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The Selenide Saga: A Contribution Toward a History of the Selenide Isotope Generator

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Abstract

In the mid to late 1970s, the U.S. Department of Energy and its predecessor agencies supported studies on a Selenide Isotope Generator (SIG) that was to power the proposed Jupiter Orbiter Probe Mission (renamed the Galileo mission in 1978). Early studies projected a 1981 generator design with an efficiency of 10.5% (compared to then state-of-practice efficiencies of under 7%) and a specific power of 6.6 We/kg (compared to then state-of-practice specific powers of 4.2 We/kg). Higher efficiencies were promised in the future: 11% (1983) and 13.5% (1985). As the experimental work continued evidence of degradation of the thermoelectric materials began to emerge, leading to the cancellation of the SIG program in 1979. This paper is a contribution to a history of the SIG program with suggestions for lessons that can be learned to aid future radioisotope power source (RPS) programs.

"The disadvantage of men not knowing the past is that they do not know the present."
--*G. K. Chesterton*, All I Survey

1. Introduction

The Selenide Isotope Generator (SIG) program was established circa 1977 to deliver flight radioisotope thermoelectric generators (RTGs) in support of the then planned 1982 Jupiter Orbiter Probe mission (renamed the Galileo mission in 1978). Each RTG was to provide a minimum of 214 We at acceptance. Higher thermal-to-electrical conversion efficiencies were anticipated with these RTGs because they were to "… use a newly developed high temperature thermoelectric material. This material, which is composed of copper, silver and selenium for the p-type and gadolinium-selenium for the n-type, was developed by the 3M Company and its use for space system applications is actively being pursued by the Teledyne Corporation under the sponsorship of ERDA" (Energy Research and Development Administration).[1] [NOTE: Technically the SIG program introduced ***two*** entirely different thermoelectric materials both of which happened to contain selenium.]

Ultimately, the selenide thermoelectric elements did not perform as originally envisioned and work on the SIG program ceased on 29 January 1979 because of a Stop Work Order issued by DOE[2]. Using publicly available documents (since the author had no involvement in the SIG program) this paper aims at being a contribution toward a top-level summary of the SIG program with the view of identifying lessons that can be learned to aid future radioisotope power source (RPS) programs.

(A list of acronyms in provided in Appendix A.)

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2. The Selenide Isotope Generator Program

This section provides a brief description of the Selenide Isotope Generator Program as gleaned from publicly available documents.

2.1 Selenide Isotope Generator

The Selenide Isotope Generator (SIG) proposed for the Galileo mission combined selenide-based thermoelectric materials with a copper-water heat pipe radiator and a modified Voyager-style Multi-Hundred Watt (MHW) heat source.[2] As described in the *Program Final Report* prepared by Teledyne Energy Systems (TES), the advantages of SIG would come from "Predicted thermoelectric conversion efficiencies in the 11% range, resulting in RTG system efficiencies of ~9% and power-to-weight ratios greater than 2 watts/pound" [4.4 We/kg] which "offered a potential for substantial improvement over prior silicon germanium (Voyager) and TAGS/lead telluride (Pioneer) flight units which exhibited system efficiencies of less than 7% and power-to-weight ratios less than 2 watts/pound" [4.4 We/kg].[2] The proposed SIG flight configuration is shown in Figure 1. [The reader should be aware that different values for the various parameters have been reported in the literature. The author has tended to use the values in the TES *Program Final Report*.]

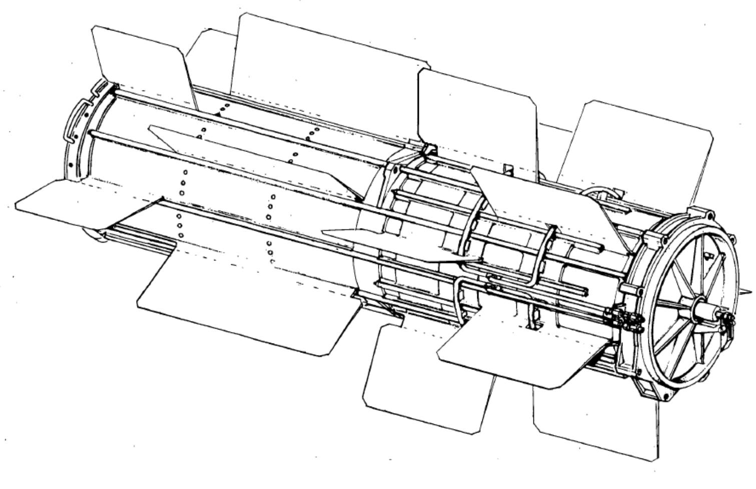

Figure 1. Selenide Isotope Generator RTG Flight Configuration.[2]
The SIG RTG was to be 1.27-m long by 0.62-m across the fin tips with a mass of 46.5 kg.
The Galileo Orbiter was to carry two SIG RTGs, each producing ≥214 We at beginning of life.
The SIG RTG design consisted of two sections: a power section and a radiator extension.

The SIG RTG flight configuration shown in Figure 1 "… is a finned, right circular cylinder 50 in. [1.27 m] long and 24.3 in. [0.62 m] across the fin tips; nominal system weight is 102.5 lbs [46.5 kg]. The 50 in. [1.27 m] –long finned cylindrical housing consists of two approximately equal length cylindrical sections: (1) the power section housing, containing the thermoelectric converter and radioisotope heat source, and (2) the radiator extension, a thin-wall finned, hollow cylinder sized to increase the effective heat rejection area of the power section housing to that required to maintain the nominal 125ºC radiator fin root operating temperature. The two cylindrical sections are joined by riveting. Twenty longitudinal copper-water heat pipes provide the means for high conductance of the unconverted heat from the thermoelectric conversion process to the heat rejection surfaces of the power section housing and radiator extension; ten pipes extend the full length of the generator while ten are the length of the power section housing. Two circumferential water coolant lines, located on the power section housing in the area of the two thermoelectric rings, provide a mean[s] for heat removal during certain ground handling operations and prelaunch/launch operations up to time of spacecraft removal from the Shuttle in earth orbit".[2] Both the power section housing and the radiator extension housing were constructed from what were basically thin-wall Type 6061-T6 aluminum hollow cylinders.[2] Figure 2 shows the Galileo spacecraft cruise configuration with the two SIG RTGs indicated.[2]

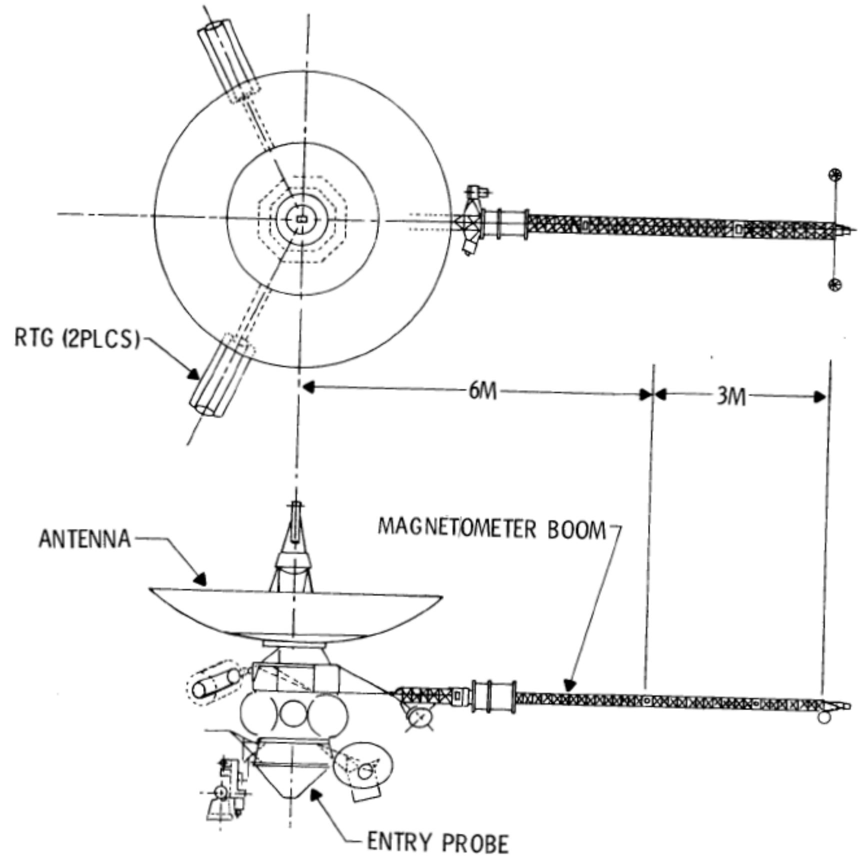

Figure 2. Galileo spacecraft cruise configuration with the two SIG RTGs shown attached on booms.[2] Each SIG RTG was to produce ≥214 We at BOL for a total spacecraft power of ≥428 We.

Major hardware deliverables included[2]

- an Engineering ETG (S/N 1) for test at TES
- a Qualification RTG (S/N 2)
- two ETGs (S/N 3 and S/N 4) for delivery to JPL
- two flight RTGs and one backup flight RTG (S/N 5, S/N 6 and S/N 7)

The flight RTGs were to be delivered to the Kennedy Space Center (KSC) in late 1981 for a planned Space Shuttle launch of the Galileo spacecraft in January 1982.[2]

Each of the two flight SIG RTGs was to produce ≥214 We at beginning of life (BOL) which was "… defined for a selenide system as after 1000 hours of operating time because of a seat-in (improvement) phenomenon associated with the thermoelectrics, on the order of a 10-20% power increase".[2] The power requirement of ≥214 We followed "… directly from the thermoelectric converter efficiency assessment, ≥10.1%, and the minimum system fuel inventory of 2450 watts(t)".[2] [Elsewhere, Tables V-3 and V-4 of Ref. 2 list the MHW heat source thermal inventory as 2460 Wt.] Table IV-1 ("SIG RTG Performance Requirements") of Reference 2 lists the end-of-mission (EOM) power requirement as ≥185 We at 30 ± 0.5 V DC after 58,760 operating hours. The report states that EOM "… time is defined as 50,000 hours after launch with launch occurring 1 year after heatup".[2] TES assigned a reliability of 0.95 at a 50% confidence level of achieving that EOM power. "Fuel decay over this total time span would account for an electrical power loss of 17 watts. Additionally, an allowance of 12 watts was assigned for all degradation effects. Ten of the twelve watts were allocated to 3M for thermoelectric effects and the remaining two watts(s) to TES for other system effects, primarily insulation conductivity change".[2]

The electrical power was to be produced by two selenide-based thermoelectric rings in the power section that would encircle the MHW radioisotope heat source as shown in exploded views in Figures 3 and 4. As described in the TES *Program Final Report*, "Each of the two rings contains 28 six-couple modules, for a total of 336 couples per generator. The couples are connected in a parallel-series arrangement (168 pairs of parallel couples are electrically con[n]ected in series). The thermoelectric couples in each ring are retained in position between the generator housing and a circular graphite hot frame by springs in the module cold end hardware which load the individual N and P couple legs".[2] Various insulators and getters were used as described in Reference 2. While cover gases were employed during ground operations and launch, once in space the SIG RTG was to be vented to operate in a vacuum.[2]

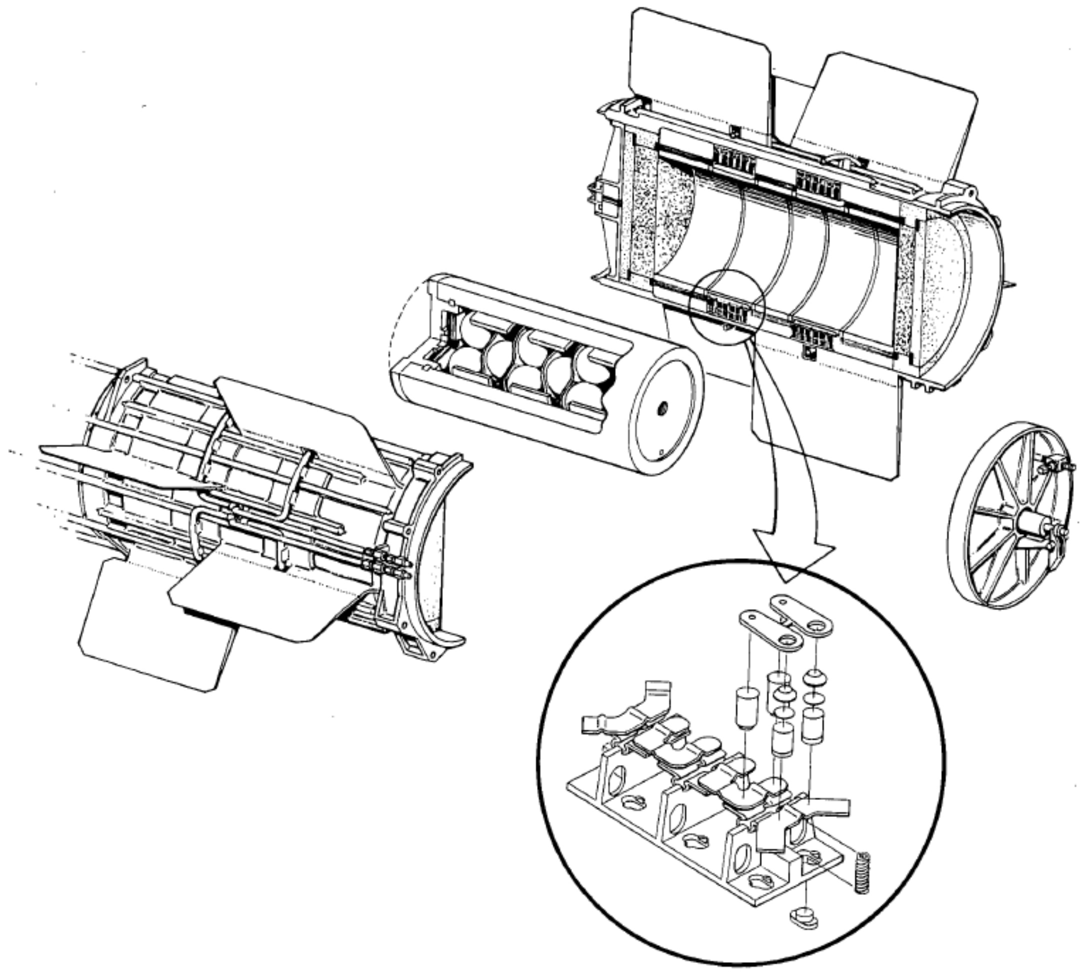

Figure 3. SIG RTG Power Section (exploded view).[2]
Two selenide-based thermoelectric rings encircle the radioisotope heat source.
Each RTG would have a total of 336 selenide-based thermoelectric couples.

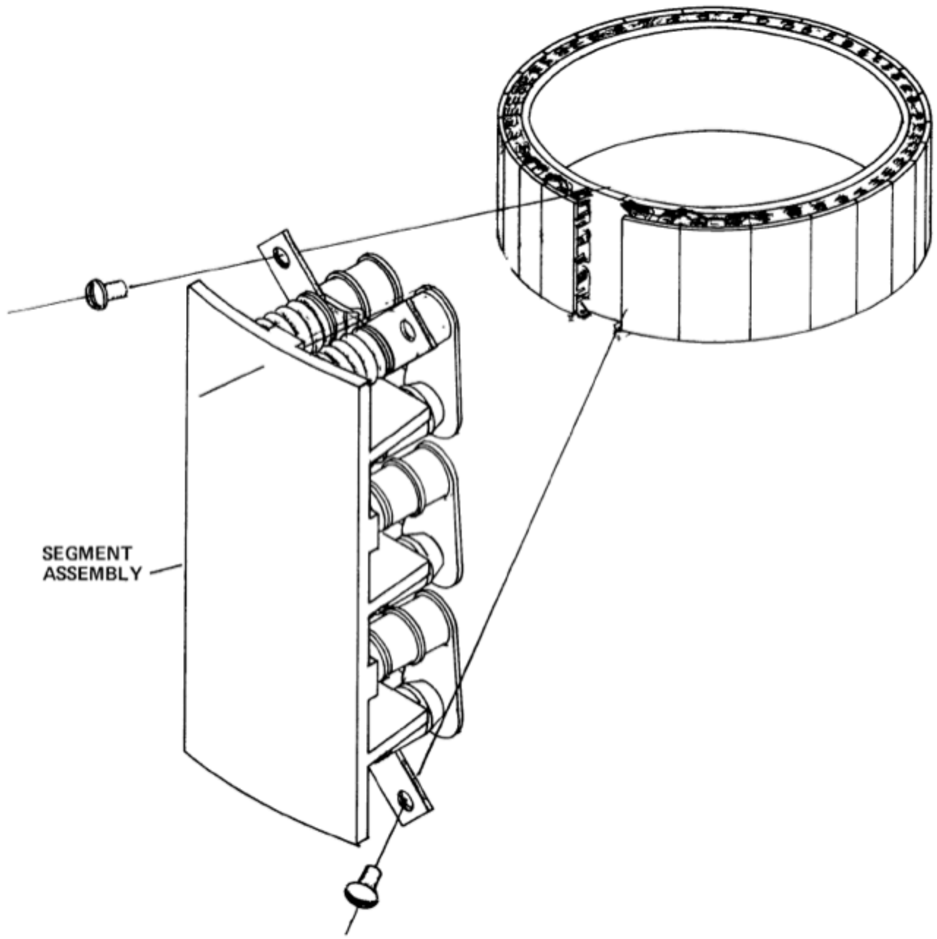

Figure 4. The 3M thermoelectric converter ring assembly (exploded view).[3]
The ring assembly consisted of a POCO support ring with 28 segment assemblies located around its periphery.
The POCO AXF-Q1 cylinder was 7.6-cm long, 19.46-cm inside diameter, and 0.58-cm minimum thickness.
Two converter rings were to be wired in series within the TES housing (see Figure 3.)
The internal circuit connection of the converter ring was to be a series parallel with two parallel strings.

The thermoelectric elements were[2,3,4]

P-leg (TPM-217)	Copper-Silver Selenide ($Cu_{1.97}Ag_{0.03}Se_{1+y}$)	0.76 cm long x 0.65-cm diameter
N-leg	Gadolinium Selenide ($GdSe_x$)	0.76-cm long x 0.76-cm diameter

NOTE: The lengths and diameters are from Table IV-2 of Reference 2. Table IV-4 of Ref. 2 shows a changing set of dimensions, temperatures and efficiencies. Chemical compositions were taken from References 2 and 4.

The hot junction temperature was to be 860 ˚C which "… is one of the major design parameters in determining system performance. Hot junction temperature previously had been specified at 900 ˚C but it appeared prudent to reduce it 40 ˚C based on 3M Company test data".[2] The cold junction temperature was listed as 160 ˚C and the radiator fin root temperature at BOL was listed as 123 ± 3 ˚C.[2] Table IV-4 of Reference 2 lists the 22 January 1979 parameters as 815 ˚C for the P-leg hot junction temperature and 845 ˚C for the N-leg hot junction temperature.

Converter efficiency dropped to 8.4% and the BOL power dropped to 188 We.[2] ["TPM-217" was the designation for the proprietary 3M P-leg material and sometimes "TPM-217" was used as the designator for both legs.]

2.2 Selenide Isotope Generator Participants

The two key contractors in the SIG RTG program were Teledyne Energy Systems (TES) and the 3M Company. (The full list of SIG/GM participating organizations is provided in Appendix B.) Both were, in essence, DOE prime contractors[2], each having its own separate contract with DOE. (The TES contract with DOE was DE-AC01-78ET33009 , formerly ET-78-C-01-2865, and the 3M contract was DE-AC01-78ET-33008, formerly No. ET-78-C-01-7864.) The responsibilities of TES, 3M and DOE were established in a *DOE/TES/3M Interface Agreement for the Selenide Isotope Generator for the Galileo Mission* (GM).[5] While the Interface Agreement permitted DOE to "… have on-site at both TES and 3M a full time DOE program office representative and a quality representative"[5] no mention was made of TES having the same prerogative.

According to the *Interface Agreement*, "Teledyne is responsible to DOE as the SIG/GM system integration contractor. As such, TES will design, develop, fabricate, assemble, test and deliver to DOE the SIG/GM power system in compliance with contractual performance specifications and delivery milestones. Major system components to be supplied to TES as GFE are the processed selenide thermoelectric converters delivered in hermetic shipping containers".[5]

The *Interface Agreement* stated that "The 3M Company will design, fabricate, test and deliver to DOE the SIG thermoelectric converters in accordance with the approved specifications. The 3M Company will provide all necessary analyses, data and drawings to the DOE such that a successful interface between TES and 3M hardware can be accomplished and that performance and reliability of the SIG system can be predicted and verified in accordance with the User's requirements and specifications".[5]

3. Selenide Isotope Generator History

The TES SIG *Program Final Report* stated that "The SIG design was initiated in 1973 from a general study contract to evaluate various RTG concepts, Refs. [6-8], with the emphasis on Cm-244 fuel. Additional design studies were performed in 1974 and 1975 with the Pu-238 fuel form, Refs. [9-12]. Then in 1976, a major redirection was given to use the developed MHW heat source. Studies with this constraint were presented in Refs. [13-15] and formed the basis for the design proposed for the Galileo mission contract. These studies also were used to formulate the contractual performance objectives for the Galileo mission".[2] [NOTE: The reference numbers in the quote have been adjusted to align with the reference numbers in this paper.]

The interest in the selenide-based thermoelectric elements appears to have come from early experiments and studies showing the improved performance of the selenide materials over existing RTG thermoelectric materials. For example, one 3M report stated that "… the selenide materials offer the potential for far superior performance over the Silicon Germanium systems" and noting that "… the Silicon Germanium system has as a characteristic an unavoidable and predictable degradation mechanism that brings about a significant loss in efficiency over a five year period".[3] [For comparison, it should be noted that the silicon-germanium-based MHW-RTGs on the two Voyager spacecraft are still providing power over 38 years after launch and Pioneer 10 with its lead telluride/TAGS thermoelectric elements sent signals over 30 years after launch.]

A 1977 JPL paper stated that "The major factor in selecting the use of the selenide thermoelectric material is its improved conversion efficiency. This improvement is primarily a result of this material's much lower thermal conductivity. The successful long-term operation of an RTG depends strongly on the stability of the electrical and thermal properties of the thermoelectric material. The long-term behavior of the thermal conductivity, therefore, becomes of vital concern."[1]

3.1 Selenide Thermoelectrics – The Beginnings

Beginning in January 1968, the U.S. Atomic Energy Commission's Division of Space Nuclear Systems supported an advanced technology program related to the TPM-217 materials at the 3M Company.[16] 3M reported that some in-gradient tests had been run for up to 10,000 hours and that "Excellent long-term stability is being demonstrated".[16] An efficiency of 12.9% was listed for the P material with the anticipation that the N material would be at the same level of development as the P type by about the end of 1972. Total couple performance characteristics for long-term operation were expected by the end of 1973.[16]

On 25 September 1972, a patent application was filed for "Thermoelectric Generators Having Partitioned Self-Segmenting Thermoelectric Legs" listing Edward F. Hampl, Jr., William C. Mitchell and Robert S. Reylek as the inventors and assigning the patent to the U.S. Atomic Energy Commission (AEC) because the invention "… was made in the course of, or under, a contract with the United States Atomic Energy Commission" (U.S. Patent Application No. 291,938). The patent was granted on 25 March 1975.[17] The patent cited as an example "… a P-type thermoelectric leg consisting of about 65.5 atomic percent copper, 1 atomic percent silver, and 33.5 atomic percent selenium …" The patent described how a thermoelectric element with selenium and copper could be partitioned with a thin barrier to prevent the migration of copper from the hot end to the cold end. Partitioning was necessary because the migration of copper was found "… to be responsible for certain problems that limit usefulness of thermoelectric legs experiencing the gradation".[17] Without the partition "… the hot end of a copper-silver-selenium leg …" would "… undergo creep-deformation after a period of sustained power-generating operation … after 100 hours of such operation [≥800 ˚C] the diameter of the hot end of the leg might increase by as much as 15 percent".[17] As copper migrated toward the cold end the hot end was left with a higher proportion of selenium which made the composition more susceptible to a creep-deformation.[17] "While a copper-silver-selenium thermoelectric leg … has a very low vapor pressure in isothermal tests, surprisingly, when the leg is operated at matched load in a thermoelectric generator with the hot end heated to 800 ˚C or higher, there is a significant loss of selenium from the hot end of the leg, which causes the operation of the leg to be unstable. Again, it has now been found that this loss of selenium can be traced to the migration of copper atoms in the composition which increases the proportion of selenium at the hot end …"[17] It was noted that some of these problems "… could be avoided by not heating the hot end of the leg to the described temperature, but the result of such a procedure would be a reduced efficiency of power-generation by the leg".[17]

By 1974, an AEC representative stated that "One of the most exciting developments in thermoelectric technology is the advent of the selenide thermoelectrics. TPM-217, the P-leg of this couple, is now ready for system application …", noting that "A unique feature of TPM-217 in contrast to other thermoelectric materials is that the mobility of a major constituent, copper, is high. The copper responds to electrical and thermal driving forces in very short times and quickly reaches a steady state. An interesting result of this steady state condition is that the dopant concentration increases significantly from the cold to the hot end. Thus the selenide infinitely segments itself in the temperature gradient"[18]. Furthermore, it was reported that "TPM-217 has demonstrated stable performance for over three years of ingradient testing … Preliminary efficiency measurements indicate couple efficiencies utilizing the current vintage selenide materials to be about 9%, with growth potential to about 11% at 800˚C hot junction temperatures".[18]

This view was reinforced in 1976 when an ERDA representative called attention to "A significant increase in RTG performance … has now been made possible with the discovery and development of a new class of thermoelectric materials, the selenides. These high temperature, high conversion efficiency materials comprise p-type thermoelectric materials composed of copper, silver, and selenium and n-type compositions composed of gadolinium and selenium. These materials exhibit excellent thermoelectric properties over a broad temperature range that extends to 1000˚C, allowing the technology of thermoelectric generators to advance to the point that RTG efficiencies could about double while their specific power (watts/pound) could improve by at least 50% over existing systems".[19]

By 1977, a Selenide Isotope Generator concept had been developed as part of the Low Cost High Performance Generator technology program that had begun in 1973.[14] Performance characteristics were established for three time intervals: 1981, 1983, and 1985. It was reported that "The 1981 generator has been designed for the Jupiter Orbiter Probe Mission and exhibits an efficiency of 10.5 percent, a specific power of 3 watts per pound … for a nominal 250 watt electrical unit … The 1983 system promises efficiencies of 11 percent, specific power of 3.5 watts per pound … In 1985 system efficiencies of 13.5 percent, specific power powers of 3.8 to 4.5 watts per pound … can be expected”.[14]

In 1978, it was stated, “The selenides, as a class of highly efficient thermoelectric materials, have been discussed in the open literature …”[20] and four references were cited [References 21-24 in this paper]. The results of testing of the P-type composition $Cu_{1.97}Ag_{0.03}Se_{1.0045}$ and N-type compositions $GdSe_{1.49}$ (designated TPM-217) “… over a broad temperature range that extends to over 1000˚C” were reported to “… have values of figure of merit and conversion efficiency that are superior to those of prior art compositions both in the low temperature portions and in the high temperature portions of these large thermal gradients. This high conversion efficiency advances the technology from a system efficiency of 4 to 6% to values in the range of 9 to 13%”.[22] [NOTE: Reference 22 states that both the P-type material and the N-type material were referred to as “TPM-217”.]

Attention was called to the fact that “P-type TPM-217 is fundamentally different from standard thermoelectric materials in that the dopant concentration is itself a function of the applied thermal and electrical gradients. The dopant level at an arbitrary position within the leg is not fixed by the original processing of the leg … but instead varies rapidly with the applied current and thermal gradient … It is essential to understand these effects to predict both the short-term thermoelectric properties and the long-term thermodynamic stability of p-type TPM-217”.[22]

3.2 Selenide Thermoelectrics – Early Issues

In 1976, at the request of ERDA, RCA researchers investigated the proposed selenide thermoelectric material using their own knowledge of what to expect on the behavior of the compositions being considered in their proposed operating environment. They concluded that the selenides would not succeed.[25]

It was noted in 1977 that “One major difference of this new thermoelectric conversion material is that the ‘n’ and the ‘p’ leg of a [thermoelectric couple] are made of a different alloy rather than being merely positively or negatively doped identical materials as was the case for the Pb/Te and the Si/Ge systems”.[26] This meant that the P material and the N material would have their “… own different set of characteristics”, making studies more difficult, particularly when only the P material was then made available to JPL for independent testing.[26] It was further noted that in the case of the copper-selenides, the selection of the desired thermoelectric material composition “… becomes somewhat more complicated; this material possesses the peculiarity of having its composition altered, or modified, by the presence of a current gradient across the material. In addition, the composition will also slightly depend upon temperature gradients. Because of this, not only do the thermoelectric performance parameters, such as Seebeck voltage, resistivity and thermal conductivity, depend on temperature and an initial alloy composition, but they are also subject to large modifications by current gradients and temperature gradients. This peculiarity of the material poses an additional complication in evaluating the performance data from a single element to a full-up generator. Since ‘standard’ property data are not really ‘standard’, but are subject to gradients, which in turn are dictated by geometry and temperature profiles, the design as well as the performance prediction of an RTG becomes rather involved”.[26]

The P leg which JPL examined in 1977 had a partition made of a tungsten foil coated with copper on the side facing the hot side (the possible need for a partition was mentioned in the original patent, see Section 3.1 and Reference 17). The 1977 JPL report stated that “The reasons for partitioning the p-leg and thus changing the composition of the leg are twofold: 1) the mechanical properties, namely the creep strength of the material, degrade with increased selenium content; 2) the weight loss rate due to sublimation increases with an increase in excess selenium. Both of these mechanisms are highly undesirable, particularly for high temperature applications. In addition to the mechanical and thermophysical property changes, the basic thermoelectric properties, i.e., Seebeck voltage, resistivity and to a minor extent the thermal conductivity, also change as the selenium composition is varied”.[26]

In addition to the usual problems of materials, coatings and insulation, the SIG program ran into sublimation issues with the P-leg and cracking with the N-leg.[2]

From the conceptual design update in June 1978 to the technical review meeting on 22 January 1979, the changes listed in Table 1 were made in the thermoelectric converter parameters.[2] Table 1 shows that the design of the thermoelectric elements was in flux and that the temperature was being reduced to accommodate sublimation (and other) issues with the thermoelectric elements. This, in turn, led to a drop in efficiency and in power.

Table 1. Chronological Comparison of S/N 1 Thermoelectric Converter Parameters[2]

Parameter	Conceptual Design Update 22Jun1978	PDR 30Nov1978	Technical Review Meeting Status 22Jan1979
T_{HJ}/T_{CJ} (˚C)	860/160	860/160	815(P)/160 845(N)/160
DN/DP (cm)	.772/.645	.800/.660	.508/.660
LN/LP (cm)	.762/.762	.762/.762	.762/.663
Voltage (V)	30	30	30
Couples	336	336	336
Converter Mass (kg)	7.3	6.4	7.3
Converter efficiency (BOL)	10.1	10.2	8.4
RTG power output (We) (BOL)	229	230	188

The TES final report stated that "… a number of changes were incorporated by 3M into a proposed new reference design …" that included "… a skewed and lower hot junction temperature, a segmented N-leg, unpartitioned P-leg, reduced P-leg length, and lower power output".[2]

All of the work would come together in the Ground Demonstration System (GDS) as discussed in Section 3.3.

3.3 Ground Demonstration System (GDS-1)

As described in the TES SIG final report, "The GDS-1 was the first attempt to design, fabricate and assemble a large-scale thermoelectric generator employing the 3M Company selenide thermoelectric materials technology. The generator was designed and built under the SIG development program and was tested under the subsequent SIG/Galileo flight program".[2] Table 2 summarizes the generator design features. Figure 5 shows a cutaway of the GDS (the GDS-1 heat pipe fin was not installed on the GDS, rather it was tested separately).[2]

Table 2. Ground Demonstration System (GDS-1) Design Summary[2]

Output power objective (We)	108 at 13.5 V
Number of thermoelectric couples	156
D_n/D_p (cm)	0.900/0.660
Hot junction temperature (˚C)	850
Cold junction temperature (˚C)	150
Housing/Radiator temperature (˚C)	125
Gas fill	Xe/He mixture
Extraneous thermoelectric module resistance (%)	25
Spring pressure, N/P (MPa)	2.07/1.03
Element length (cm)	0.76
Circuit	2 strings, series/parallel
P-leg	Unwrapped, one partition
Cold end hardware	Sliding follower
Heat source	Electrical

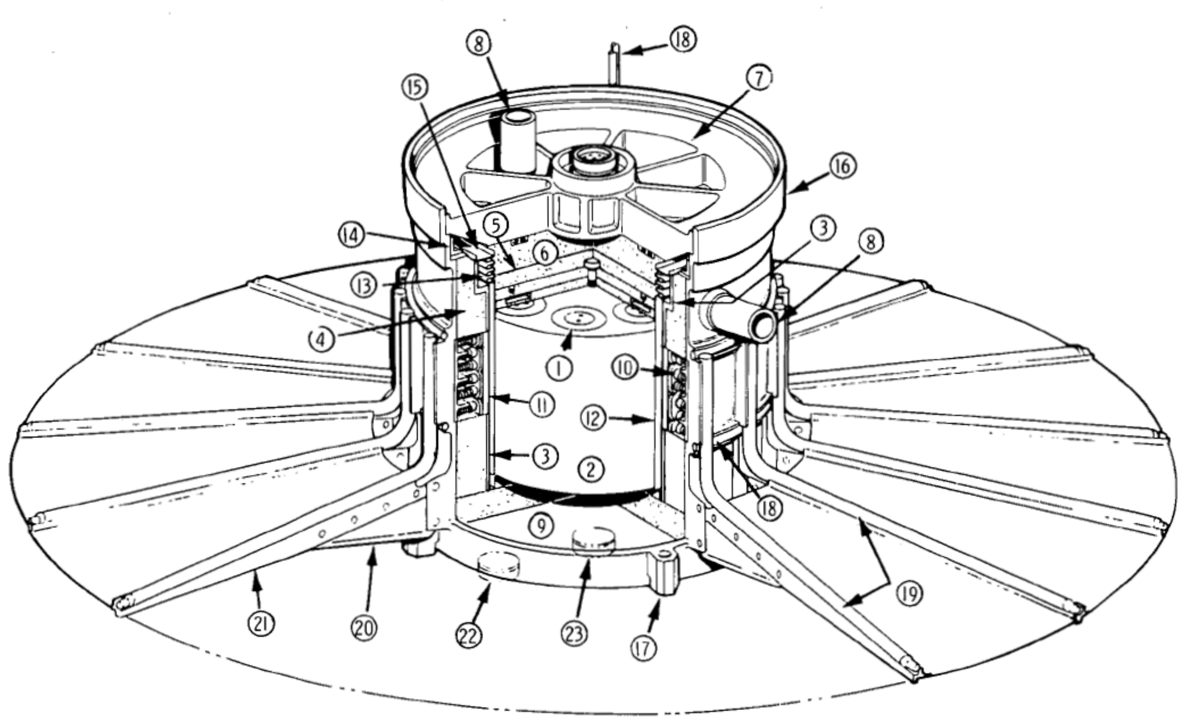

Figure 5. Selenide Isotope Generator (SIG) Ground Demonstration System (GDS-1)[2]

1. Electrical Heater Element
2. Electrical Heater Body
3. Insulation Retainer Barrier Sleeve
4. Side Insulation Rings
5. End Insulation High Temperature
6. End Insulation Low Temperature
7. End Cover With Heater Power Input Connector
8. Outgassing Port
9. End Cover With Power Output & Instrumentation Connectors
10. Thermoelectric Module
11. Thermoelectric Module Support Ring
12. Isolation Hot Frame (I.H.F.) Liner
13. I.H.F. Bellows
14. I.H.F. Bi-Metal Seal Ring
15. I.H.F. Transition Ring
16. Housing Shell
17. Mounting Lugs
18. Auxiliary Cooling System
19. Heat Pipe
20. Fin Support Gusset
21. Fin Structure
22. Instrumentation Connector
23. Power Output Connector

As reported in the TES SIG final report, "The original plan for GDS-1 called for outgassing and then seat-in of the thermoelectric module after which performance characteristics of the generator would be determined. This was to be followed by a life test and finally a random vibration test. However, during the seat-in operation it was observed that the normalized power output peaked at 86.7 watts on July 26, 1978 and then began to decrease uniformly at about 0.2 watts/day. This rate of degradation continued for about 25 days; then the decrease became greater and at the same time more erratic. Finally on September 12, 1978 the generator open circuited. By manipulating the electrical load conditions, the generator could be brought back on load for intermittent periods".[2]

Subsequent to this, GDS-1 was cooled down and disassembled. While the TES SIG final report stated that it was "… not possible to relate the observations to direct causes for the degradation" some observations could be made. These included that a "… significant amount of sublimation of the P-legs [had] occurred during the relatively short life of 2000+ hours as shown by the bullet nosing of the legs and deposits on the cold end hardware…" and that “All exposed N-legs display[ed] cracks and/or chips”.[2] It was also noted that “A great deal of misalignment of both N and P-legs was seen both visually and with radiographs”.[2]

Table 3 lists other observations made during disassembly.

Table 3. <u>Observations on Exposed Legs of Ground Demonstration System (GDS-1) Module</u>[2,27]

- Misalignment with hot/cold shoes
- Legs titled away from the radius
- Deposits on cold straps and followers
- Orange ring around N-legs (one ring appeared white)
- Cracks/chips in N-legs (longitudinal and transverse)
- Copper extrusion at cold end of P-legs
- Bridging around P-leg partition
- Deposits on edges of BeO discs
- Dark crystalline deposits on hot end of P-leg
- Bullet nosing of hot end of both P-leg segments
- P-leg hot segment has larger diameter than cold segment
- Mushrooming of P-leg just below partition
- Individual instance of P-leg undercut at cold end and with dark purple ring around cold end

Although the TES SIG final report stated that “… no definite conclusions can be made at this time concerning the cause for the rapid degradation of performance, several observed conditions within the module listed in Table [3] could possibly contribute to that fact.[2] They are:[2]

- Cracks in N-legs (increased resistance)
- Deposits on edges of BeO disc (shorting of thermoelectric circuit)
- Bullet nosing of P-legs (increased resistance)

Reference 27 is the final TES disassembly report for GDS-1 containing more detailed data and pictures.

In the midst of these issues, JPL voiced its concerns about the selenide thermoelectric material.[29]

On 29 January 1979, a Stop Work Order was issued by DOE.[2,3]

4. Observations on the Selenide Isotope Generator Program

From this distance in time and not having any involvement in the SIG program the author's observations are based on available documents. Still, with that constraint, some general observations can be made and these are divided into (1) management and (2) technical in the following sections.

4.1 Management Aspects of the Selenide Isotope Generator Program

The contractual structure for the SIG program differed from some earlier RTG programs. For example, on the SNAP-27 program and the MHW-RTG program, the system contractor (General Electric) was the prime contractor who, in turn, subcontracted for the thermoelectric work (to 3M for SNAP-27 and to RCA for the MHW-RTG). In the SIG program, DOE contracted with TES and 3M directly which, in a sense, made the two contractors coequal. Ideally one would like to have the system contractor be the system integrator with the overall responsibility and authority to manage the program answerable to the government for deliverables, schedule and budget.

The SIG interface agreement solidified this coequal contractual arrangement and did not specifically mention that TES could have a site representative at 3M.[5] A flight RTG program such as SIG, which had a very tight schedule, requires close coordination. Onsite representatives can often expedite resolving interface issues.

If, as seems to have been the case with the SIG program, a DOE Headquarters person was the overall program manager, then DOE needed the resources to fulfill that role. Specifically, the DOE contracts with TES and 3M should have been managed out of DOE Headquarters to facilitate coordination between the program manager and the contracting officer.[28]

Equally important is having a tightly knit government management structure.[29] In the case of the SIG program, program management appears to have been centralized at DOE's Germantown Maryland site although DOE field office personnel were involved because the contract had been moved to the field.[28] For those programs involving two or more government agencies, it is essential that a joint program office and joint project office be established in which the program and project people are colocated. Historically for RTG programs combining the program and project management functions has worked very well.[29]

A classic example of the successful integration of a space nuclear program involving two agencies (NASA and AEC) was the NERVA program where it was determined that "A single program/project organization was mandatory for efficient and effective program implementation".[30] The joint program/project organization was necessitated by the fact that "… it was impractical to separate reactor development from the development of the nonnuclear engine components because of the interactions and critical interfaces that existed among components of a rocket engine".[30] A simplified and updated version of the nuclear rocket program organization (from Figure 2 of Reference 30) is shown in Figure 6 below. A key feature of the nuclear rocket program organization was that people from both agencies (AEC and NASA) were colocated and they had badges and security clearances from both agencies. Instead of what was then the NASA red badge or the AEC green badge, it was said that the government nuclear rocket people had "purple badges".

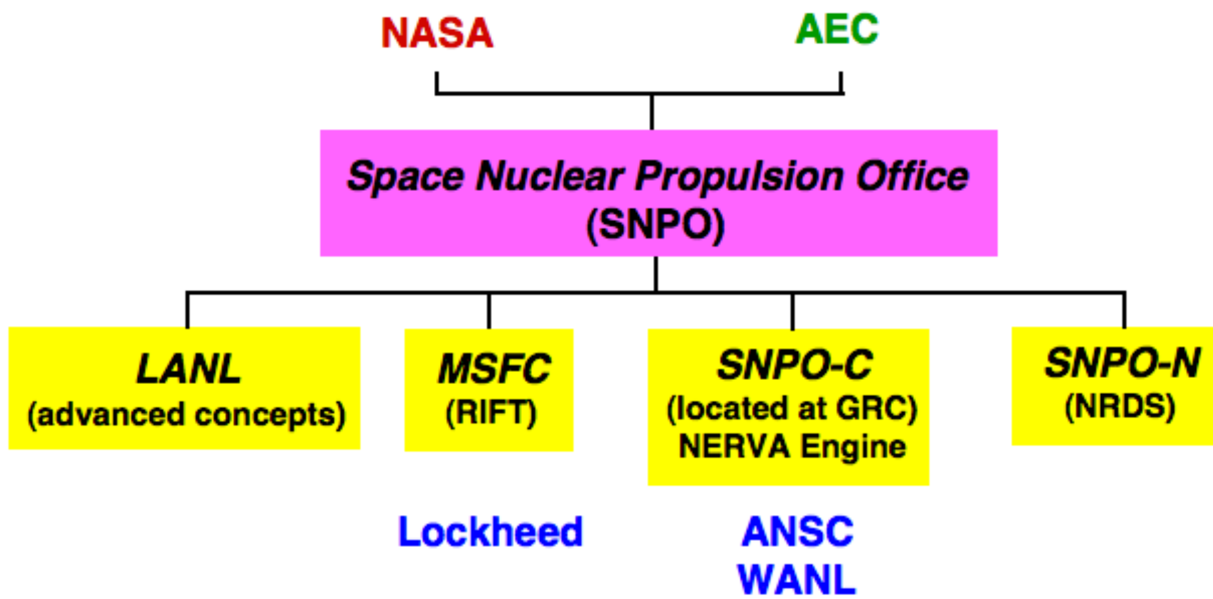

Figure 6. Nuclear Rocket Program Organization.
(Redrawn, updated and simplified from Reference 30.)
The acronyms are defined in Appendix A.

A combined program/project organization with colocated personnel from the participating agencies can overcome the sorts of problems that reportedly afflicted one program in which the headquarters officials were located at their respective agency headquarters, the contract was managed at a government field office, the system project office was at one government laboratory and the nuclear project office was at another government laboratory. Thus, when the system contractor submitted a change order it had to go to the government field office, then to the system project office then to the nuclear project office then back through the chain and to headquarters where more discussions could take place among the involved agencies. It was rumored that this convoluted management structure once led to months passing before a change order was finally approved. Flight programs (or even technology programs with fixed budgets) cannot be run on a 8-to-5, five-day workweek.

In any undertaking like the SIG program, we need, as the late system scientist C. West Churchman wrote, "… to set down the explicit steps that we will be willing to take and capable of taking when plans fail. This is perhaps one of the most neglected aspects of the system approach to design and planning. The planners are often far too optimistic about their success so that when failures occur they are in no position whatsoever to take the necessary steps because they have never thought about them before. In other words, to reiterate the point, *when you postpone thinking about something too long, then it may not be possible to think about it adequately at all*."[31] [Emphasis in original.]

As an example of what Prof. Churchman was describing, following the completion of the MHW-RTGs for the Voyager program, the silicon-germanium alloy (Si-Ge) thermoelectric production line at RCA was shutdown. Various reasons have been given for the withdrawal of RCA from the RTG program but despite those reasons something should have been done to maintain a backup thermoelectric capability until the selenide thermoelectric elements had been flight qualified. At the very least, a high-level formal DOE review should have been conducted that identified viable fallback options that could be quickly activated should the selenide technology not pan out. When it became clear that the Selenide Isotope Generator would not be available, leftover MHW-RTGs were proposed for the Galileo mission.[28] In the end, DOE was forced to resurrect the Si-Ge thermoelectric technology (this time at GE) for both Galileo and the International Solar Polar Mission (which became the Ulysses mission) with a new, higher-powered (300-We) RTG known as the General-Purpose Heat Source Radioisotope Thermoelectric Generator (GPHS-RTG).[32] [Sadly, as if to prove once again Hegel's observation that we learn nothing from history, following the GPHS-RTG production campaign in the 1990s, DOE eliminated the Si-Ge option for a second time leaving the U.S. without a high-temperature, high specific power RTG.]

Judging from the publicly available papers and reports, the Selenide Isotope Generator program appears to have been initiated by technologists (which brings to mind the observation of a longtime technologist: To have your technology adopted by a flight program you must give it up). The requirements of a flight system are usually more rigorous than the goals of a technology development program.[33] For example, in the MHW-RTG program it was initially thought that the years of technology development by RCA of the Si-Ge Air-Vac thermoelectric elements (which GE would term “unicouples”) would make a flight application easy.[34] As it turned out, when the flight program was initiated a number of problems were identified that required immediate fixes.[35] Any technology being considered for flight needs to be scrubbed by experienced people with a space system background. As will be discussed in the next section, it is very likely that had a serious investigation of the selenide technology been undertaken by experienced spacecraft engineers the selenide saga might not have turned out the way it did.

Clearly, there was an inadequate evaluation of the Technology Readiness Level (TRL) of the selenide technology.[28] As the *NASA Systems Engineering Handbook* states, “*It is impossible to understand the magnitude and scope of a development program without having a clear understanding of the baseline technological maturity of all elements of the system.* Establishing the TRL is a vital first step on the way to a successful program.”[33] [Emphasis in original] Based on the available SIG reports, it appears that the Technology Readiness Level for SIG was probably 2 (or less) (“Technology concept and/or application formulated”). For a mission-enabling component of a critical national flight system, the TRL should ideally be at least 8 (“Actual system completed and ‘flight qualified’ through test and demonstration (ground or flight)”).

There is a tendency for projects to compensate for the lack of the necessary technical and flight experience by employing committees and advisory groups. When I see organization charts embellished with advisory committees and review groups, I am reminded of the observation given to me by the NERVA systems engineer Clark Archer: “Collective wisdom does not flow from pooled ignorance”.

At the beginning of my career in management, I was given two interrelated pieces of advice: (1) before you can manage something you have to understand it and (2) you need to penetrate a program to understand it (“the devil is in the details”). Granted, from our present vantage point over 35 years away from the SIG program, we have 20/20 hindsight, yet there were warning signs in the selenide technology development and in the early stages of the SIG program that warranted a deeper technical penetration into the program. In the deluge of technical information and the push to meet a schedule there is a tendency to believe in a technology and to ignore any warning signs. A person can become “invested” in the technology (psychologists refer to this inability to see negative results as “confirmation bias” which traces back to a 1960 study).[36] There is a vast literature on how humans will believe even in the face of disconfirming evidence (see, for example, References 37 and 38). In the face of this “cognitive dissonance”, it is helpful to consider the view of the late history professor Peter Viereck who said, “I can think of nothing more gallant, even though again and again we fail, than attempting to get at the facts; attempting to tell things as they really are. For at least reality, though never fully attained, can be defined. Reality is that which, when you don’t believe in it, doesn’t go away”.[39]

4.2 Technical Aspects of the Selenide Isotope Generator Program

The 1972 patent application for the selenide thermoelectric material highlighted a number of operational issues including migration of copper, creep deformation, and loss of selenium.[17] Of particular note is that unlike most thermoelectric materials in which holes and electrons move, the copper-silver-selenide material operated by ionic conduction, i.e., the copper itself moved.[28] This deviance from past experience should have been a warning flag that much more testing was needed before committing the material to a flight program. This point was emphasized in a 1977 report from JPL to DOE which listed a number of issues and concluded, “From the information which has been presented in this report, it can readily be realized that the successful application of the selenide thermoelectric materials will to a large extent hinge on a thorough understanding of the exact behavior of this system”.[26]

While the 1972 patent application described the migration or gradation (termed “self-segmenting”) of copper as beneficial because “… it automatically achieves the variation in level of current carriers that was previously obtained only by mechanically assembling discrete thermoelectric leg segments that included different levels of doping agent"[17], in practice the copper would "extrude" from the thermoelectric element and "… form something akin to 'steel wool' like copper metal strands".[28] The author has heard these copper metal strands described as "whiskers". Whiskers are to be avoided because they can cause electrical shorts and become a source of debris and contamination.[40]

Two ways to minimize the extrusion of copper would be to maintain the cold side temperature of the thermoelectric elements at or above 150 ˚C[29] (as was done) or to reduce the hot side temperature.[17] However, as discussed in Section 3.2 and Table 1, such changes can affect the efficiency. From the changes proposed in Table 1, it is clear that there were issues with the selenide thermoelectric elements that necessitated lowering the hot junction temperature.

Material loss was a major problem identified and fixes were identified. As noted earlier, the thermal and electrical gradients produced a copper gradient by driving the copper away from the hot end. This, in turn, led to a higher concentration of selenium which tended to evaporate at the operating temperatures causing a solid-state precipitation of copper. Partitioning (which was discussed in the 1972 patent application) could alleviate the problem at the cost of a decreased efficiency and the possible increase in the copper concentration at the partition.[26,41]

In a recent review of the selenide work, Brown et al. concluded "If Cu_2Se is to be used in thermoelectric generators, these problems must be solved or evaded. Possible solutions would be the development and use of different diffusion barriers and contact materials, and the operation of the material only at lower temperature. The authors' own work shows that physical degradation of Cu_2Se can be induced with currents similar to those needed to build a practical thermoelectric generator".[42] [From the documents reviewed, the author is not persuaded that Reference 41 led to the demise of the SIG program as implied by Brown et al.[42] More likely the failure of GDS-1 led to the demise of the SIG program.]

In 2014, Dennler et al., which cited Brown et al., discussed the SIG program in the context of whether or not copper selenides are really new thermoelectric materials and concluded, "Thus, after more than 14 years of research activity, the Cu_2Se based TEG program at 3M was terminated because of the very same intrinsic property of the materials identify by [S.-Y. Miyatani and Y. Suzuki] 25 years earlier".[43]

All of which reminds me of the observation of the late Stan Szawlewicz: "The half-life of technical information is about seven years". After about seven years technical people forget what was done and reinvent the technology.

Concluding Remarks

The Selenide Isotope Generator (SIG) program began with the admirable intentions of improving both the efficiency of radioisotope thermoelectric generators and increasing the specific power (We/kg). These goals are still important to the U.S. space science program.[44,45] Yet in developing the technologies to meet these goals, researchers need to keep in mind the lessons of the SIG program (and other technology programs that did not achieve their goals).

Warning signs that the selenide materials were unlike previously used thermoelectric materials appear not to have been heeded to the degree needed. Even though the SIG program was under a tight schedule the issues that were unresolved had been identified years earlier; in fact, some of those issues had been identified almost a quarter of a century earlier.

[It is worth noting that the SIG N-leg TPM-217 material, $GdSe_x$, and the material $LaTe_x$ (which is currently being considered for future RTG applications) are members of exactly the same thorium phosphide (Th_3P_4) structure type[46] and they utilize elements from the same chemical groups (selenium and tellurium are both Group 6A while both gadolinium and lanthanum are members of the Lanthanide Series).]

The original goals of the SIG program raise the question about the goals proposed for future radioisotope power source (RPS) programs. For example, the SIG program began with the goal of providing ≥214 We (at an RTG efficiency of ~9% and a specific power of 4.4 We/kg) and ended at a projected power of 188 We.[2] Efficiencies and specific powers were to be greater than those of (then) state-of-practice RTGs but in the end (see Table 1) the Selenide Isotope Generator had a lower power and a lower projected specific power than the 300-We GPHS-RTG with its state-of-practice thermoelectric elements (see Ref. 32 for comparison).

There is a tendency to overstate the benefits of a proposed new technology and to compare the new technology too favorably against existing technologies. Admiral Hyman G. Rickover highlighted this in a famous comparison of "academic reactors" and "practical (real) reactors". Admiral Rickover stated that "An academic reactor or reactor plant almost always has the following basic characteristics: 1) It is simple. 2) It is small. 3) It is cheap. 4) It is light. 5) It can be built very quickly. 6) It is very flexible in purpose. 7) Very little development is required … 8) The reactor is in the study phase. It is not being built now".[47] To counteract the tendency to exaggerate the attributes of paper concepts, Rickover urged the nuclear community "… to state the facts as forthrightly as possible".[47]

From the SIG experience, where goals were not achieved despite the expenditure of precious resources, one could argue that to make it worth the effort in advancing RTG technology the goals should be an efficiency greater than 9% and a specific power greater than 5.5 We/kg (which was the specific power for GPHS-RTG F-1, see Ref. 32). The argument for dynamic conversion (e.g., Brayton, Rankine, Stirling) hinges on two factors: higher efficiency (hence using less Pu-238) and the potential for power growth. To make a significant dent in the use of the limited store of Pu-238, it would seem that the goal for a dynamic conversion RPS should be to have efficiency somewhere in the range of 15% to 30% (the higher the better, obviously). While important, specific power may not be a primary requirement for a dynamic conversion system if the desired high efficiency, high reliability and lifetime can be achieved. The lifetime goal for RPS should be at least 10 years (which is what the Pioneer SNAP-19 RTGs and the Si-Ge RTGs have demonstrated).

Both thermoelectric and dynamic conversion systems present challenges in determining the reliability and the lifetime. The ability to accelerate the aging (e.g., testing to higher temperatures or higher dynamic environments) may not be applicable to thermoelectric and dynamic conversion systems. For example, in the MHW-RTG program we found that raising the test temperature in an effort to accelerate the aging produced effects (e.g., chemical species moving about the converter) that would not occur during normal operation. Dynamic conversion systems face similar constraints in aging tests; e.g., it makes no sense to spin a turbine-alternator faster in the hope of accelerating the aging.

The experience gleaned in the NERVA program may offer some approaches.[48] A number of components and subsystems can be tested with careful attention paid to the statistical distribution of the properties of the test articles. Data from related hardware can be examined and Monte Carlo analyses performed, all with the goal of developing the degree of stress/strength overlap (see Figure 7).

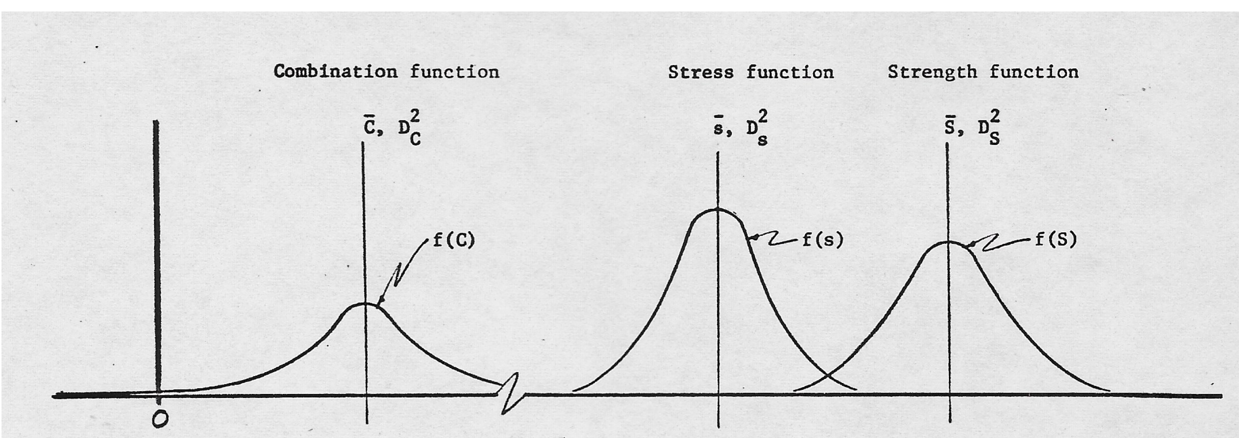

Figure 7. Diagram of the stress-strength interference method for calculating structural reliability.[48]

A key element of an RPS test program should be independent testing conducted by an independent organization. Independent testing by JPL, for example, was helpful in the MHW-RTG and GPHS-RTG programs. Just having a second suite of test chambers and instruments coupled with the proverbial “second pair of eyes” can greatly enhance the credibility of the overall test program.

It cannot be emphasized too much that testing must be in the relevant environment. Just operating an RPS at the same temperature for thousands of hours may not uncover problems associated with a changing mission environment (e.g., lunar day-night cycle). Heat source interactions must also be accounted for.

The difficult effort to recreate the Si-Ge thermoelectric production capability along with the issues associated with restarting the telluride/TAGs production following years of downtime argue for having a facility dedicated to producing thermoelectric elements so the country doesn’t have “to keep reinventing the wheel”. Perhaps, like government wind tunnels, the thermoelectric facility could be made available to RTG contractors who want to produce thermoelectric elements for their flight RTGs. When flight production is not needed, such a facility could be used for research to advance thermoelectric technology, to maintain skills and to keep the equipment in an operationally ready state.

If the objective is to power a mission, the RPS technology development program must be aimed at that objective which means very close interactions with the mission. Figure 8 illustrates the systems engineering and configuration management procedure employed in the NERVA program. NERVA was guided by the NERVA Program Requirements Document (NPRD) which listed the technical and programmatic requirements and described the missions. The NRPD also developed the design philosophy and principles which were to be used in designing and developing the engine.[49]

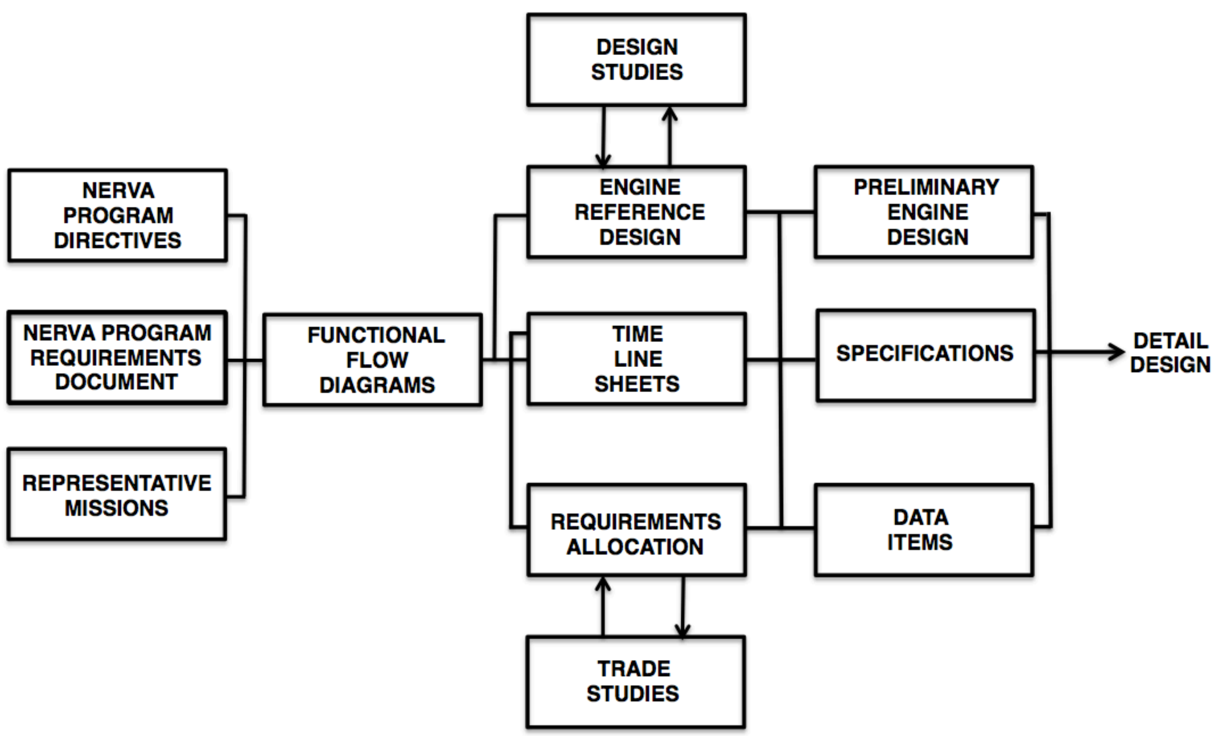

Figure 8. NERVA System Engineering Flow Diagram Definition Phase.
(Redrawn from Reference 49)

Deciding how long to pursue a technology is challenging; stop too soon and one may miss the next major advance; continue too long and waste resources if the technology doesn't deliver. (Despite the problems uncovered in the SIG program, DOE-sponsored work on selenide technology continued for over two more years.[50]) In an effort to bound the problem let me cite an incident that occurred around the end of 1970, when the Cleveland Extension of the Space Nuclear Propulsion Office received word that there would be a major reduction in the NERVA (Nuclear Engine for Rocket Vehicle Applications) program. One of my coworkers told me that he had spent 10 years working on the nuclear airplane which had cost over a billion dollars and that program had been cut. Then he had spent 10 years on the nuclear rocket program and that program, which had cost over a billion dollars, was being cut. He offered a "rule": When any technology program goes 10 or more years and/or costs a billion dollars or more and doesn't deliver something it is a candidate for termination.

In the end, it comes down to Richard Feynman's observation that "For a successful technology, reality must take precedence over public relations, for nature cannot be fooled".[51] To which we can add Peter Viereck's "definition" cited in Section 4.1: "Reality is that which, when you don't believe in it, doesn't go away".[39]

Acknowledgments

The author acknowledges the many very helpful pieces of advice on thermoelectrics (and other topics) he has received over the years from Mr. Gil Silverman and Dr. Cronin Vining. However, neither of those two gentlemen is in any way responsible for the opinions and interpretations presented in this paper; any defects are the responsibility of the author alone. The author would also like to acknowledge the advice he received from the late Gerry Stapfer whose career spanned thermoelectrics to gravity waves. ("May he go among the imperishable stars,/May he journey in the Boat of a Million Years."[52])

Appendix A

List of Acronyms

AEC	=	Atomic Energy Commission (sometimes written USAEC) (1947 - 1974)
ANSC	=	Aerojet Nuclear Systems Company
BOL	=	Beginning Of Life
Cm-244	=	Curium 244
DOE	=	Department of Energy (sometimes written U.S. DOE) (1977 – present)
EOM	=	End Of Mission
ERDA	=	Energy Research and Development Administration (1974 – 1977)
ETG	=	Electrically heated Thermoelectric Generator (non-nuclear)
GE	=	General Electric
GFE	=	Government-Furnished Equipment
GM	=	Galileo Mission
GPHS-RTG	=	General-Purpose Heat Source Radioisotope Thermoelectric Generator
GRC	=	John H. Glenn Research Center at Lewis Field (NASA)
KSC	=	John F. Kennedy Space Center
MHW	=	Multi-Hundred Watt
MSFC	=	George C. Marshall Space Flight Center (NASA)
NERVA	=	Nuclear Engine for Rocket Vehicle Applications
NPRD	=	NERVA Program Requirements Document
NRDS	=	Nuclear Rocket Development Station
POCO	=	Pure Oil Company (manufacturer of graphites)
Pu-238	=	Plutonium 238
RCA	=	Radio Corporation of America
RIFT	=	Reactor In Flight Test
RPS	=	Radioisotope Power Source (or System)

RTG	=	Radioisotope Thermoelectric Generator
SIG	=	Selenide Isotope Generator
Si-Ge	=	Silicon-Germanium alloy used in the MHW-RTG and GPHS-RTG thermoelectric elements
S/N	=	Serial Number
SNAP	=	Systems for Nuclear Auxiliary Power
SNPO	=	Space Nuclear Propulsion Office
TAGS	=	Tellurium Antimony Germanium Silver (Ag) thermoelectric material
TES	=	Teledyne Energy Systems
3M	=	Minnesota Mining and Manufacturing (Company)
WANL	=	Westinghouse Astronuclear Laboratory
We	=	Watts of electrical power
Wt	=	Watts of thermal power

Appendix B

SIG/GM Program Participating Organizations[2]

Item	Organization
Government RTG Supplier	Department of Energy
User/Agency/Spacecraft Integrator	NASA/Jet Propulsion Laboratory
RTG System Contractor	Teledyne Energy Systems
Thermoelectric Converter Contractor	3M Company
Heat Source Assembly	Monsanto Research Corporation (Mound)
Fuel and Cladding	Savannah River Plant
Heat Pipes	B & K Engineering, Inc.
Insulations and Technology	Oak Ridge National Laboratory
Fibrous Insulation Testing	Dynatech R/D Company
Multifoil Insulation	Thermo Electron Corporation
Electroplating and Chemistry	Battelle Columbus Laboratories
Graphite Characterization	Southern Research Institute Battelle Columbus Laboratories
Safety Testing	
• Blast overpressure	Los Alamos Scientific Laboratory
• Flyer plate	Air Force Weapons Laboratory
• Plasma arc	NASA/Ames Research Center Air Force Dynamics Laboratory/ Wright-Patterson Air Force Base

Dynamic Testing	Naval Surface Weapons Center
Technology Development, Test and Assessment	General Atomic Company Jet Propulsion Laboratory Fairchild Space & Electronics Company Battelle Columbus Laboratories
Electrical Heat Source	General Electric Company
Quality Assurance	Sandia Corporation

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