

# Performance Test Results of a Skutterudite-Based Unicouple with a Metallic Coating

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**Abstract.** A performance test of a Skutterudite-based unicouple (MAY-04) with a metallic coating to suppress the sublimation of antimony from the legs near the hot junction is performed in vacuum ( $\sim 9 \times 10^{-7}$  torr) for  $\sim 2,000$  hours at hot and cold junction temperatures of  $892.1 \pm 11.9$  K and  $316.1 \pm 5.5$  K, respectively. The p-leg is made of  $\text{CeFe}_{3.5}\text{Co}_{0.5}\text{Sb}_{12}$  and the n-leg is made of  $\text{CoSb}_3$ . Presented are the measured voltage-current characteristics, electrical power, open-circuit voltage, and Seebeck coefficients of the legs as functions of cumulative test time. Also presented is the estimate of the conversion efficiency, shortly ( $\sim 96$  hrs) after the start of test. To demonstrate the effectiveness of the metallic coating, the measurements for MAY-04 are compared with those of two uncoated unicouples of the same leg materials (MAR-03 and JUN-03), which had been tested earlier. The cross-sectional areas of the legs in MAY-04 are larger than those in MAR-03 and JUN-03, tested in argon cover gas at  $\sim 0.051$ - $0.068$  MPa for 450 and 1200 hours, respectively. The open circuit voltage,  $V_{oc}$  (204 mV) of MAY-04 at Beginning-Of-Test (BOT) is almost the same as that of MAR-03, but higher than that of JUN-03 ( $\sim 180$  mV). Although the argon gas effectively decreased antimony loss from legs of MAR-03 and JUN-03, marked degradations in performance occurred with time in these tests. Conversely, the metallic coating in MAY-04 effectively reduced the performance degradation with cumulative test time. The estimated peak efficiency of MAY-04, shortly after BOT (10.65%) is only  $\sim 0.37$  percentage point lower than the theoretical value, assuming zero side heat losses and zero contact resistance per leg. The peak power of MAY-04 decreased by only  $\sim 12\%$ , from its BOT value of  $\sim 1.6 W_e$  to  $\sim 1.4 W_e$  after  $\sim 2,000$  of cumulative testing.

## INTRODUCTION

Radioisotope Thermoelectric Generators (RTGs) with SiGe thermoelectric unicouples had been used successfully in numerous NASA space exploration missions for more than four decades (Carpenter, 1970; Schock, 1980; and Bennett, Lombardo, and Rick, 1987). Because of the low efficiency of SiGe, new Radioisotope Power Systems (RPSs) are being developed using more efficient Skutterudite-based Unicouples (SKUs) to not only reduce the mass of the  $^{238}\text{PuO}_2$  fuel, but also to increase the specific electric power. For SKUs, currently under development, it is important to suppress the sublimation of the volatile antimony from the legs near the hot junction, thus minimizing the degradation in the performance with time in a multi-year space missions. For SiGe unicouples used very successfully in RTGs on board of numerous spacecraft during the last four decades (Carpenter, 1970; Schock, 1980; and Bennett, Lombardo, and Rick, 1987), a similar issue of the sublimation of germanium had been dealt with satisfactorily by applying a thin  $\text{Si}_3\text{N}_4$  coating (a few to tens of microns) on the legs near the hot junction.

A sublimation suppression metallic coating, that is compatible with the material of the legs in SKUs (p- $\text{CeFe}_{3.5}\text{Co}_{0.5}\text{Sb}_{12}$  and n- $\text{CoSb}_3$ ), has been developed at the Jet Propulsion Laboratory (JPL), California Institute of Technology, in Pasadena California. It has been shown to significantly reduce the loss of antimony at higher temperatures up to 973 K in tests conducted at JPL and at the University of New Mexico (Fleurial et al., 1996 and 1997; Caillat et al., 1999 and 2000; El-Genk, Saber, and Caillat, 2004; El-Genk et al., 2003 and 2004). A number of tests involving segmented and non-segmented SKUs, with and without the metallic coating, have been conducted in the high temperature vacuum facility at the University of New Mexico's Institute for Space and Nuclear Power Studies (ISNPS). These tests had been conducted for hundreds of hours (El-Genk, Saber, and Caillat, 2004; El-Genk et al., 2003 and 2004) at average hot and cold junction temperatures of  $\sim 973$  K and 300 K, respectively (Fig. 1). The purpose of these tests, conducted as a part of joint program between JPL and ISNPS, was to verify

Beginning-Of-Life (BOL) performance relative to the theoretical predictions. Tests of coated and uncoated uncouples are conducted both in vacuum and in argon cover gas (0.051 - 0.068 MPa), but the uncoated uncouples had been tested only in argon cover gas. As shown in Figure 1, the test performed at ISNPS included un-segmented SKUs, with p-legs of  $\text{CeFe}_{3.5}\text{Co}_{0.5}\text{Sb}_{12}$  and n-legs of  $\text{CoSb}_3$ , and segmented SKUs, in which the p-legs had two segments of  $\text{CeFe}_{3.5}\text{Co}_{0.5}\text{Sb}_{12}$  and  $\text{Bi}_{0.4}\text{Sb}_{1.6}\text{Te}_3$  and n-legs had two segments of  $\text{CoSb}_3$  and  $\text{Bi}_2\text{Te}_{2.95}\text{Se}_{0.05}$ . These tests are performed at almost the same average hot and cold junction temperatures in vacuum ( $\sim 9.0 \times 10^{-6}$  torr) and /or in argon gas (at 0.051 MPa and 0.068 MPa) (El-Genk, Saber, and Caillat, 2004; El-Genk et al., 2003 and 2004). Although not designed as life tests, the results provided valuable performance data for segmented and non-segmented SKUs for the longest test durations ever been reported to date.

The estimates of the peak efficiency of the segmented SKUs tested at ISNPS (Fig. 1) decreased from 13.5% at BOT, which is within 10% of the theoretical predictions (using the JPL materials properties database and the measured contact resistances in the tests and assuming zero side heat losses), to 10.8% after 406 hours of cumulative testing. Results showed that the side heat losses in these tests were responsible for the much lower actual peak efficiencies for example  $\sim 7.3\%$  and  $6.3\%$  for the non-segmented MAR-03 and JUL-03, respectively. The test results of JAN-04, a segmented SKU with metallic coating on the legs near the hot junction, showed significantly lower degradation in performance with cumulative test time compared to similar uncoated uncouples tested at almost the same hot and cold junction temperatures but in argon cover gas instead of vacuum (Fig. 1) (El-Genk, Saber, and Caillat, 2004; El-Genk et al., 2003 and 2004). The estimated peak efficiency of segmented, but uncoated SEP-03 SKU decreased from  $\sim 13.8\%$  at BOT to 5.8% at End-Of-Test (EOT) and its peak power decreased from 1.64  $W_e$  at BOT to 0.48  $W_e$  at EOT, however, the open circuit voltage,  $V_{oc}$ , decreased by  $\sim 14\%$ . The open circuit voltage of the coated and segmented JAN-04 decreased only by  $\sim 3\%$ , the estimated peak efficiency ( $\sim 12\%$ ) changed very little, and the measured peak electrical power decreased by  $\sim 20\%$  after  $\sim 1,000$  hours of cumulative testing. In addition, unlike the uncoated SEP-03, the measured total and contact resistances of the legs in JAN-04 changed very little.

This paper presents the results of the most recent test conducted at the ISNPS. It involved a non-segmented, coated SKU (MAY-04) (Fig. 1). This test lasted for  $\sim 2,000$  hours is conducted in vacuum ( $\sim 9.0 \times 10^{-7}$  torr) at hot and cold junction temperatures of  $892.10 \pm 11.90$  K and  $316.10 \pm 5.50$  K, respectively. To quantify the effectiveness of the metallic coating applied to the legs of MAY-04 near the hot junction, the test results are compared with those of two earlier tests of uncoated uncouples (MAR-03 and JUN-03) with same materials of the n- and p-legs. The cross-sectional areas of the legs in MAR-03 and JUN-03 are almost half those of MAY-04, but total lengths are similar.

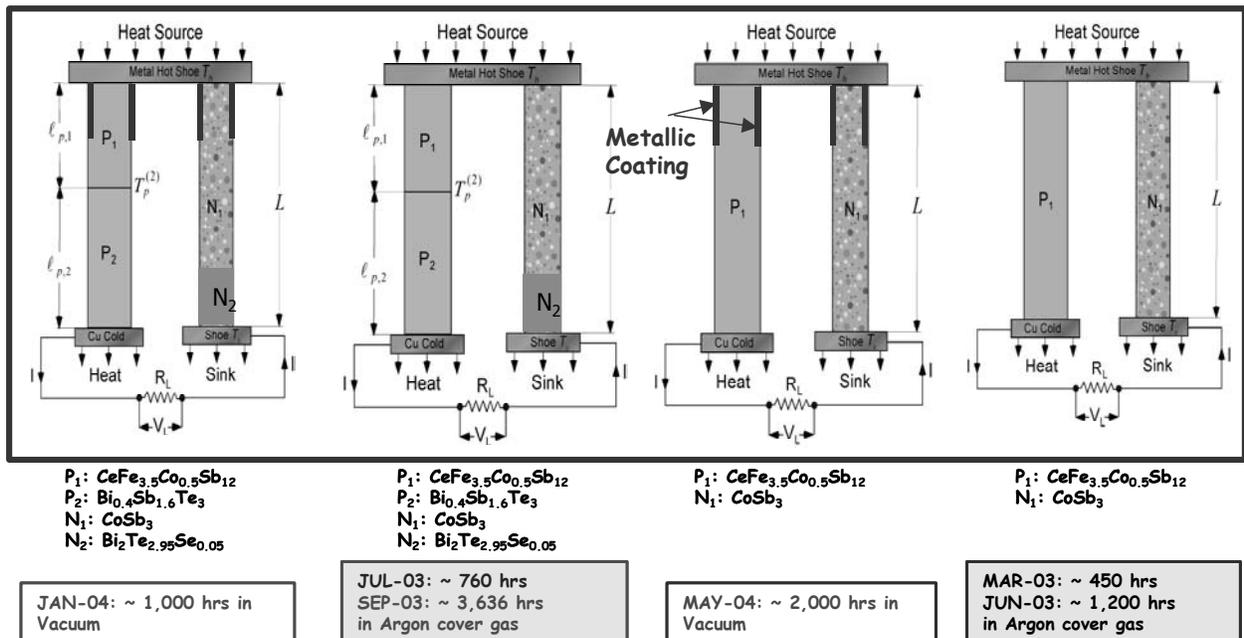


FIGURE 1. Skutterudite-Based Segmented and Non-Segmented Uncouples Tested at University of New Mexico.

## EXPERIMENT SETUP

The performance tests of the uncoated SKUs (MAR-03, JUN-03) (Fig. 1) were performed in argon cover gas at 0.051-0.068 MPa in order to minimize the loss of the volatile antimony (Sb) from the legs near the hot junction. In MAR-03, the 17.7 mm long n-leg is made of  $\text{CoSb}_3$  and the 19.1 mm long p-leg is made of  $\text{CeFe}_{3.5}\text{Co}_{0.5}\text{Sb}_{12}$  (El-Genk et al., 2003; El-Genk, Saber, and Caillat, 2004). In order to facilitate good thermal and electrical contacts between the hot and cold copper (Cu) shoes and the legs, the n-leg had 1.2 mm and 1.4 mm thick metallic electrodes at the hot and cold ends, respectively, and the p-leg had thin metallic electrodes at the hot end, but none at the cold end. The hot shoe is a copper disk that is 12.2 mm in diameter and 2.86 mm thick. In the tests performed in the high temperature vacuum facility at the ISNPS (Fig. 1) the legs were soldered to the Cu cold shoe. Good solid-solid contact is maintained between the hot shoe and the metallic electrodes of the-legs in the tests using four compression springs (Fig. 2c). The springs also accommodate the expansion of the legs in the test. The hot shoe is heated using an electric heater to which the DC input power is continuously regulated to maintain a constant hot junction temperature. The fully assembled unicouples are surrounded with fiberglass insulation to reduce side heat losses in the tests (Fig. 2c). The rejected thermal power by MAY-04 to the cold shoe in the test is removed using a chilled coolant (50% Ethylene Glycol and 50% distilled water) circulating through an underlying aluminum cold plate. The set up is the same for SKUs tested in the ISNPS facility (Fig. 1). The total lengths of the legs in the unicouples tested are similar, but the cross-sectional areas of the legs in MAR-3 and JUN-03 are almost half those in MAY-04. Figure 2a shows a schematic of MAY-04 SKU with the temperature and voltage measurements probes attached and Figure 2b shows the composition and dimensions of the n- and p-legs.

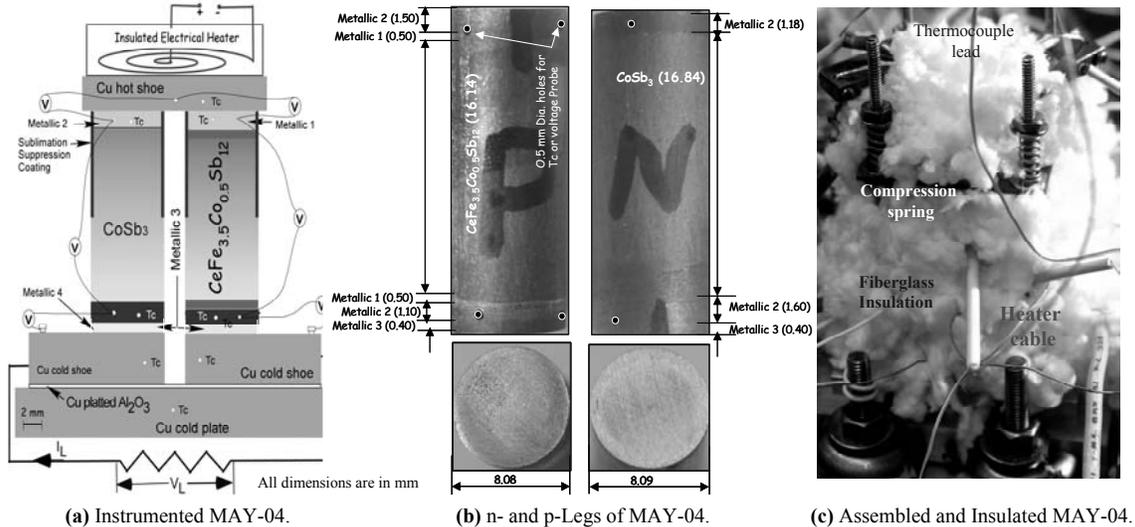


FIGURE 2. Assembled and Instrumented MAY-04 Before Testing in the ISNPS Vacuum Facility.

The measurements of the hot and cold shoes temperatures as well as those of the hot and cold junctions, the voltage across the legs and the MAY-04 unicumple, and the hot and cold contact resistances between the hot and cold shoes and the unicumple legs are recorded in the tests (Fig. 1). The test parameters are controlled, collected, and stored using a computer program that is based the LabView commercial software. The electrical power to the heater is continuously adjusted to maintain a constant hot junction or hot shoe temperature in the tests. A number of sweeps of the voltage-current characteristics are conducted per week at a user prescribed times. As indicated earlier, MAR-03 and JUN-03 tests

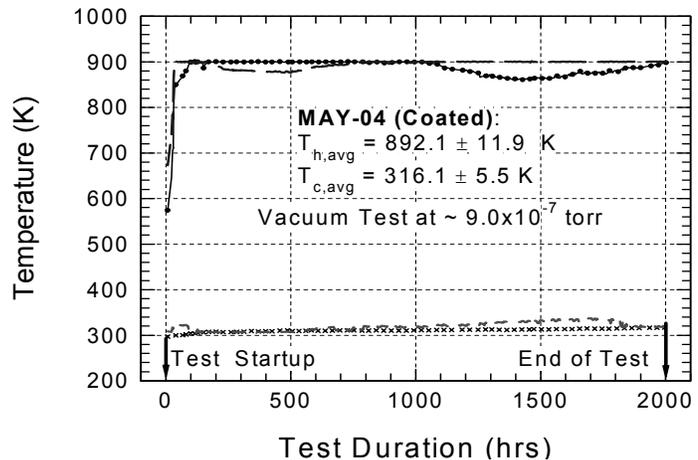


FIGURE 3. Testing History of MAY-04.

were conducted in a tight bell jar at 0.051 –0.068 MPa of argon cover gas. MAY-04 is conducted in vacuum at  $\sim 9.0 \times 10^{-7}$  torr at a hot and cold junctions temperature of  $892.10 \pm 11.90$  K and  $316.10 \pm 5.50$  K (Fig. 3), respectively. The test of MAR-03 lasted for 450 hours, of which 261 hours were at hot and cold junction temperatures of  $972.10 \pm 10.0$  K and  $301.10 \pm 5.10$  K, respectively (El-Genk et al., 2003). The test duration for JUN-03, at almost same average hot and cold junction temperatures, is 1,200 hours (El-Genk, Saber, and Caillat, 2003). Table 1 lists the dimensions of the legs of MAR-03, JUN-03, and MAY-03 SKUs.

**TABLE 1** Dimension of the Legs of MAR-03, JUN-03, and MAY-03 SKUs and Test Conditions.

Parameter	MAR-03 (Uncoated SKU)		JUN-03 (Uncoated SKU)		MAY-04 (Coated SKU)	
	p-leg	n-leg	p-leg	n-leg	p-leg	n-leg
Cross section Area (mm <sup>2</sup> )	23.82	24.83	21.16	22.33	51.23	51.36
Materials of Legs	CeFe <sub>3.5</sub> Co <sub>0.5</sub> Sb <sub>12</sub>	CoSb <sub>3</sub>	CeFe <sub>3.5</sub> Co <sub>0.5</sub> Sb <sub>12</sub>	CoSb <sub>3</sub>	CeFe <sub>3.5</sub> Co <sub>0.5</sub> Sb <sub>12</sub>	CoSb <sub>3</sub>
Length of TE Material in Legs (mm)	19.10	17.70	18.02	18.28	16.14	16.84
Total length of the legs (mm)	20.30	20.30	20.79	20.80	20.14	20.02
Test Environment	argon gas		Argon cover gas		Vacuum	
Test Pressure (Pa)	51, 000 and 68,000		51, 000 and 68,000		$1.18 \times 10^{-4}$ ( $\sim 9 \times 10^{-7}$ torr)	
Av. Hot Junction Temperature (K)	$974.9 \pm 10.0$	$970.3 \pm 10.0$	$976.6 \pm 6.20$	$971.5 \pm 6.20$	$895.1 \pm 11.90$	$889.1 \pm 11.90$
Av. Cold Junction Temperature (K)	$301.8 \pm 5.10$	$300.3 \pm 5.10$	$301.8 \pm 8.30$	$301.8 \pm 8.30$	$320.5 \pm 5.50$	$311.6 \pm 5.50$
Total Test Duration (hrs)	$\sim 261.0$		$\sim 1,200.0$		$\sim 2,000.0$	

## RESULTS AND DISCUSSION

The measured open circuit voltage ( $V_{oc}$ ) and V-I characteristics of MAY-04 SKU are used to obtain average Seebeck coefficients of the materials of the n- and p-legs and the electric power generated versus the load current, respectively. The estimate of the conversion efficiency of MAY-04 SKU is calculated shortly after the start of the test ( $\sim 96$  hours) when the hot and cold junction temperature reached  $892.10 \pm 11.90$  K and  $316.10 \pm 5.50$  K, respectively. These temperatures are kept almost constant at these values through the end of the test. The estimate of the conversion efficiency is based on the measured contact resistances at the hot and cold junctions of the n- and p-legs and neglecting the contact resistances of the metallic electrodes at the hot and cold junctions of the legs (Fig. 2b). The estimate of the conversion efficiency at BOT also assumes zero side heat losses and uses the JPL thermoelectric properties database for the materials of the legs in MAY-04. It should be noted that the contact resistance between the metallic electrodes and the legs and between the electrodes and copper hot shoe changes with time in the test. This has been confirmed by the post-test visual observation of the MAY-04 SKU. Extensive chemical reactions occurred at the interfaces of the metallic electrodes and both the legs and the copper hot shoe. The change in the contact resistance with the Cu hot shoe has been measured as a function of time in the test, but that between the electrodes and legs was not measured because of insufficient holes in the electrodes and in the legs near the interface to insert voltage probes. The performance measurements in the tests of the uncoated SKUs, MAR-03 and JUN-04 are similar to those recorded for MAY-04, and have been previously published (El-Genk et al., 2003; El-Genk, Saber, and Caillat, 2004) thus would not repeated here in its entirety. Only, selected measurements will be compared with those of MAY-04.

### V-I Characteristics

Figures 3a and 3b present the measured V-I and  $P_e$ -I characteristics for MAY-04 at different times during the test. As indicated earlier, the cumulative test duration of MAY-04 is  $\sim 2,000$  hours. BOT open circuit voltage,  $V_{oc}$ , of  $\sim 204.2$  mV (Fig. 3a) is similar to that measured for MAR-03 (203.6 mV), but higher than that for JUN-03 (180 mV). The open circuit voltage for MAY-04 stayed almost constant at its BOT value during the first 800 hours of the test, then decreased slowly with time to reach  $\sim 188$  mV, when the test was terminated  $\sim 1200$  hours later. Conversely, the open circuit voltage of the uncoated MAR-03 and JUN-03 SKUs, decreased rapidly with time in the test. For example,  $V_{oc}$  of MAR-03 decreased linearly with time to 183 mV, only after 261 hours. Similarly,  $V_{oc}$  for JUN-03 decreased initially linearly to 155 mV after  $\sim 400$  hours, then changed very little after through the end of test  $\sim 800$  hours latter. As shown in Figs. 3a and 3b, the short circuit current for MAY-03 decreased from  $\sim 32$  A at BOT to  $\sim 30$  A after 1961 hours of cumulative testing. Such a decrease reflects the measured decrease in the open circuit voltage (Fig. 3a) as well as the change in the contact resistances of the n- and p-legs. However, the increase in the

slope of the V-I characteristics is only indicative on the increase in the total resistances of the legs, including the contacts. As delineated in Fig. 3a the slope of the V-I characteristics of MAY-04 increased with test time, decreasing the peak electric power and shifting it to lower current (Fig. 3b).

The measured peak electric power of MAY-04 ~ 96 hours after BOT is ~ 1.63  $W_e$  and occurs at a load current of 16.0 A. This value, however, decreased to ~ 1.40  $W_e$  (a decrease of ~ 14%) after 1961 hours of cumulative testing (Fig. 3b). The measured decrease in  $V_{oc}$  of MAY-05 with time in the test is indicative of the decrease in the Seebeck coefficients of the legs and /or the increase in the side heat losses in the tests. As Fig. 2c shows, to minimize the radiation side heat losses in the test a molybdenum foil surrounds the legs of MAY-04. However, the deposition of reaction products and sublimed antimony onto the inner surface of the foil could have increased the surface emissivity and, hence increased the side heat losses from the legs with cumulative test time.

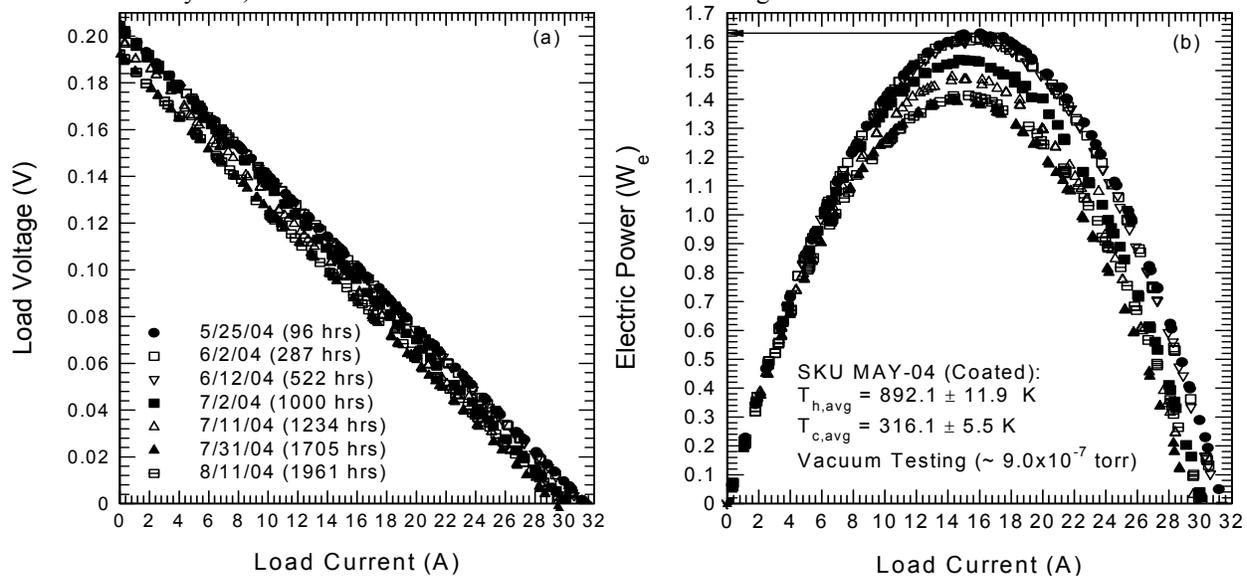


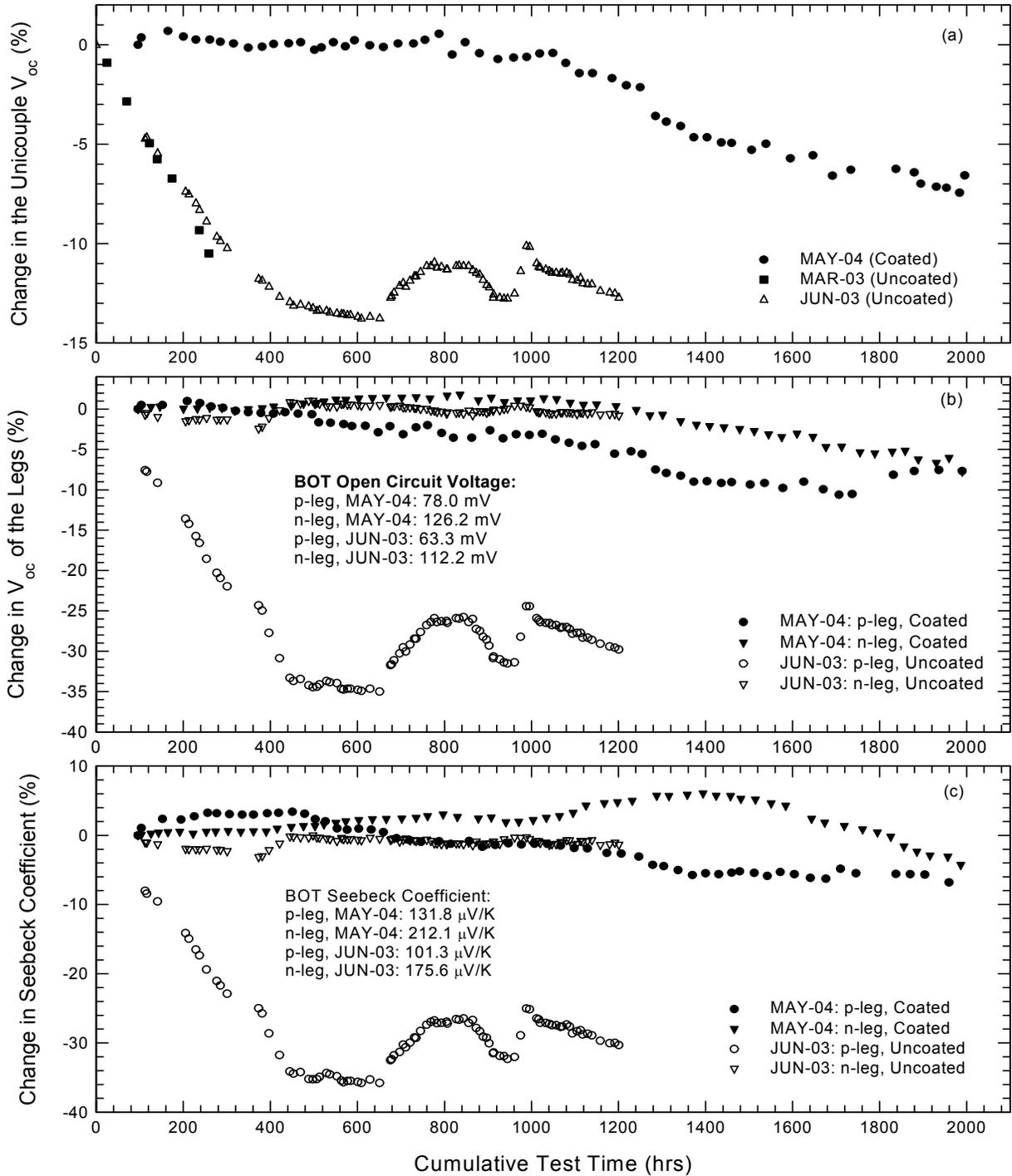
FIGURE 3. Measured V-I Characteristics for MAY-04 SKU with Metallic Coating.

### Changes in Open Circuit Voltage and Legs Seebeck Coefficients

Figure 4a compares the measured changes in Seebeck coefficient of MAY-04 with those of the uncoated SKUs MAR-03 and JUN-03, as functions of time in the respective tests. These changes are relative to the measured  $V_{oc}$  values at BOT listed in Figs 4b - 4c. Figure 4a indicates that the open circuit voltage of JUN-03 decreased linearly ~ 14% during the first 400 hours of testing then increased somewhat, cutting the decrease to ~ 12- 13%. The decrease in  $V_{oc}$  of MAR-03 during 261 hours of testing at almost the same hot and cold junctions temperatures, is consistent with that measured for JUN-03. Unlike MAR-03 and JUN-03,  $V_{oc}$  of MAY-04 decreased very little in the test. It increased initially only slightly and remained almost constant for the first 800 hours of the test. Subsequently, it decreased almost linearly with test time to ~ 7.5% at the end of ~ 2,000 hours of cumulative testing. These results suggest that the metallic coating applied to the legs of MAY-04 near the hot junctions have been effective in reducing the changes in Seebeck coefficients of the legs.

Figure 4b compared the changes in the open circuits voltages of the legs of MAY-04 and JUN-04, as functions of time in the respective tests. Figure 4c present the corresponding changes in Seebeck coefficient of the materials of the legs. These Figures clearly show that the measured decreases in the open circuit voltage and the Seebeck coefficient in JUN-03 were solely due to the p-leg ( $CeFe_{3.5}Co_{0.5}Sb_{12}$ );  $V_{oc}$  and Seebeck coefficient of the n-leg ( $CoSb_3$ ) was almost unchanged in the test. The total decreases in  $V_{oc}$  and the Seebeck coefficient of the p-leg in JUN-03 are the same ~ 30%. For MAY-04, the values of  $V_{oc}$  of the legs remained unchanged during the first 400 hours of testing. After that,  $V_{oc}$  of the n-leg ( $CoSb_3$ ) increased slowly by ~ 2% after an additional 700 hours (or 1,100 hours from BOT), then decreased linearly to a total of ~ 7% below its BOT value at end of test (~2,000 hours). After 400 hours from BOT,  $V_{oc}$  of the p-leg ( $CeFe_{3.5}Co_{0.5}Sb_{12}$ ) in MAY-04 decreased gradually ~ 11% below its BOT value after ~ 1,800 hours, then increased slightly cutting the total decrease to ~ 8% below BOT value

at the end of the test (~ 2,000 hours) (Fig. 4b). The corresponding decreases in the Seebeck coefficients of the n- and p-legs in MAY-05 are shown in Fig. 4c. These coefficients remained almost unchanged during the first ~ 400 – 450 hrs of testing, then that of the p-leg decreased with test time to ~ 7% below its BOT value at the end of the test. After 400 hours of testing, the Seebeck coefficient of the n-leg in MAY-04 increased by ~5% after ~ 1,400 hrs from BOT, then decreased to ~ 4% below its BOT value, 600 hours latter, or at the end of the test (~2,000 hrs).



**FIGURE 4.** Measured Changes in Open Circuit Voltage and Seebeck Coefficients of MAY-04 legs.

These results clearly demonstrate once again the effectiveness of the metallic coating applied to the legs of MAY-04 SKU. The decrease in Seebeck coefficients of the legs after ~2,000 hours of cumulative testing was  $\leq 7\%$  (Fig. 4c),

significantly lower than those experienced by the uncoated JUN-03 SKU. For this unicouple, the Seebeck of the p-leg material decreased by 30%, during 1,200 hours of cumulative testing (Fig. 4b). It is worth noting that during the 1,200 hours of testing of JUN-03 the Seebeck coefficient of the n-leg remained almost unchanged, consistent with the result of n-leg in MAY-04, during the same time of cumulative testing (Fig. 4c).

### Peak Electrical Power and Conversion Efficiency

Figure 5a presents the measured peak electrical power of MAY-03 as a function of cumulative test time and Fig. 5b presents the estimates of the conversion efficiency ~ 96 hours after the start of the test (solid circle symbols). These estimates are based on the measured hot and cold junction temperatures and the measured contact resistances in the test at the hot and cold junctions. These estimates also used the JPL thermoelectric properties database for the n- and p-leg and assumed zero side heat losses. The theoretical limits of the conversion efficiency are those shown in Fig. 5b for zero side heat losses and zero total contact resistance per leg (or  $r_{\text{cont}} = 0.0 \mu\Omega\text{-cm}^2$ ), but at the same hot and cold junction temperatures measured in the test. For a practical consideration, the efficiency estimates for  $r_{\text{cont}} = 150 \mu\Omega\text{-cm}^2$  per leg are also shown in Fig. 5b.

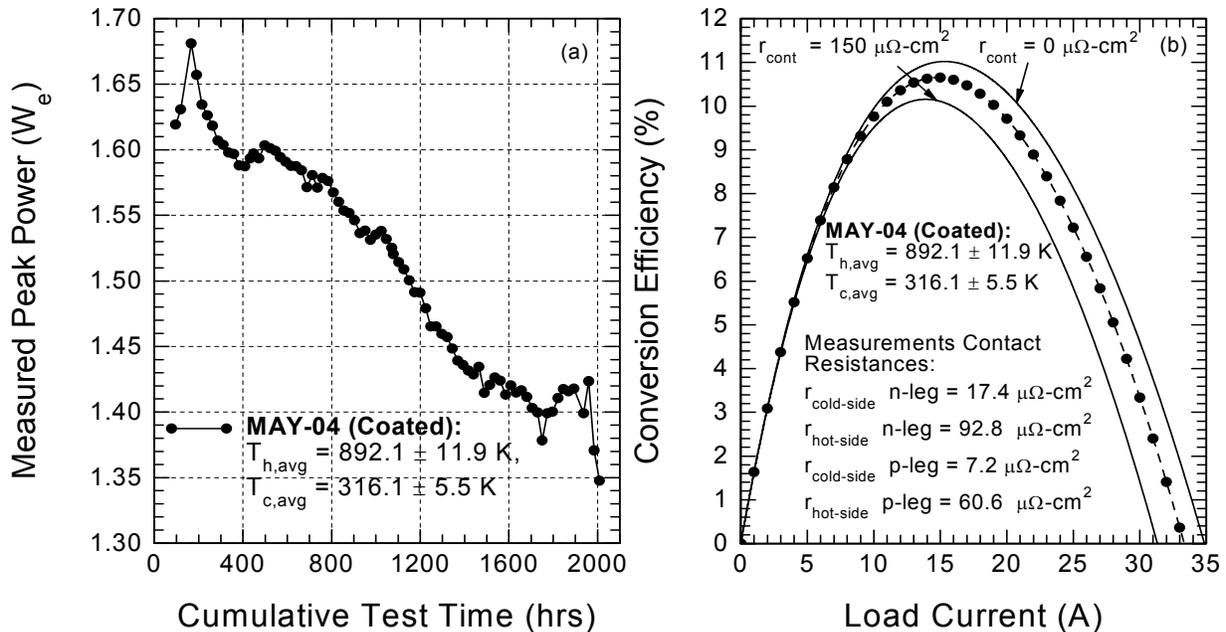


FIGURE 5. Measured Peak electrical Power and Efficiency Estimates for MAY-04 SKU.

The peak electric power, measured ~ 96 hours after starting MAY-04 test, was 1.62  $W_e$ . This power is measured when the hot junction temperature reached the value indicated in Fig. 5a for the first time. It took 96 hours to incrementally increase the hot junction temperature in the test and established contact with the copper hot shoe. At each temperature the test lasted for a few hours and one or two sweep of the voltage-current characteristics are collected. The early measurements at lower hot junction temperatures are not show in the Figures in this paper.

Figure 5a shows monetary increase in the measured peak electric power to ~ 1.68  $W_e$ , followed by an almost linear decrease with time to ~ 1.4  $W_e$  at the end of the test (after ~2,000 of cumulative testing). Such decrease is attributed to the increases in the contact resistances of the p- and n-legs at the hot junction with the copper hot shoe. Those between the cold junctions and the copper cold shoe (Fig. 5b) do not change with time in the test because of the low temperature of the cold junction. The increases in the contact resistances in the test are indicated by the increase in the slope of the measured V-I characteristics (Fig. 4a) as well as the decrease in the peak electric power and shift to lower current (Figs. 3a and 3b). Figure 4a-4c show changes in the measured  $V_{oc}$  and Seebeck coefficients of the legs in the MAY-04 with time in the test, suggesting that changes in the material properties of the legs might have occurred. Thus, the measured increases in the total resistances of the legs of MAY-04 with test time could be partially attributed to changes in the electrical resistivity of the leg materials. However, in order to quantify these changes, detailed measurements of the legs resistances, excluding the contact resistances with the metallic

electrodes, are needed. Such measurements would require making 0.5 – 0.6 mm holes in the legs, just below and above the hot and cold metallic electrodes, respectively, which might compromise the structure integrity of the legs and of the metallic coating.

The estimate of the peak efficiency of MAY-04, 96 hrs after BOT, accounts for the measured contact resistances in the test and assume zero side heat losses (solid circle symbols in Fig. 5b). The estimated peak efficiency of MAY-04 of 10.65% is only within 0.37 percentage point of the theoretical peak efficiency (11.02%) calculated at the same hot and cold junction temperatures in the test, but assuming zero contact resistances. Both estimates used the JPL thermoelectric materials properties database. As shown in Fig. 5b, the measured contact resistances, 96 hrs after the start of the test, are as follows: the cold ( $r_{\text{cold-side}}$ ) and hot ( $r_{\text{hot-side}}$ ) contact resistance of the n- and p-legs are 17.4 and 92.8  $\mu\Omega\text{-cm}^2$  and 7.2 and 60.6  $\mu\Omega\text{-cm}^2$ , respectively. Note that the cold contact resistances are very low confirming the quality of the bismuth-lead solder of the legs with the copper cold shoe. The hot contact resistances are also reasonable, considering that the legs of MAY-04 were not brazed to the copper hot shoe. In an actual converter assembly, the legs will be brazed to the hot shoe, which is likely to be molybdenum instead of the copper used in the present test, the hot contact resistances could be much smaller than indicated in Fig. 5b. For the purpose of comparison, estimates of the conversion efficiency of MAY-04 SKU, assuming a total contact resistance per leg,  $r_{\text{cont}} = 150 \mu\Omega\text{-cm}^2$  and zero side heat losses are calculated at the same hot and cold junction temperature in the test and shown in Fig. 5b. The results indicate that such relatively high contact resistances would decrease the peak conversion efficiency to  $\sim 10.16\%$  and shift it to a lower current. This peak efficiency is  $\sim 5\%$  lower than that estimated in the test and  $\sim 8\%$  below the theoretical value of 11.02%.

## SUMMARY AND CONCLUSIONS

Presented and discussed in this paper are the test results of a Skutterudite-based unicouple (MAY-04) with a metallic coating to suppress the sublimation of antimony from the legs near the hot junction. This test was conducted in the ISNPS high temperature facility in vacuum ( $\sim 9 \times 10^{-7}$  torr) at hot and cold junction temperatures of  $892.1 \pm 11.9$  K and  $316.1 \pm 5.5$  K, respectively, for  $\sim 2,000$  hours. To quantify the effectiveness of the metallic coating in MAY-04, the test measurements are compared with those of two uncoated unicouples of the same leg materials (MAR-03 and JUN-03), which had been tested earlier. MAR-03 and JUN-03 were tested in argon cover gas at  $\sim 0.051\text{-}0.068$  MPa for 450 and 1200 hours, respectively. The BOT  $V_{\text{oc}}$  of MAY-04 (204.2 mV) is almost the same as that of MAR-03, but higher than that of JUN-03 ( $\sim 180$  mV). Results indicated that the metallic coating in MAY-04 effectively reduced the performance degradation with cumulative test time. The estimated peak efficiency of MAY-04,  $\sim 96$  hours after BOT (10.65%) is only  $\sim 0.37$  percentage point lower than the theoretical value (11.02%), assuming zero side heat losses and zero contact resistance per leg. The peak power of MAY-04 decreased by only 13.6%, from its BOT value of 1.62  $W_e$  to  $\sim 1.4$   $W_e$  after  $\sim 2,000$  of cumulative testing.

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