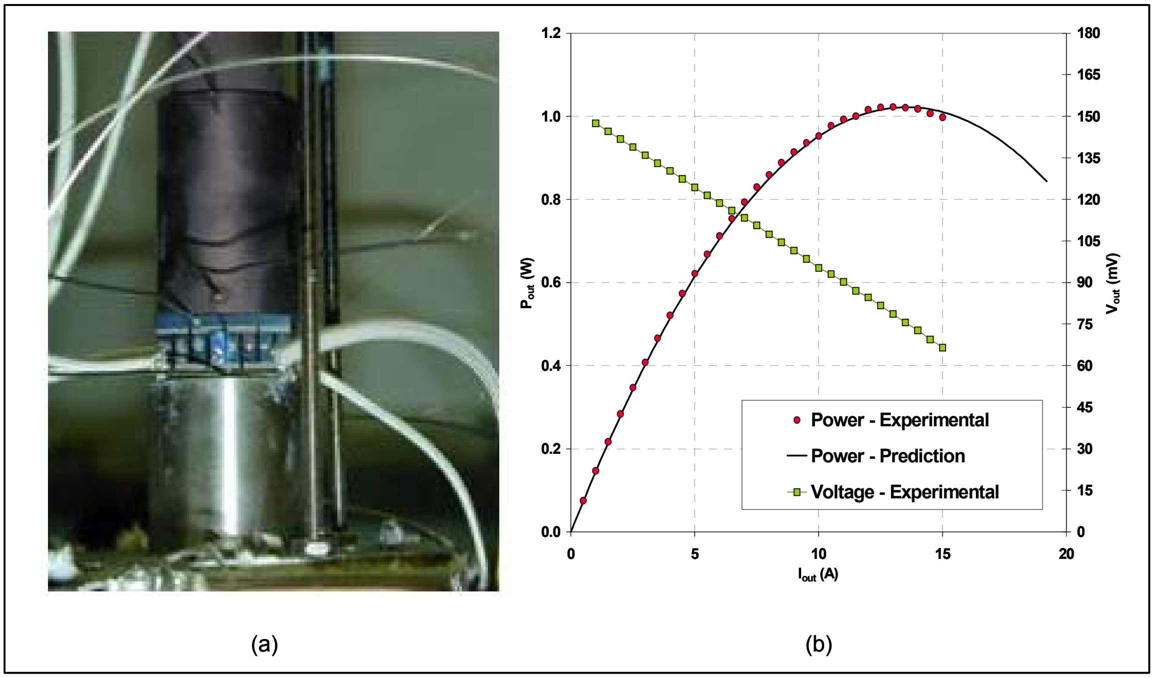

Figure 7 (a) - 2x4 LT-TMC module mounted and instrumented in the high temperature test station. Cylindrical calorimeters made out of Mo (top) and stainless steel 304 (bottom) are used for determining the actual heat flux across the module; (b) I-P curve at 822 K/479 K hot and cold side heat exchanger temperatures. Maximum power output was 1.016 W and the internal resistance was 5.429 mΩ, within 2% of predicted performance.

Table 2 - Predicted and experimental values for 2x2 LT-TMC #4 show good agreement

2x2 LT-TMC #4	**Voltage (mV)**	**Current (A)**	**Power (W)**	**R_{int} (mΩ)**	**R_{Load} (mΩ)**	**Q_{in} (W)**	**Q_{out} (W)**	**Efficiency (%)**	**Effective ZT**
Prediction	84.04	12.00	1.008	5.529	7.003	21.37	20.36	4.720	0.623
Experimental	84.65	12.00	1.016	5.429	7.054	22.10	20.10	--	--

temperature and up to 1000 K; c) the single crystal sapphire HVISAs were easily cracked in a direction perpendicular to the cut during the in-house dicing process, leading to sometimes significant thermal resistances on the hot side of the modules and non uniformity of the temperatures across the legs; d) the commercially procured polycrystalline beryllia HVISAs performed flawlessly, with no apparent damage observed post-assembly or post-test; d) the internal electrical resistance of all the fully bonded modules steadily increased during extended tests – possible degradation mechanisms include interdiffusion of the thermoelectric material to metal interfaces, an evolution of the Ti-Pd interfaces, sublimation of Sb from the skutterudite materials at the hot junctions, microscopic cracks propagating through the legs due to CTE mismatches. Further tests and analyses will be required.

CONCLUSIONS

The overall objective of this task was to develop an advanced segmented thermoelectric multi-couple converter (STMC) required for future space reactor power systems. The STMC is projected to provide a 9% thermal to electric conversion efficiency (SOA Si-Ge ~ 4.5%) and capable of reliable operation for more than 15 years for space nuclear electric propulsion missions. The space reactor power system (SRPS) employing STMC is projected to provide 20-30% mass savings compared to the SP-100 system based on Si-Ge multicouple converters. The updated conceptual design of the 100 kWe SRPS based on an STMC selected a liquid metal cooled reactor SRPS configuration with the thermoelectric converters conductively coupled to both the primary and secondary heat transfer loops to minimize the overall system mass. The specific objectives throughout the past eighteen months were grouped in two focus areas, high efficiency high temperature thermoelectric materials and the development of multicouple devices using the low temperature skutterudites capable of operating up to 975 K as an intermediate demonstration step towards fully segmented modules. Following completion of a multi-pronged study, the materials selected for the high temperature STMC are n-type $La_{3-x}Yb_yTe_4$ segmented with n-type $CoSb_3$ skutterudite and p-type $Yb_{14}MnSb_{11}$ Zintl compound segmented with p-type $CeFe_3RuSb_{12}$ filled skutterudite, with a combined average ZT value of 1.0 across the 1275 to 750 K projected operating temperature range, clearly exceeding the minimum performance goal of 0.85. A thermal, hydraulic and mechanical analysis performed at the largest power converter assembly level (PCA) was used to design a STMC module structure that minimizes thermally induced stresses under operating conditions and conceptualize novel assembly processes that utilize fewer and simpler fabrication steps with the final assembly using a novel vaporizable egg-crate component for indexing and alignment of all component sub-assemblies. Significant progress was made in demonstrating that the STMC technology is compatible with large batch processing of the component sub-assemblies: TE legs, high voltage insulator/electrical interconnect pads, cold and hot side heat exchangers and egg-crates. A number of fully functional 2x2 and 2x4 modules were successfully fabricated and post-assembly thermal packaging of the LT-TMC modules by backfilling the leg network with refractory aerogel materials was demonstrated. Preliminary high temperature performance test results were encouraging, but significant efforts are still needed to continue the development of the segmented multicouple technology and demonstrate performance at the highest temperatures with minimal degradation for up to fifteen years of operation. A key product of the effort was the initiation of technology transfer between JPL, Teledyne and P&W Rocketdyne by developing and documenting LT-TMC fabrication and assembly procedures and conducting joint fabrication runs at each facility.

ACKNOWLEDGMENTS

The work described in this paper was performed at the Jet Propulsion Laboratory, California Institute of Technology under contract with the National Aeronautics and Space Administration (NASA ROSS-2002 NRA, In-Space Propulsion Technologies). The authors would like to acknowledge the thermoelectric materials university research group leads that contributed to this effort: Profs. Terry Tritt (Clemson), George Nolas (South Florida), Ctirad Uher (Michigan), Mercouri Kanatzidis (Michigan State), Rüdiger Dieckmann and Francis DiSalvo (Cornell), Robert Cava (Princeton) and Susan Kauzlarich (Davis). The authors would also like to thank Drs. Andrew Kindler and Elizabeth Yen for development of the polymeric egg-crate concept, Drs. Steve Jones and Jay Pak for aerogel packaging of the modules and Eric Stewart and his team at NASA/MSFC for support in developing the PCA and TCA designs.

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