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FLAME HEATED THERMIONIC CONVERTER RESEARCH

Final Report
(1 July 1961 – 30 June 1963)
Contract No. DA-36-039 SC-88982

Department of the Army Task No. 1G6-22001-A-053-03
U. S. Army Electronics Research and Development Laboratory
Fort Monmouth, New Jersey

ATOMICS INTERNATIONAL
A DIVISION OF NORTH AMERICAN AVIATION, INC.

NO OTS

AI-8681

FLAME HEATED THERMIONIC CONVERTER RESEARCH

Final Report
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Contract No. DA-36-039 SC-88982

By
W. R. MARTINI

Power Sources Division Technical Guidelines for PR & C
No. 61-ELP/D-4623 dated 23 December 1960

Department of the Army Task No. 1G6-22001-A-053-03

Object: To develop the technology required for portable flame heated thermionic power sources.

ATOMICS INTERNATIONAL
A DIVISION OF NORTH AMERICAN AVIATION, INC.
P.O. BOX 309 CANOGA PARK, CALIFORNIA

CONTRACT: DA-36-039 SC-88982
JANUARY 1964

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PURPOSE

The purpose of the procurement is to investigate the various problems encountered in the design and construction of thermionic generators capable of producing from 5 to 200 watts of power. These problems cover:

1) The selection of suitable materials for thermionic diode envelopes and heat ducts where high temperatures and corrosive gases are encountered.
2) The design of a fossil-fuel burner capable of providing the required temperatures and heating rates.
3) The establishment of design parameters for thermionic generators of various power levels from 5 to 200 watts.

PROGRAM OUTLINE

TASK A – THERMIONIC CONVERTER DEVELOPMENT

1. Development of flame heated converter construction technique.
2. Building converters for test.

TASK B – HEAT SOURCE DEVELOPMENT

1. Premix burner development.
2. Fuel injection burner development.
3. Theoretical studies on heat economy and combustion.

TASK C – MATERIALS DEVELOPMENT AND EVALUATION

1. Procurement and evaluation of emitter thimble materials and coatings.
2. Gas permeation measurements.

TASK D – PROTOTYPE DEVELOPMENT

1. Testing flame heated converters.
2. Series connection studies.
3. Push-pull connection studies.
4. System design and construction.

TASK E – PROJECT COORDINATION AND REPORTS

Coordinate the various parts of this contract and write seven quarterly technical reports and one final technical report.

OTHER PROGRAMS

The following programs at Atomics International are related to the present contract:

1) Company sponsored research on flame heated thermionic converters.
2) Office of Naval Research sponsored research on the basic physics of thermionic converters
3) Solar-heated thermionic converters for the Jet Propulsion Laboratory.
4) Research on uniform work function diodes for Aeronautical Systems Division, US Air Force.

5) Company sponsored research on new emitter and collector materials.

6) Company sponsored research on low temperature diode construction methods and on the evaluation of materials for flame corrosion and gas permeation.

7) ASD sponsored program on development and testing of a two-thermionic converter module simulating a nuclear reactor fuel element (joint with Thermo Electron Engineering Corp. and Battelle Memorial Institute).

ABSTRACT

The internal flame heated thermionic converter concept has been shown to be feasible. A hot shell to be used on this type of converter has been tested at a temperature of 1300°C for 607 hr and then at 1400°C for a total of 640 hr. The shell is still leak-tight. The permeation rate of gas through this type of hot shell is insignificant, being only 0.003 cm^3 (stp)/hr. The successful hot shell is molybdenum coated first with molybdenum disilicide and then with pyrolytically deposited silicon carbide.

A burner has also been perfected for the internal flame heated thermionic converter. It employs an internal and an external heat exchanger to salvage flue gas heat. Propane is injected into the combustion chamber where mixing and burning are promoted by a converging flow vortex. Heating efficiencies of up to 56% and burner lifetimes greater than 600 hr at 1300°C emitter temperature have been obtained.

A conceptual design showing the advantage of using the internal flame heated thermionic converter is presented.

PUBLICATIONS, LECTURES, REPORTS, CONFERENCES
(For 8th Quarter)

W. R. Martini attended the 17th Annual Power Sources Conference at Atlantic City, New Jersey, May 21-23, 1963. He presented a paper entitled "Internally Heated Fuel-Fired Thermionic Converter Systems."

On May 20, W. R. Martini visited the Minnesota Mining and Manufacturing Company, St. Paul, Minnesota, for the purpose of discussing corrugated ceramic materials.

On June 17, 1963, Mr. Joseph P. Angello from USAELRDL visited Atomics International for the purpose of discussing progress on the contract and final arrangements for conclusion of same.

TECHNICAL PROGRESS

INTRODUCTION

This report summarizes the significant developments accomplished during the 2-yr life of the contract. It is not intended in this report to summarize everything previously presented in the prior seven quarterly type reports.[1-7] Only those lines of development which have proved to be significant will be summarized, and those lines of development which are no longer of any major interest will simply be mentioned with reference to the proper quarterly report. The program outline used during the second year of the contract will serve as a basis for reporting progress.

TASK A – THERMIONIC CONVERTER DEVELOPMENT

During the early phases of the project, a 150-watt thermionic converter was built and tested. As shown in Figure 1, this converter employed a combination of resistance heating from a tantalum foil filament and low voltage electron bombardment to heat the molybdenum emitter. The emitter had an area of 84 cm^2. Four sapphire spacers were set into the collector assembly to keep the emitter and collector from shorting. The sapphires functioned properly even though they were found to have shattered. The performance of this converter is described under Task D. Failure of the converter was due to a small leak in the molybdenum-nickel weld at the very top of the converter. This allowed cesium to escape slowly into the vacuum envelope until the cesium in the diode was completely depleted.

A total of,10 flame heated thermionic converters were constructed for the purpose of furnishing a flame-heated thermionic converter for the first year's sample product as required by contract. Converter 6 was delivered as the sample product. Converters 7, 8, 9, and 10 were built in an attempt to make a better sample product. A section of one of these converters in its test stand is shown in Figure 2. Notice that the combustion chamber is inside the converter and the emitter is the bottom end of the hot shell thimble. The emitter and collector are held apart by a sapphire spacer, while air pressure acting upon the expansion diaphram keeps the sapphire spacer in compression at all times. The collector temperature and the cesium reservoir temperature are controlled by movable radiation shields. Table I summarizes the construction features of the 10 converters.

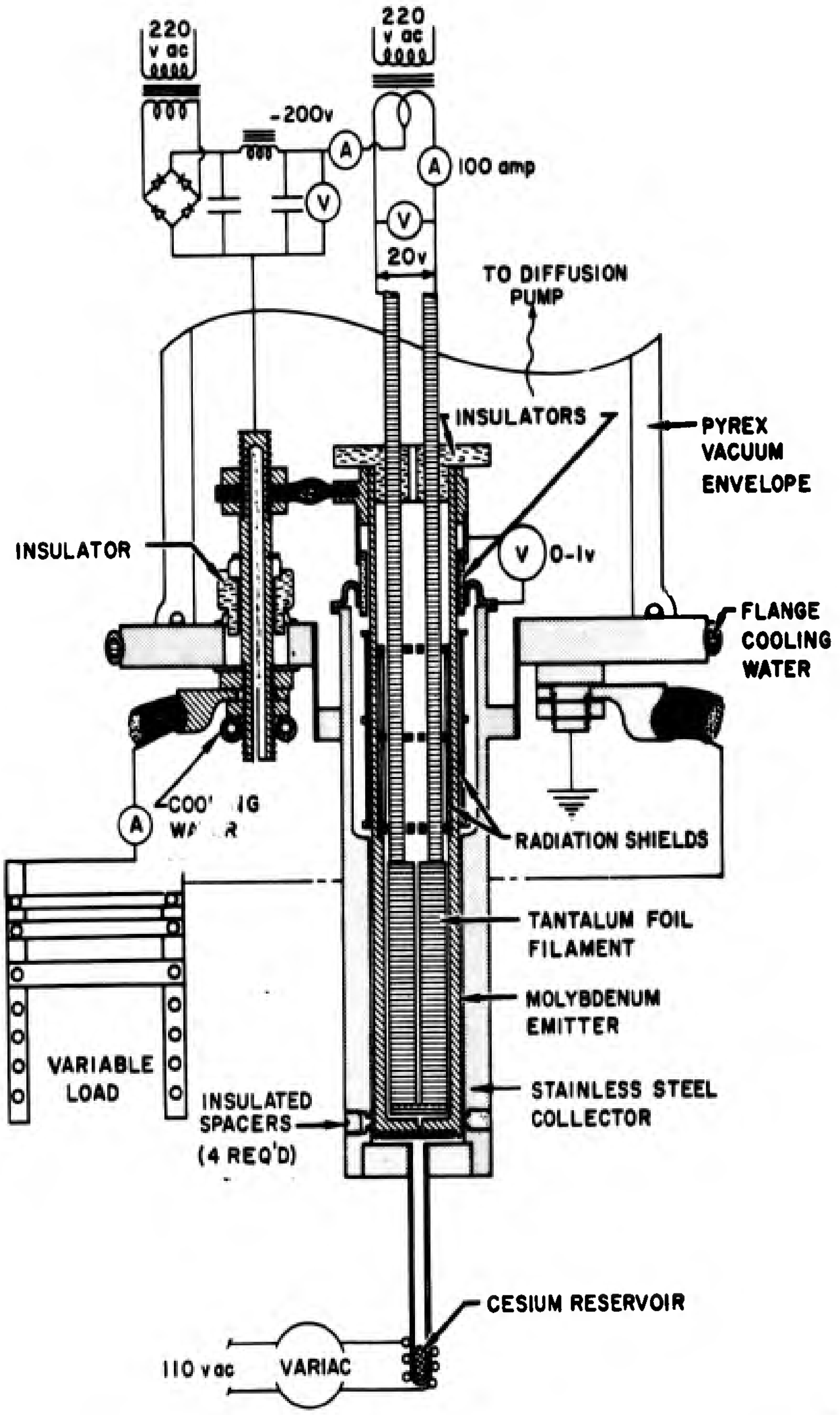

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Figure 1. Cutaway of the 150 w (e) Thermionic Converter

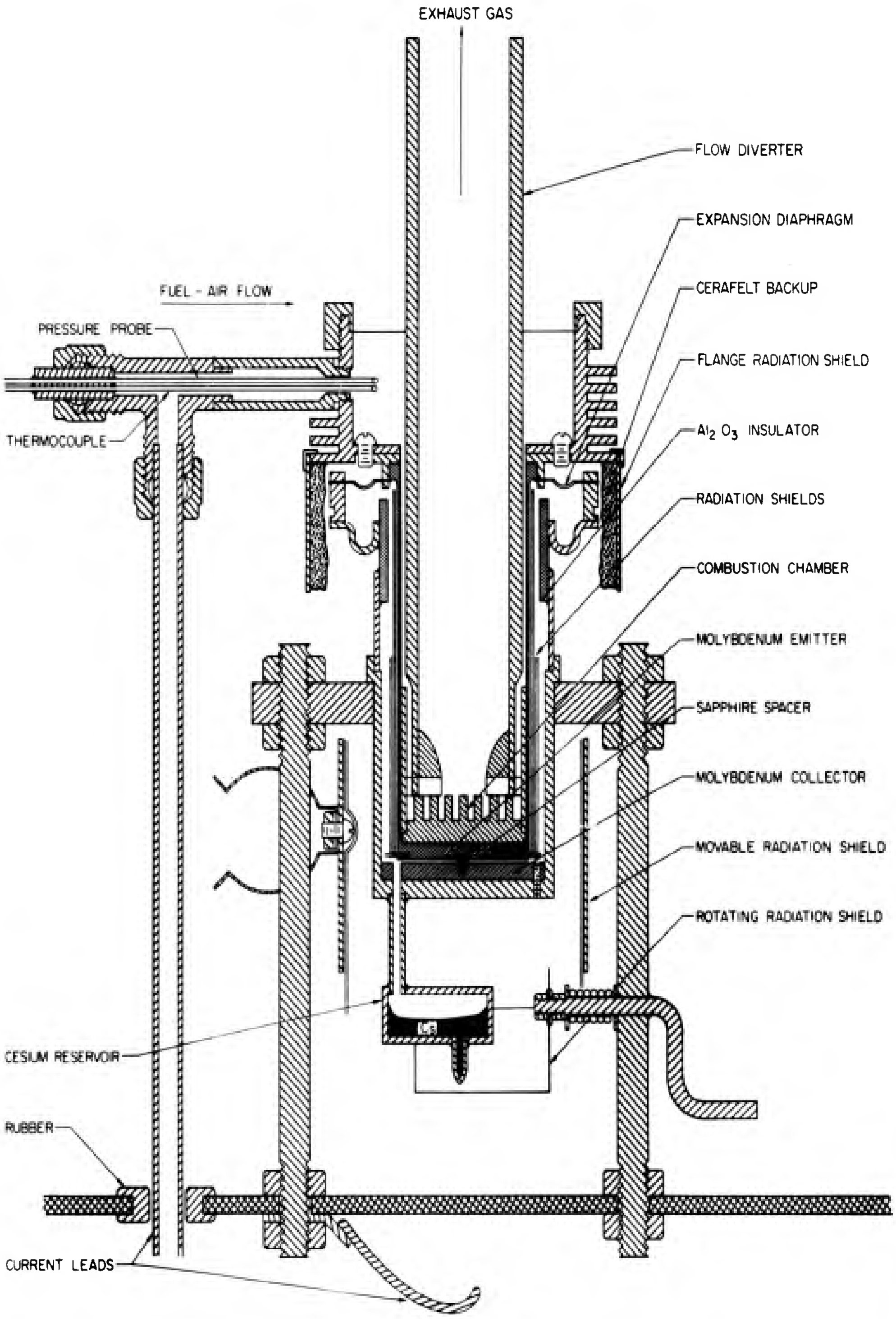

Figure 2. Second Sample Product Diode Test Apparatus

TABLE I

CONSTRUCTION FEATURES OF FLAME HEATED THERMIONIC CONVERTERS*

Diode No.	Sapphire Spacer	Ti-Zr Getter	Spacing
1	yes	no	0.010
2	yes	no	0.010
3	yes	yes	0.010
4	yes	yes	0.010
5	no	yes	variable
6	no	yes	variable
7	no	yes	variable
8	no	yes	variable
9	no	yes	variable
10	no	yes	variable

*For hot shell description see Table IX

Improvements were made in molybdenum hot shell fabrication. Machined molybdenum hot shells were found to be unsatisfactory since most of the shells developed cracks after the coating was applied. Consequently, an end cap weld of a deep drawn molybdenum tube to an arc-melted molybdenum cap was developed after some trial and error. The welds were a source of leaks in certain cases. Therefore, the deep-drawn molybdenum thimbles used in the last 2 converters was considered to be somewhat more satisfactory. The types of coating employed and the performance experienced will be discussed in Task C.

The sapphire spacer shown in Figure 2 proved to be quite fragile. After the first 4 converters, the spacer was left out and the spacing was controlled by a jack screw assembly working on the expansion diaphragm.

Also, after the first 2 diodes, titanium-zirconium alloy shavings were placed in the cesium reservoir to getter hydrogen and some nitrogen as it permeated through the hot shell.

The joining of the molybdenum to the diaphragm assembly, a kovar-molybdenum weld, proved to be the most difficult operation in the construction of the converter. At first, a copper brazed joint was tried, but difficulty was experienced with this type of a joint because the copper braze could not be successfully heated by induction. For the welded and deep drawn hot shells, the technique of melting the kovar onto the molybdenum, using a tungsten inert gas (TIG) arc in a retort, usually produced leak-tight welds on the first try. For some of the very thick pyrolytic silicon carbide coated hot shells which were tested near the end of the project, welding fractured the coating too badly and the return to a copper braze was required. The copper braze was applied in a dry hydrogen atmosphere after the molybdenum had been preplated with nickel. The braze was generally satisfactory.

It should be emphasized that the task of building flame heated thermionic converters was part of the first year's work. In the second year, the emphasis was on hot shell development.

TASK B – HEAT SOURCE DEVELOPMENT

Phase 1 – Premix Burner Developments

A series of 27 runs involving 7 separate burner experiments was conducted using the experimental apparatus shown in Figure 3. The foam silicon carbide baffle shown in Figure 3 gave the best results. That is, the combustion chamber temperature was the highest at a reasonable pressure drop and efficiency. The brightness temperature of the bottom of the combustion chamber, as measured by a pyrometer looking down through the exhaust gases, is taken as the combustion chamber temperature. The percent of the lower heating value of the propane and air mixture which was absorbed by the heat sink is defined as the heating efficiency. The best results were a temperature of 1450°C at a heating efficiency of 12%, a heat input of 1515 watts, and a supply pressure of 3.3 in. of water. At this operating point, no tendency to flash back or whistle was observed.

After this experiment, diodes designed for flame-heating became available and the burner was adapted to these diodes. Generally, it was found that the same input of heat was required to obtain the same combustion chamber temperature when the diode was on open circuit. However, three problems arose when the burner perfected in the standin apparatus shown in Figure 3 was applied to the actual diode. They were:

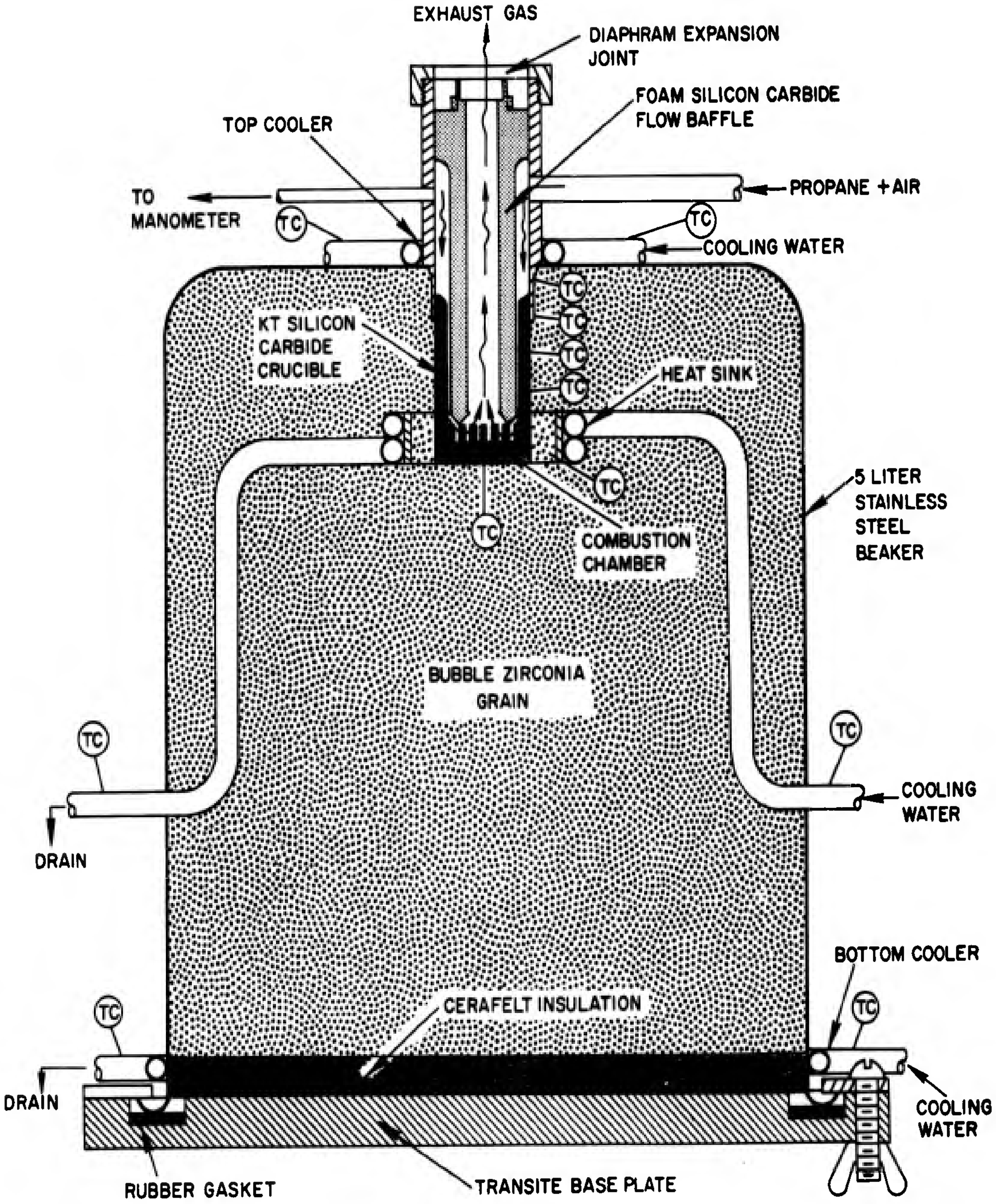

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Figure 3. Demonstration Diode Heater Test Assembly

1) Flash-back of the premixed fuel-air mixture became a problem.

2) The burner tended to burn with an ear-piercing screech unless a muffler was attached to the outlet.

3) The converters behaved as if the actual emitter temperature was very much lower by several hundred degrees than the apparent temperature of the bottom of the combustion chamber.

The problem of flash-back was solved by passing the fuel air mixture through very narrow annuli down to 0.020 in. and by using thin-wall silicon carbide flow dividers with a wall thickness down to 0.040 in. Of course, these provisions increased the required air supply pressure to a large degree.

In Diode No. 9 (see Table II), a flow divider was used with a 0.020 in.-clearance and a 0.040-in.-thick wall. An input of 1100 watts was required to obtain a temperature of 1500°C at the bottom of the combustion chamber which was made of 1/4-in.-high posts, 1/16 by 1/16 in. on 1/8-in.-square centers. The combustion chamber was cemented to the bottom of the molybdenum thimble with silicon carbide cement composed of silicon carbide grain and an oxide binder. No flash-backs developed, but excess air was needed to keep the flame from walking too far back into the heat exchanger.

TABLE II

RESULTS OF DIODE BURNER TESTS

Diode No.	Combustion Chamber Temperature-°C (brightness)	Heat Input (watts)	Main Air Supply Pressure (in. H_2O)	Duration	
				Time (hr)	Cycles
1	1300	1310			
2	1415	1090	14.6	3	3
3	1415	1000	3.1	2	3
4	1550	700		6	2
5	1410	1300	3.6	1	1
6	1566	1950		4	2
7	1600			~ 6	1
8	1650	1950	>8	6	2
9	1500	1100	17.5	2	1
10	1550	1470	15.6	1	1

The heater for Diode No. 10 was an improvement over that used in Diode No. 9 because the combustion chamber and the attendant difficulties of transferring heat from the silicon carbide combustion chamber to the molybdenum was removed by removing the combustion chamber. With the heat input of 1470 watts, a temperature of 1550°C (brightness) was observed on the molybdenum disilicide hot shell with a pressure drop of 15.6 in. of water. Evidently, the high pressure drop in premix burners of this type is due to the ignition of the gas-air mixture in the close clearance between the diode thimble and the flow divider. Ignition of the mixture abruptly increases the temperature, which decreases the gas density and increases gas velocity. The energy required to accelerate this gas must be supplied by an increased burner back pressure. This high burner back pressure is evidently inherent in this type of premixed fuel and air burner to heat a thimble for a thermionic converter. Also inherent in premix burners is the low overall efficiency, unless extremely high burner back pressures are employed (see Phase 3). For these reasons it was decided to return to the development of a fuel injection burner for the internally heated thermionic converter.

Phase 2 – Fuel Injection Burner Development

In the first part of the project five propane aspiration burner tests were described.[1] The intent of this development was to use a propane powered jet pump to move the combustion air through a regenerative heat exchanger and a combustion chamber. Although eventually a temperature of 1000°C was obtained at a heating efficiency of 20% and a projected heat flux of 8 watts/cm^2 using a propane fuel pressure of 100 psig, the concept was unsatisfactory because even at these conditions, which are short of the required conditions, the stable mode of combustion was very noisy. The noise is produced by unstable combustion in the mixing tube of the jet pump. This combustion drastically decreases the efficiency of the jet pump and is quite undesirable. Some though was given to the possibility of developing a burner with very little regenerative heat exchange, which would be powered with a jet pump. However, as will be explained in Task 3, this type of burner can never have a very high heating efficiency and therefore was not developed.

Three tests were made on a type of reversing flow burner with two regenerators.[3] A combustion chamber temperature of 820°C was obtained at a heat flux of 23 watts/cm^2. A heating efficiency of 39% was obtained with an air supply pressure of from 9 to 13 in. of water. The fuel was injected directly into the combustion chamber and the air flow through the regenerators was reversed every 30 sec. A swing in the combustion chamber temperature of 30°C was observed due to incomplete combustion with one direction of air flow. It was decided that added complexity of the reversing flow and the possibility of obtaining an oscillating emitter temperature overruled the advantage of high heating efficiency. Therefore, the steady-flow regenerative heat exchanger was preferred.

A type of fuel injection burner was employed in the very first demonstration flame heated thermionic converter test ever conducted using a high temperature converter.[1] The very large furnace used in this demonstration was subsequently instrumented to determine its overall heating efficiency. The furnace was 2 ft tall, 10 in. in diameter, and weighed 30 to 40 lb. A simulated emitter temperature of 1100°C* was obtained with 2100 watts input and 8% heating efficiency (60 watts/cm^2). An air supply pressure of 15 in. of water was required. However, the heat exchanger in this burner test was very fragile in that it would crack due to thermal shock after a few hours of operation. It was felt that a very much smaller and more rugged burner could be developed to heat the converter.

The development of the fuel injection burner for the interally heated converter was done concurrent with testing of hot shells for the internally heated converter. A summary of these burner tests is given in Table III. Many different types of fuel injection burners were tested. However, the fuel injection burner stabilized by a converging flow vortex illustrated in Figure 4 gradually evolved and has become standard for the testing of hot shells. It has an even heating capability, a large amount of reserve capacity, and will operate for long periods of time without deterioration. For instance, in VB 35 one burner has operated for over 600 hr without repairs. Heating efficiences of these burners were not measured.

Phase 3 – Theoretical Studies on Heat Economy and Combustion

When a fuel is burned with air, both of which are initially at ambient temperature, the flame has a well-defined maximum temperature. Table IV gives the experimentally determined temperatures for the stoichiometric combustion of methane, propane, and gasoline, and shows that the adiabatic flame temperatures are all about 1900°C, for these common fuels.

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*Temperatures up to 1600°C were obtained on other tests

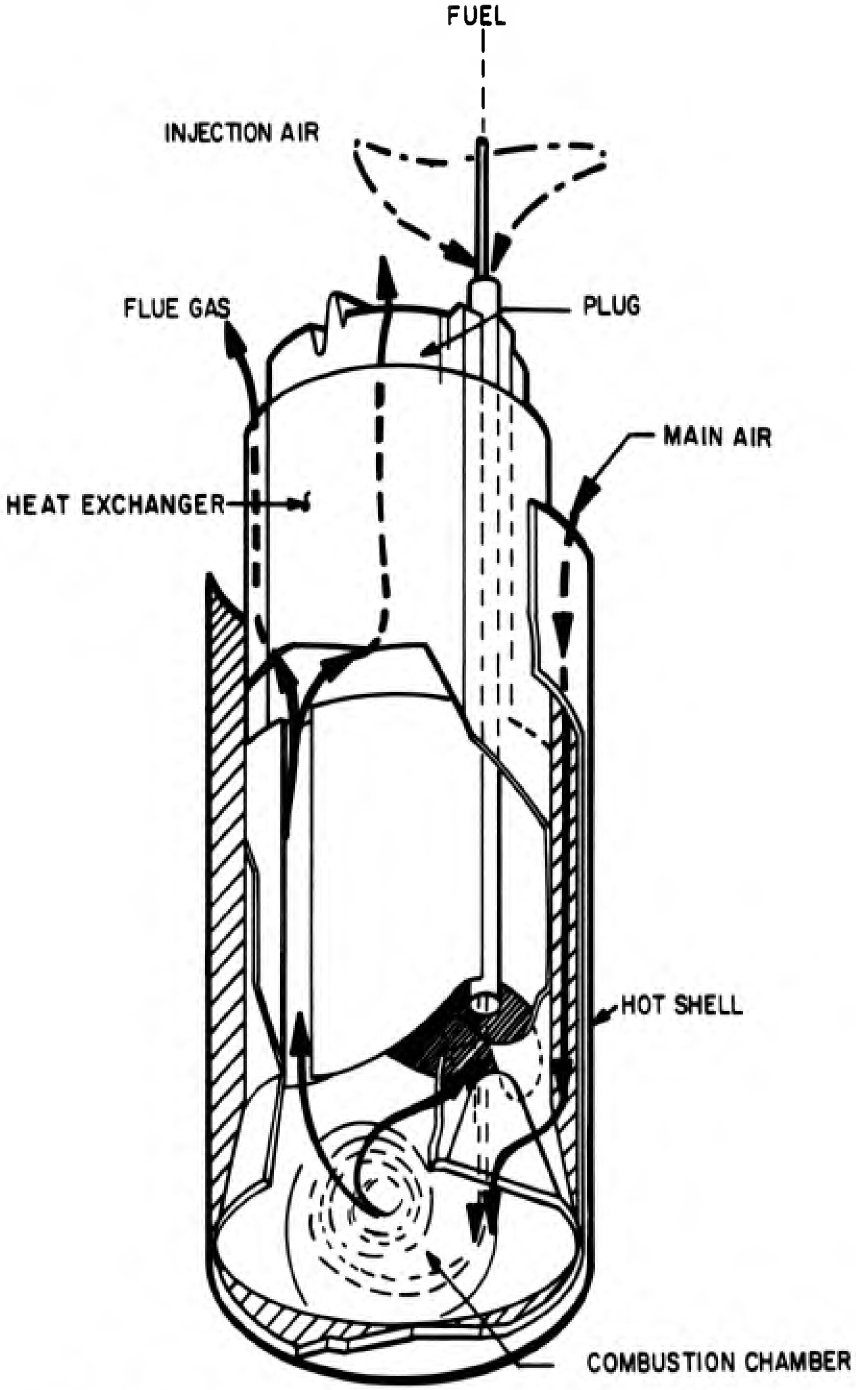

Figure 4. Fuel Injection Converging Vortex Burner

TABLE III

VACUUM INSULATED BURNER TESTS

Run No.	Emitter Temperature °C (brightness)	Heat Input (watts)	Heat Flux* (watts/cm^2)	Heating Efficiency (%)	Main Air Supply Pressure (in. H$_2$O)	Injection Air Supply Pressure (in. H$_2$O)	Propane Supply Pressure (psig)	Duration		Did Burner Fail?	Burner Type
								Time (hr)	Cycles		
VB 1	955	485	4.6	6	0.8	6	8	3	2	No	Premix
2	1180	1350	9.8	4	4.5			6	1	No	
3	1150	840	4.9	4	1.6			8	1	No	
4	1170	1330	9.8	4	4.5			7	4	Yes	
6	940	2040	64	19	0.6			4	1	Yes	
7	1180	1490	57	23	0.4			2	1	Yes	
8	1160	1220	29	14	1.2			2	1	Yes	
9	1130	980	17	11	2.7			4	3	No	Fuel Injection
10	1300	900	18	12	2	6		42	2	No	
11	1155	1590	36	14	1	6.4		136	1	No	
12	1130	1730	32	11	0.9	0.3		22	1	No	
13	1365	1030	23	13	1.9	10.6		61	1	No	
14	1300	1720	46	16	4.0	22.6		18	1	No	
15	1400	1020	93	56	1.8	7.0		36	1	No	
16	1260	1740	35	12	30	24.2		56	1	No	
17	1355	1040	20†	12†				16	1	No	
18	1285	1940	16†	5†	2.0			6	1	No	
19	1310	1820	18†	6†	2.0	2.8		70	1	No	
20	1200	1820	13†	4†		2.4		1	1	Yes	
21	1325	1820	18†	6†	0.1	3.8		36	1	No	
22	~1300							1	1	No	
23	1380	1890	21†	7†	2.0	14.2	8	45	1	No	Fuel injection with converging vortex combustion chamber
24	1480	470		1.2	9.8			4	2	Yes	
25	1310	300			0.9	§	11§	1	1	Yes	
26	1310	370			1.8	5.6	5	10	1	No	
27	1320	500			2.8	§	20§	23	1	Yes	
28	1335	340			1.6	15.3	13	110	1	No	
29	1300	320			1.0	7.2	0	95	1	No	
30	1300	470			0	7.9	1.5	24	1	No	
31	1300	290			0.5	8.8	0	176	8	No	
32	1300	300			0.6	12.2	2.5	4	1	No	
33	1300	320			0.8	12.2	0	109	1	Yes	
34	1300							0.5	1	No	
35	1300-(1400)	410			0.8	18.7	0	640	3	No	
36	1300	300			0.3	10.5	0	276	3	No	
37	1300	300			1.8	10.6	0	3	1	No	
38	1300-(1400)							133	3	No	

*Heat Transfer Area = 6.1 cm^2
†Assuming free radiation with ϵ = 0.5
§Fuel-air mixture injected together

TABLE IV

SELECTED FUEL PROPERTIES

Fuel	Experimental Flame Temperature With Air[8] (°C)	Ignition Temperature With Air[8] (°C)	Fuel Deposition Temperature (°C)	Moles Air Required (10% Excess) Per Mole Fuel
Methane	1870	632	390*	10.5
Propane	1925	480	390*	26
Gasoline				
Regular	1950	280	171(9)	65
100 Octane	1950	429	-	65

*Based upon no cracking below this temperature

In the limiting case of perfect heat transfer from the flame to the emitter, the maximum heat available is that obtained in cooling the gases from the flame temperature to the emitter temperature. In practice, however, only a rather low heat transfer coefficient can be realized for transferring heat from the flame to the emitter. Consequently, a large temperature difference must be maintained in order to develop the required heat flux. This effect makes the heating efficiency, η_H, materially less than the theoretical maximum shown in Figure 5. The heating efficiency, η_H, is defined as the ratio of the heat transferred to the emitter, Q_e, to the potential chemical energy of the fuel, Q_F; and is given for this ideal, limiting case along the ordinate of Figure 5. Note that, for an emitter temperature of 1500°C, the maximum η_H varies over the narrow range of 0.25 to 0.28, depending upon the fuel used.

As shown in Figure 5, η_H can be increased by introducing a heat exchanger between the exhaust gases and the incoming combustion air and fuel. This has the effect of increasing the flame temperature and the heat available to the emitter. Theoretically, η_H can approach unity as the heat exchanger efficiency, η_X, approaches unity (the heat exchanger efficiency is defined as the ratio of the heat absorbed by the incoming air and fuel, Q_P, to the heat available in the gases leaving the combustion zone, Q_R). A heat exchanger is thus seen to be an essential component of an efficient flame-heated thermionic converter system.

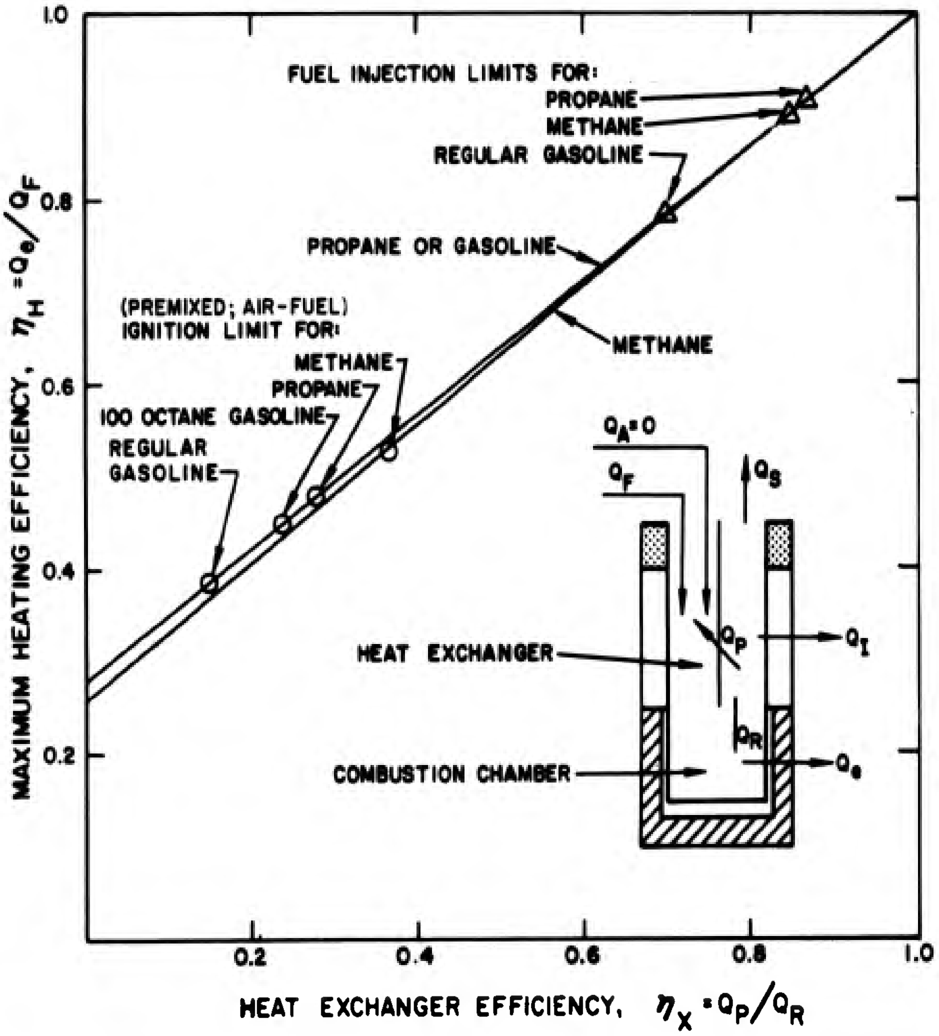

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Figure 5. Maximum Heating Efficiency for Various Fuels (T_e = 1500°C, 10% excess air, perfect flame-emitter heat transfer)

Heat Exchanger Operation, Premixed Air and Fuel

It is of interest to examine ways of operating the heat exchanger. The first of these involves the premixing of the fuel and air and the subsequent heating of this mixture by the Q_P heat. Under these conditions, the mixture can be heated only to the ignition point (Table IV), in order to avoid premature combustion in the heat exchanger. If premature combustion is allowed to occur, the direction of the heat flow in part of the heat exchanger reverses, and an increasing fraction of the combustion heat is lost to the exhaust gases. Once started, combustion in the heat exchanger tends to be self-perpetuating, and can lead to dangerously high temperatures in the exchanger. Of the common gaseous fuels, methane has the highest ignition point, 632°C. When this air-fuel mixture is preheated to 632°C, and the emitter temperature is 1500°C, methane can yield a maximum heating efficiency of 53%. The maximum heating efficiency of premixed regular gasoline and air is 38%.

The ignition points given in Table IV are the temperatures at which a mixture of fuel and air burns, if held at that temperature for some time. If preheating is done rapidly, preheat temperatures that are higher than the ignition temperature might be successfully employed. In the fifth quarterly report, we analyzed the effect of ignition delay on the premix burner used in Diode 10. We found that the mixture was preheated mostly by transfer of heat from the hot shell and very little from the hot exhaust gases. That is, heat that would be lost by conduction along the hot shell was salvaged to preheat the incoming gas mixture. Near the combustion chamber a very rapid heatup rate was computed. This rapid heatup rate would favor preheat without combustion beyond the nominal ignition temperature. Literature values on ignition delay were employed to compute the onset of ignition which was found to be at a gas temperature of >1200°C. The nominal ignition temperature for propane and air is 480°C. (See Table IV.)

The maximum heating efficiencies, mentioned previously, are based upon a fixed emitter temperature of 1500°C. However, if the emitter temperature can be lowered, the heating efficiency increases. Figure 6 shows the maximum heating efficiency for gasoline fuels as a function of heat exchanger efficiency and emitter temperature. The two curved lines represent a conservative estimate of the preignition temperature limitations for regular and 100-octane gasoline, and show the maximum heat exchanger efficiencies known to be possible for premixed fuel at each of the emitter temperatures shown (no allowance for ignition delay). Note that, as the emitter temperature T_e decreases, the η_H increases, and that the known safe value of η_X for premixed air and fuel also increases slightly.

Heat Exchanger Operation, Fuel Injection

It is clear, from the preceding discussion, that the overall efficiency for flame-heated power sources, using premixed fuel and air, is limited. To obtain the ultimate η_o, fuel and air must be preheated separately. The fuel can only

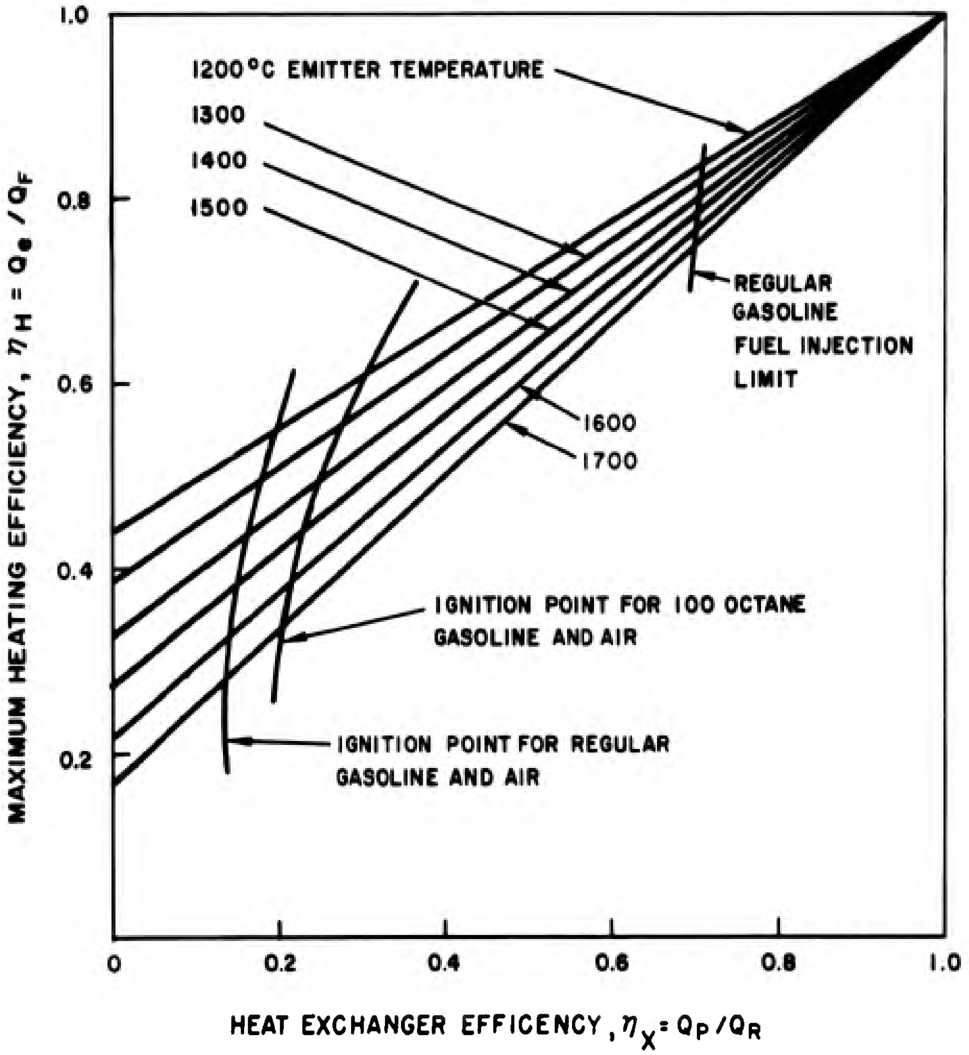

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Figure 6. Maximum Heating Efficiency for Gasoline

be preheated to the temperature at which it begins to form a deposit in the fuel line. This maximum temperature depends upon the fuel composition (with particular attention to trace amounts of gums and additives), the wall temperature of the fuel tube, the allowable time between replacements of the fuel tube, and the possibility of burning the deposits out by running air or hot exhaust gas through the fuel tube. The fuel temperature at which deposits begin to occur is known, with certainty, only for regular gasoline (see Table IV). In any case, the fuel cannot be heated as hot as one would like to heat the air. The two streams must therefore be separated during passage through the heat exchange;

and, in general, it will be necessary to lead the fuel into the combustion zone through a cooled fuel injector. However, the fuel then has only a short time to mix and burn, making complete combustion more difficult.

The ultimate heat exchanger efficiency is attained if the 10% excess combustion air is preheated to emitter temperature by the exhaust gases which are initially at emitter temperature, and if the fuel is preheated to the deposition temperature (see Table IV). Using this definition, the maximum η_X, for a T_e of 1500°C is given in Table V and is plotted in Figure 5.

TABLE V

MAXIMUM HEAT EXCHANGER EFFICIENCY FOR A FUEL INJECTION BURNER (T_e = 1500°C)

Fuel	Maximum η_X
Methane	0.85
Propane	0.87
Regular gasoline	0.70

It is interesting to note that the maximum η_X for fuel injection is only mildly affected by T_e when regular gasoline fuel is used (Figure 6). How close one can approach the maximum heat exchanger efficiency, shown in Table V, depends upon the number of transfer units that can be built into a heat exchanger without exceeding a reasonable air supply pressure and how well one can eliminate extraneous heat leak through the insulation. Possible types of insulation are compared in Table VI.

Notice that radiation shields in the cesium vapor space of the thermionic diode have very good insulating properties. Consequently, a design which places the combustion chamber and the heat exchanger inside the diode has a definite advantage, from the standpoint of heat economy, system size, and system weight. All the flame heated diodes we are now constructing take advantage of the benefits of vacuum insulation.

Besides heat leak through the insulation, the other reason for not obtaining maximum efficiency is the heat loss in the stack gases. This can be reduced by the use of a regenerative heat exchanger. Consideration of thermodynamic principles has revealed the limitations on heating efficiency, if the air is preheated

TABLE VI

COMPARISON OF THERMAL INSULATION MATERIALS*

Insulation	Heat Loss (watts/cm^2)
Zirconia brick Type I (Temperature limit, 3270 °C)	7.1
Stagnant air (Conduction and radiation)	5.48
Vacuum (Radiation only, emissivity = 0.15)	4.47
Cerafelt (6 lb/ft^3, temperature limit, 1100 °C	3.1
Stagnant air and 9 radiation shields (Emissivity = 0.15)	1.46
Min-K Type 2000 (Temperature limit, 1100 °C)	1.12
Vacuum and 9 radiation shields (Emissivity = 0.15)	0.45
2 Torr† Cs vapor and 9 radiation shields (Emissivity = 0.15)	0.69

*The comparison assumes the insulation to be between two surfaces, one at 1500 °C and the other at 300 °C, 1 cm apart

†1 Torr = pressure = 10^{-3} mm Hg

and the fuel is injected at the maximum allowable temperature. These limitations are based upon a hypothetical heat exchanger, in which heat is transferred by an infinitesimally small driving force, ΔT_X. Now we will consider how close to the limit it is worthwhile to operate. The results of analysis for the case of regular gasoline and no insulation loss is presented in Figure 7. This figure shows the number of transfer units that are needed in the heat exchanger to attain a given heat exchanger efficiency. A transfer unit is defined by the equation:

$$N_{Tu} = UA_x/wC_p$$

where

N_{Tu} = number of transfer units

U = heat transfer coefficient (watts/cm^2-°C)

A_x = heat transfer area (cm^2)

w = flow rate (g/sec)

C_p = heat capacity at constant pressure (J/g-°C)

Since N_{Tu} is dimensionless, any self-consistent set of units can be used. The N_{Tu} is not an extensive property of the heat exchanger, as is the heat transfer area or the size; but, for a given service, it is a convenient performance indicator, by which alternate heat exchanger designs can be compared.

A number of preliminary measurements have been made to determine the effective heat transfer coefficient between the combustion chamber and the emitter. For the purpose of a very rough estimate, the gas temperature, T_R, is 200 to 400°C higher than T_e. This temperature driving force can possibly be reduced to 100°C by introducing an extended surface.

From a study of Figure 7 it appears that an N_{Tu} of 2 is as large as is practical without reaching the point of diminishing returns. For a T_e of 1500°C and a T_R of 1600°C, an η_H of 0.66 is obtained. If the length of the heat exchanger is doubled, N_{Tu} becomes 4 and η_H becomes 0.73. For a T_R = 1600°C, as $N_{Tu} \rightarrow \infty$, $\eta_H \rightarrow 0.77$. All these heating efficiencies are predicated upon negligible heat losses through the insulation, and the preheating of gasoline vapor only to its deposition temperature of 171°C.

In the design of an efficient heat exchanger for a given service, one usually has a choice of making it large with a low pressure drop or smaller with a larger pressure drop. Therefore, the available air supply pressure must be considered. At least two types of air supply are possible:

1) Use output of power source to power an electric blower
2) Use the energy derived by expanding the pressurized fuel to run a blower.

The use of an electric blower is the easiest way because many small, well-engineered, lightweight blowers are being sold. The performance of some of these blowers is shown in Table VII. One cfm of combustion air will generate ~1500 watts of heat. At 5% overall efficiency, 75 watts of electrical energy are available. For the blowers shown in Table VII, 1 to 3% of the available electric energy is required for the blower provided the blower only supplies combustion air. All these lightweight blowers are very high-speed axial flow

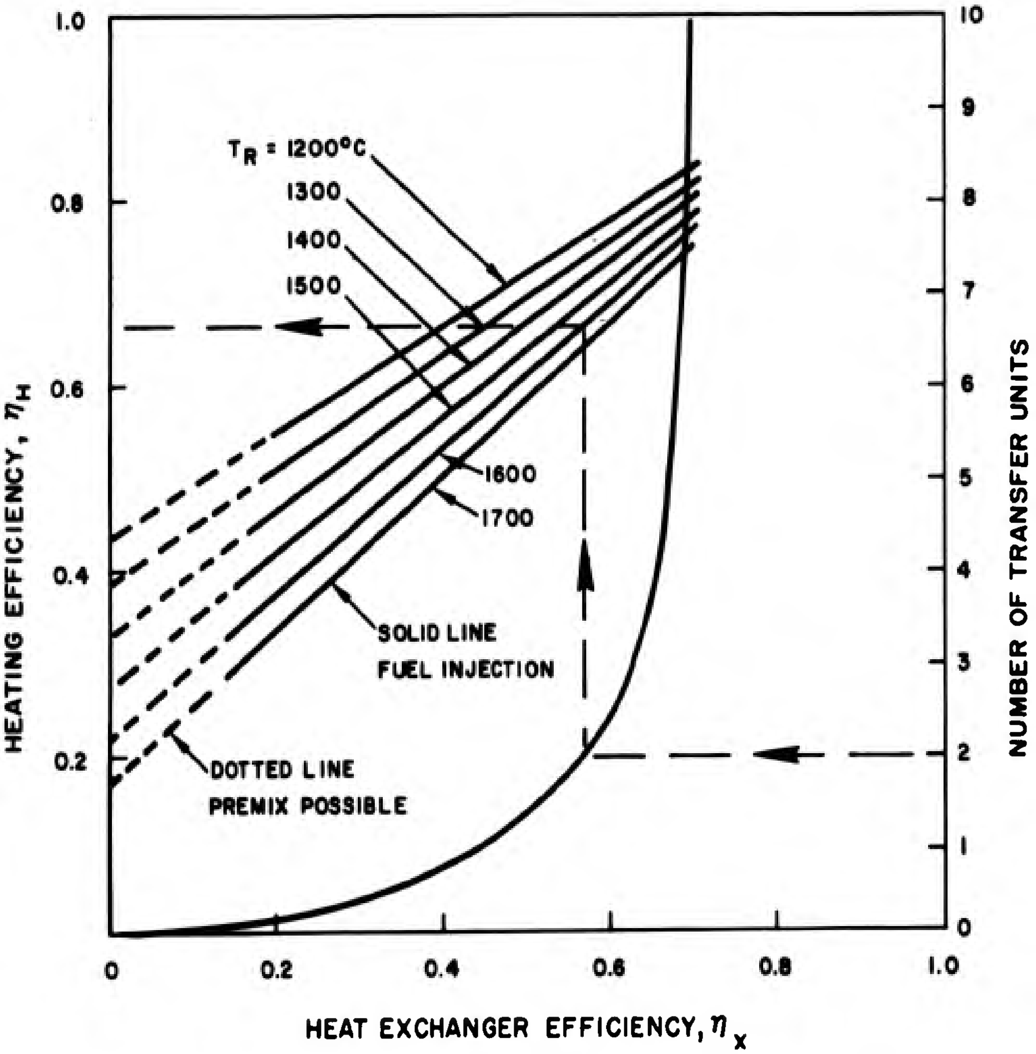

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Figure 7. Design Chart for Regular Gasoline-Fueled Diode Heaters ($Q_I = 0$)

TABLE VII

AVAILABLE LIGHTWEIGHT ELECTRIC BLOWERS

Source	Model	Optimum (ΔP in. H_2O)	Optimum Flow (cfm)	Electric Power Requirements			Weight (ounces)
				(volts)	(cycles)	(watts)	
Globe Indicator	VAX-1-DC	0.3	10	28	DC	5.6	1.4
Globe Indicator	VAX-2-MM	1.5	37	26	DC	30	5
Globe Indicator	STAX-3-FC	14.0	45	200	400	-	29
Rotron	Aximax	2.0	10	115	400	17	4

fans and therefore are inherently noisy. Under arctic conditions, a warm battery must be inserted into the power source to start operation. A quiet, low-speed positive displacement blower that could be hand cranked for startup would be the ideal type of blower but does not appear to be available in the size required.

The use of energy derived from expanding the pressurized fuel to pump the required air is an attractive possibility because it does not detract from the electric output but uses an otherwise wasted source of energy. Propane has a vapor pressure of 130 psig at room temperature. If a perfect adiabatic expansion of propane, from 130 psig to atmospheric pressure, could be coupled directly to a perfect adiabatic compression of the requisite combustion air, an air supply pressure of 28 in. of water would result. The vapor pressure of gasoline, just below its quoted coking temperature, is about 30 psia, or 15 psig. If the pressure energy of gasoline, under these conditions, could be perfectly utilized, a pressure of approximately 8 in. of water could be developed for its requisite combustion air. Since a pressure drop of 2 in. of water appears adequate to operate an efficient burner, these illustrations show that it might be possible to attain a workable air supply pressure with a mechanical expander-compressor engine, even if it were only 25% efficient.

The simplest type of expander-compressor engine is, of course, the jet pump. The pumping efficiency for a jet pump is, at best, 20% when the weight-rate of the fluid being pumped is equal to the weight-rate of the pumping fluid. For the high air-gas ratio required for burner jet pumps, the efficiency is only a few percent.[10, 11] Using the best information available, it appears that propane can aspirate 100% of the required combustion air and produce a pressure of about 1 in. of water gauge. The pressure difference generated by a gasoline vapor jet pump should be substantially lower.

For arctic operation, propane may have no vapor pressure. A hand-operated air pump can be used to pressurize the fuel tank. A mixture of compressed air and fuel can be fed to an air-motor-powered blower to furnish the combustion air. After warmup, the blower can be powered with fuel vapor alone.

TASK C – MATERIALS DEVELOPMENT AND EVALUATION

Phase 1 – Procurement and Evaluation of Emitter Thimble Materials and Coatings

The plan for the last 2 yr of work was that the coatings for refractory metals should first be applied to wires 1/16 in. diameter by 6 in. long and heated electrically in the apparatus shown in Figure 8. A number of these apparatuses were built in order to test a large number of samples of many different coatings to obtain statistically significant results with which to realistically evaluate a specific coating procedure. However, in a number of instances, the experience of the coating applied to the inside of the molybdenum thimble (used in the flame heated thermionic tests) did not agree with the experience of the same coating applied to the outside of the wire. However, the wire test was useful in screening a number of thin, self-healing coatings that do not depend particularly upon geometry for their successful application. A summary of these screening tests is shown in Table VIII. In general, the wire tests appear to last considerably longer than the hot shell tests, using the same material. For instance, the coating DURAK-B lasted as long as 629 hr in the wire test and as long as 95 hr in the hot shell test at comparable temperatures. The aluminum-tin coating on molybdenum lasted up to 196 hr in the wire test and 4 hr in the hot shell test. Conversely, TI-Kote lasted 5 hr in the wire test and 42 hr in the hot shell test.

The hot shell tests summarized in Table IX were all conducted using a molybdenum hot shell approximately 1 in. in diameter by 3 in. long. The inside of the thimble was coated with a coating found usable to prevent oxidation of the molybdenum, and also to prevent permeation of the gases through molybdenum. A burner was placed inside the coating to heat the end of the thimble to a high temperature. In all cases, this temperature was measured optically with proper corrections for windows, mirrors, and emissivity of the source. In some cases, the temperature of the combustion chamber was observed through one of the exit gas ducts. However, when at all possible, the emitter temperature was observed directly. In this case, the emitter assembly of a flame heated thermionic converter was installed in a glass vacuum chamber so that the side and the bottom of the thimble can be observed. A cutaway drawing of how one of these tests was conducted is shown in Figure 9. In this figure, one of the early fuel injection burners is shown heating the bottom of a molybdenum

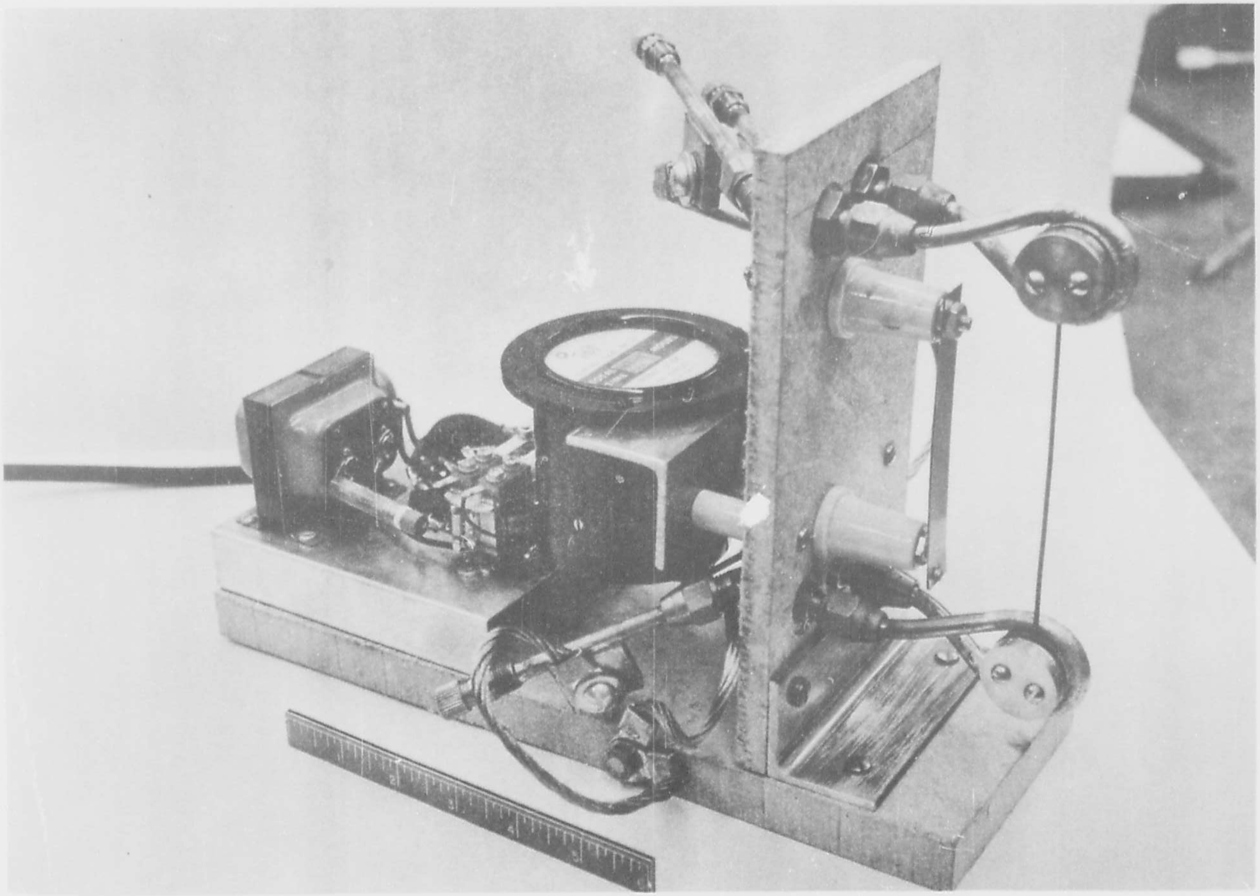

TABLE VIII

SUMMARY OF SCREENING TESTS USING COATED REFRACTORY METAL WIRES

Metal	Source*	Coating	Source*	Longest Time to Failure at 1350°C in Air (hr)
Mo	F	DURAK-B	Cr.	629
Ta	F	KS	Cr.	26
Mo	F	Al-Sn	GT	196
Mo	F	AI coat	AI	710(one only)
Mo	F	TI-Kote	TI	5
Mo	F	Pyro. SiC	R	15
Mo	F	T-61	TM	120†
Mo	F	Mod T-61	TM	887†
Ta	F	R-505C	GT	454†

* F = Fansteel
Cr = Chromizing Corporation
GT = General Telephone & Electronics Laboratory
AI = Atomics International
TM = Thermomet Co.
Ti = Texas Instruments
R = Raytheon
†Tested and procured on company sponsored program

thimble protected with a coating. The sides of the thimble are insulated with radiation shields and the end of the thimbles comes close to touching the heat sink. The heat sink has a slot in it so that one diameter of the converter can be observed through a window in the bottom of the glass vacuum envelope (not shown). A measure of the efficiency of the radiation shields can be obtained by noting the amount of heat picked up by the radiation shield cooler.

From Run VB 27 on, another type of apparatus was used which is shown schematically in Figure 10 and photographically in Figure 11. In this apparatus all rubber gaskets are eliminated and the system is designed to have a very low leak rate so that the gas permeation through the hot shell can be measured at any time by the pressure rise method. The apparatus is pumped down and degassed by the roughing pump. Then the vacuum is maintained during unattended operation with the ion pump. The diode heater inserted into the hot shell is of the type shown in Figure 4. Figure 11 shows the hot shell tester ready for testing.

TABLE IX

RESULTS OF HOT SHELL TESTS

Test Numbers	Hot Shell Description (Mo)			Maximum Temperature		Duration (hr)	Description of Failure
	Shape (1)	Mo Fabrication By (2)	Coating	(°C)	Where (3)		
Diode 1	F	M	DURAK-B + KT-SiC	1300	CC	3	Hole 2 in. from bottom
Diode 2	F	M	DURAK-B + KT-SiC	1415	CC	2	No failure
Diode 3	F	W	DURAK-B + KT-SiC	1415	CC	6	No failure
Diode 4	F	W	DURAK-B + KT-SiC	1550	CC	1	No failure
Diode 5	F	W	DURAK-B + KT-SiC	1410	CC	4	1/4 in. from bottom under SIC cup
Diode 6	F	W	DURAK-B + KT-SiC	1566	CC	~6	Sample product, not examined
Diode 7	F	W	DURAK-B + KT-SiC	1600	CC	2	Under SiC cup
Diode 8	F	W	DURAK-B + KT-SiC	1650	CC	6	Under SiC cup
Diode 9	F	DD	DURAK-B + KT-SiC (cemented)	1500	CC	2	Hole 1 in. from bottom
Diode 10	F	DD	DURAK-B	1535	CC	1	Hole 0.3 in. from bottom
VB-1-4	F	W	DURAK-B + KT-SiC	1290	E	41	No failure
VB-6-8	F	DD	DURAK-B	1180	E	8	Hole 0.1 in. from bottom
VB-9	F	DD	Sn-Al	1128	E	4	2 holes 0.68 and 0.76 in. from bottom
VB-10	F	DD	TI-Kote	1400	E	42	Holes in bottom
VB-11-14	F	DD	DURAK-B	1365	E	237(4)	Hole 1/2 in. from bottom
VB-15	F	DD	DURAK-B	1400	E	36	Hole 1/2 in. from bottom
VB-16-18	F	VD	TI-Kote†	1355	E	78	Longitudinal crack
VB-19	F	DD	DURAK-B	1310	E	70	Hole 1/2 in. from bottom
VB-20-21	F	DD	DURAK-B + KT-SiC	1325	E	37	Hole 1/2 in. from bottom
VB-23	F	DD	DURAK-B + KT-SiC (cemented)	1380	E	45	Hole 1/2 in. from bottom
VB-24-26	F	DD	DURAK-B + TI-Kote (cemented)	1480	E	15	Hole in bottom
VB-22, -27, -28	F	DD	DURAK-B + KT-SiC (cemented)	1335	E	137	Hole 1/2 in. from bottom
VB-29	R	DD	DURAK-B†	1360	E	95	2 holes 3/4 in. from bottom
VB-30	R	DD	DURAK-B†	1305	E	24	Hole 1/2 in. from bottom
VB-31	F	VD	TI-Kote†	1395	E	176	Crack in SiC 1/16 in. from bottom
VB-32	R	DD	TI-Kote†	1300	E	4	Hole 1.1 in. from bottom
VB-33	F	DD	DURAK-B + TI-Kote (cemented)	1335	E	109	Hole 0.3 in. from bottom
VB-34	R	DD	Silicon Pyrocarbide†	1300	E	0.5	Hole 0.7 in. from bottom
VB-35	R	DD	DURAK-B + TI-Kote†	1400	E	640(5)	No failure
VB-36	R	DD	Silicon Pyrocarbide†	1350	E	276	Hole in bottom
VB-37*	R	DD	TI-Kote†	1315	E	3	Slit 3/4 in. from bottom
VB-38*	R	DD	DURAK-B + TI-Kote†	1300	E	133	Corrosion at top of thimble

*Test done with company funds
†Hot shell prepared with company funds
(1) F = Flat bottomed, R = Round bottomed
(2) M = Machining, W = Welding, DD = Deep drawing, VD = Vapor deposition
(3) CC = Combustion chamber, E = Emitter
(4) 83 hr > 1300°C
(5) 607 hr at 1300°C, 33 hr at 1400°C

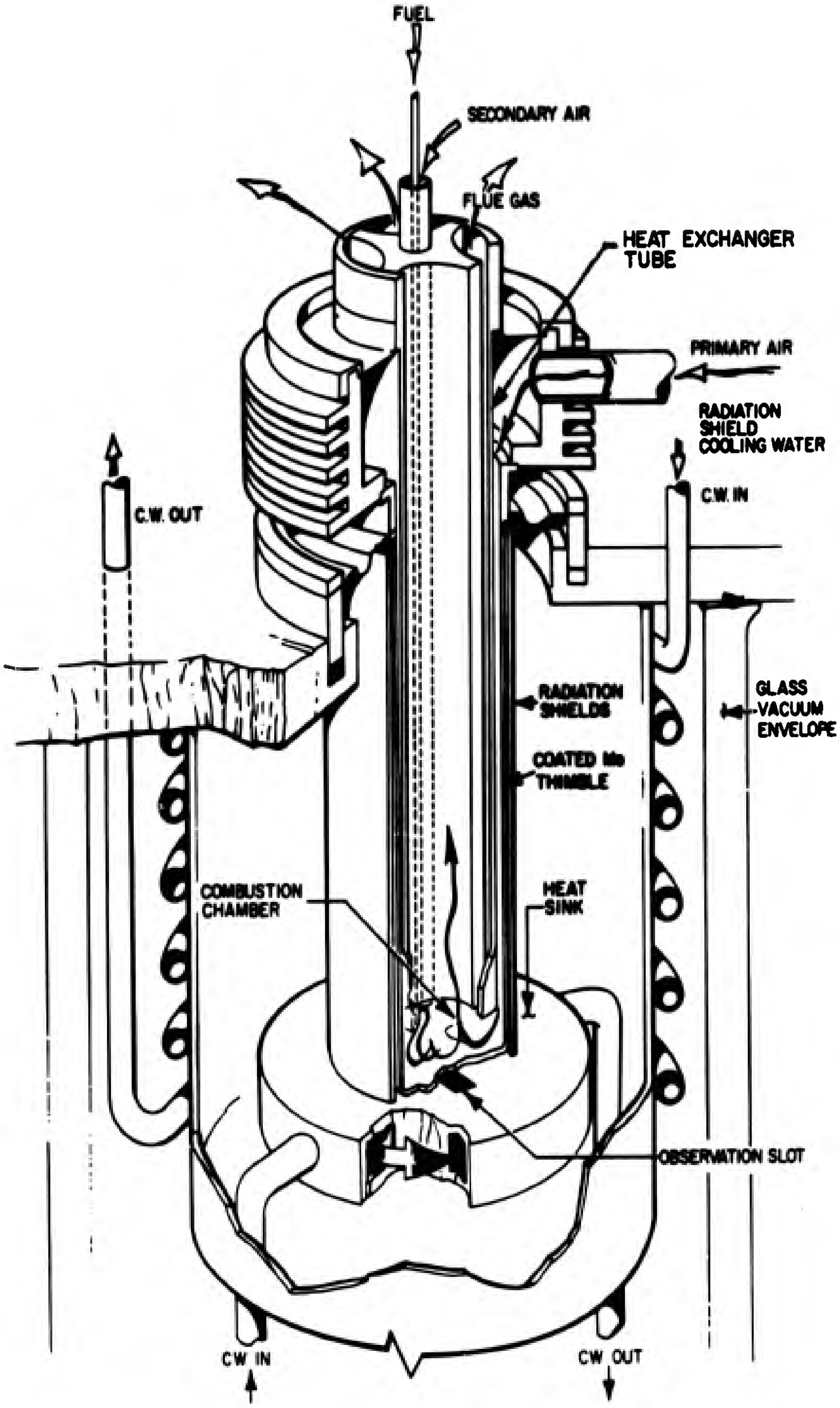

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Figure 9. Vacuum Insulated Burner Experiment

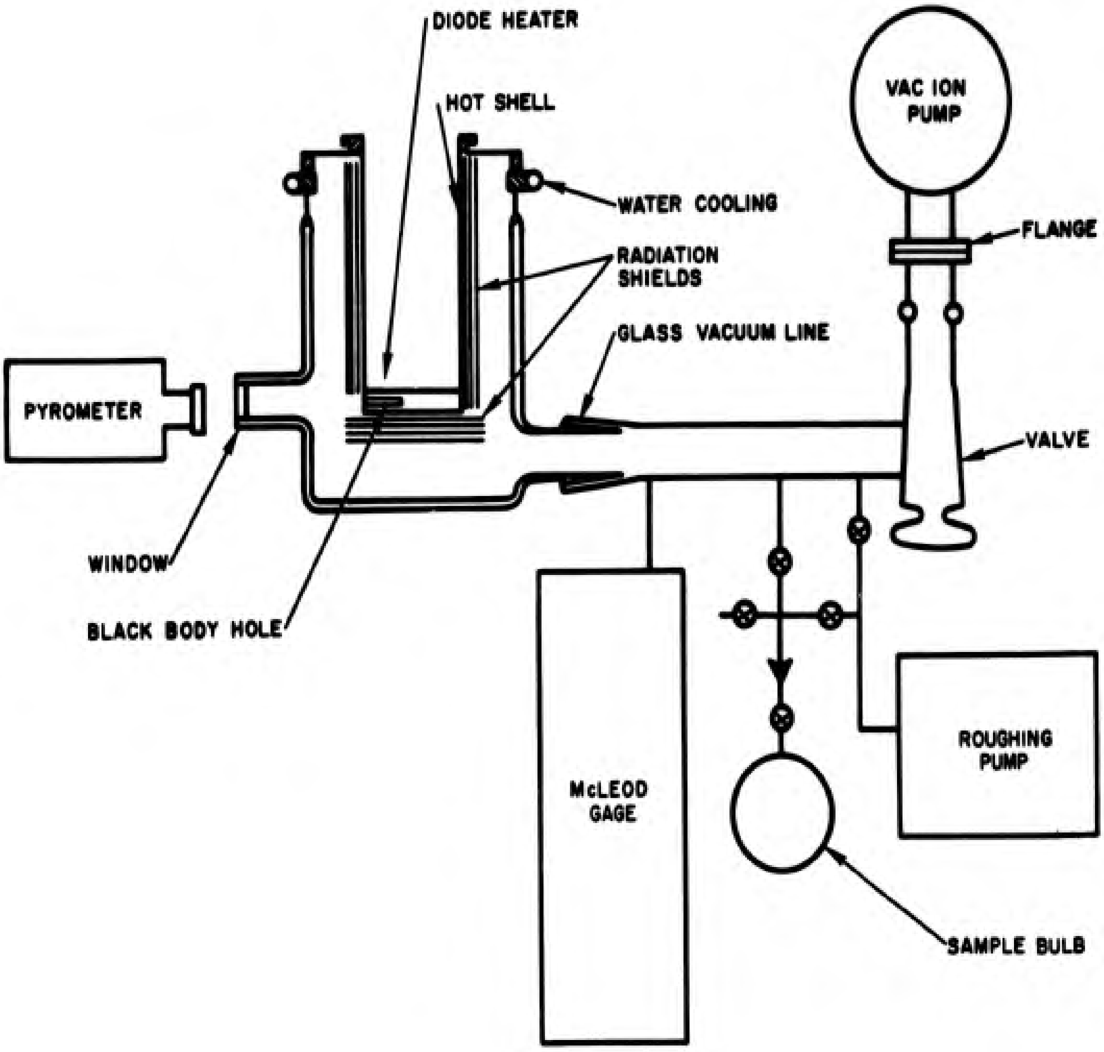

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Figure 10. Hot Shell Test Apparatus

Two of these testers are now in operation and both testers are equipped with an automatic protection circuit so that if electricity, water, or air is interrupted, or if high pressure develops in the vacuum manifold, both fuel and air flow to the burner are stopped, and a small amount of air is diverted through the fuel injector line to keep the injector cool during the slow cooldown. At the same time, the timer is stopped and neither the timer nor the air or fuel can be turned on until the latching relays are reset manually. Therefore, an orderly termination of the test will happen when necessary without attendance. Consequently, the duration of the test is known exactly even though the test fails on an off hour.

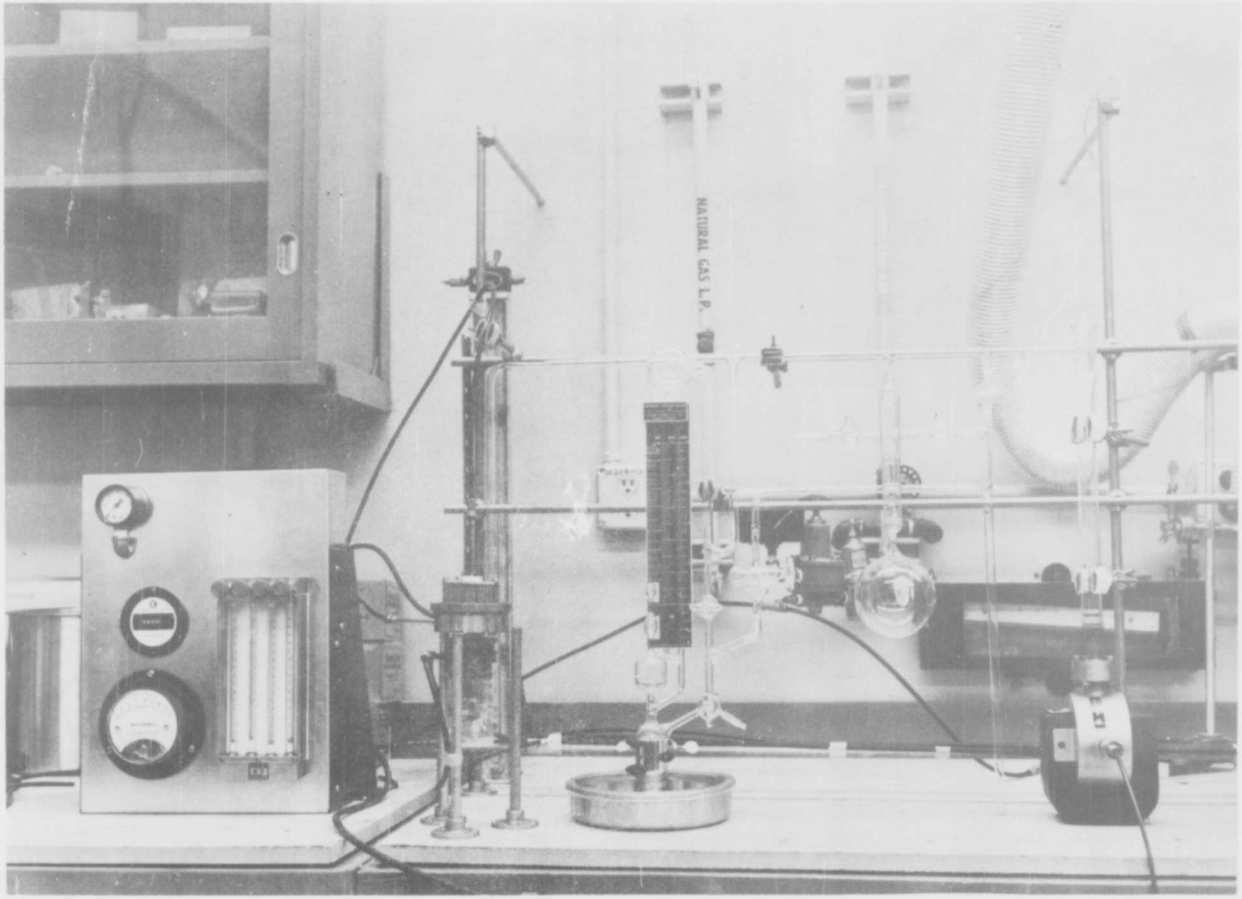

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Figure 11. Run VB-27 Ready for Testing in the Hot Shell Test Apparatus

The very poor performance of the hot shell used in the diodes was due to an error in the preparation of the DURAK-B protective coating. The coating is applied by a pack cementation process to both the inside and outside surfaces of the thimble. The inside surface was then protected by two coats of microstop lacquer. The thimble was then immersed in hydrofluoric acid for removal of the coating from the outside of the thimble. The microstop lacquer evidently had some holes in it which allowed small areas of the surface to be predisposed to failure by the action of hydrofluoric acid. When the DURAK-B coating fails, an area around the failure turns white due to devitrification of the silica which is the primary protective coating. In some of the diode tests, a hole developed without any preliminary devitrification whatever. However, in the vacuum burner tests thimbles were tested which had not been exposed to hydrofluoric acid. These tests showed hot shell lifetimes of up to 100 hr at 1300°C. Note that the 237 hr listed for VB 11 to 14 included only 83 hr in the temperature range greater than 1300°C. Tests numbers VB 22, 27, and 28 were done with one thimble which, although it was protected with silicon carbide on the bottom, failed on the side in a region where flame impingement was not possible.

The use of pyrolytically deposited silicon carbide to protect the molybdenum was given encouragement when VB 10 was run for a period of 42 hr at a temperature of 1300 to 1400°C. There are two possible ways to go, in the use of silicon carbide. One may either start with a thimble of silicon carbide and deposit the molybdenum on to it for the purpose of conducting electricity, or one may start with the molybdenum thimble and deposit the silicon carbide. The hot shell used in VB 10 was constructed by the latter process. The holes which developed in the bottom of the hot shell led to the observation that the silicon carbide does not bond well to the molybdenum. The silicon carbide works because it can be made entirely impervious to gas. Molybdenum has a somewhat higher expansion coefficient than does silicon carbide. Therefore, at ambient temperature, the silicon carbide can easily spall off a flat or a convex surface because of the lack of adhesion between the silicon carbide and the molybdenum. Consequently, some round bottom deep drawn molybdenum cups were obtained from Metallwerk Plansee and some of these were coated with TI-Kote from Texas Instruments, Materials and Sensors Division. These coatings were approximately 0.015 in. thick on the sides and up to 0.050 in. thick across the bottom. Two of these hot shells have been tested in VB 34 and VB 37 and, as can be seen, exhibit very short lives. Evidently the TI-Kote has cracks in it that are invisible to the unaided eye. These cracks may or may not open up when the thimble reaches test temperature but they would at least tend to open up. The molybdenum metal directly underneath the crack is attacked and a long groove develops in the molybdenum. The groove eventually breaks through the molybdenum and a leak occurs. Pyrolytic silicon carbide was also obtained from High Temperature Materials, Inc. They call their products silicon pyrocarbide. Two tests have been conducted with this material. VB 34 lasted only 1/2 hr. Examination of the hot shell afterward showed that the coating has spalled off the molybdenum in two areas at the point of transition between the cylindrical surface and the hemispherical surface. The bare metal under one of these areas had been corroded. VB 36 failed after 276 hr at the point of maximum flame impingement. This silicon carbide coating was 0.005 in. thick. By far, the best results so far have been obtained in VB 35 which has been operated at 1300°C for 607 hr and at 1400°C for an additional 35 hr. The hot shell is being prepared for additional testing as part of a burner test. The point of difference between this hot shell and all the others tested is that it is a 3-layer coating system: molybdenum, molybdenum disilicide (0.003 in. thick), and vapor deposited silicon carbide (0.015 to 0.050 in. thick). It is felt that the molybdenum disilicide in the form of DURAK-B does two things: (1) it promotes a better bond between the molybdenum and the silicon carbide, and (2) it helps combine the high resistance of silicon carbide to flame impingement with the self-healing feature of the molybdenum disilicide. Therefore, if a crack should develop in the silicon carbide, the molybdenum disilicide at the bottom of the crack will protect the molybdenum from oxidation. The molybdenum disilicide will oxidize slowly and fill the crack with silica glass which will greatly impede the diffusion of oxygen through the coating to the surface. It appears that the Mo-$MoSi_2$-SiC system would be more reliable than the simple molybdenum-silicon carbide system because the silicon carbide must be completely impervious if it is to have any lifetime at all. Our experience has been that even pyrolytically deposited silicon carbide can crack under the severe thermal shock of a rapid startup or shutdown. Therefore, the Mo-$MoSi_2$-SiC system would have the advantage with defected silicon carbide.

Pyrolytic silicon carbide from Texas Instruments (TI-Kote) can be obtained in a free standing condition. The availability of this material led us to attempt to make a composite of TI-Kote and vapor deposited tungsten. However, the composite was extremely fragile and the two that were made cracked longitudinally before they could be tested. Vapor deposited molybdenum was then applied over the TI-Kote by San Fernando Laboratories. The first test of this material conducted in VB 16 to 18 resulted in a 78-hr test. The second test, VB 31, resulted in a 176-hr test. This last test failed due to a crack in the silicon carbide near the abrupt transition between the thick, flat bottom of silicon carbide and the thinner cylindrical wall. A number of hot shells are being made ready for test which employ a hemispherical bottom and a uniform wall so that this apparent weak spot will be eliminated. The vapor deposited molybdenum is found to be permanently bonded to the silicon carbide so that no spalling of the silicon carbide is expected.

Phase 2 – Gas Permeation Measurements

As important as durability of a hot shell is, the ability of the hot shell to resist permeation of all gases that might be present in a flame is of equal importance. Consequently, the literature related to gas permeation of possible hot shell materials has been searched. The rate of permeation of a DURAK-B coated molybdenum thimble has been measured in such a way that the absolute permeation rate can be determined. Finally, the actual permeation rate of the number of hot shells being heated on life test has been measured. Permeation of gases through metals or ceramics takes place in three stages:

1) Absorption of the gas on the surface of the solid

2) Diffusion through the solid

3) Desorption from the outer surface of the solid.

Theoretically, any of these three steps can be controlling, but in actual practice the diffusion step is usually controlling.

The relationship expressing the amount of diatomic gas diffusing through a solid metallic barrier is customarily given by the following equation:

$$Q = \frac{DAt}{d}\left(\sqrt{P_1} - \sqrt{P_2}\right)\exp\left(-\frac{E}{RT}\right) \qquad (1)$$

Q = amount of gas permeating the barrier (std cm^3)

D = permeation of constant (cm^2/hr)

A = area of barrier (cm^2)

d = thickness of barrier (cm)

t = time (hr)

P_1, P_2 = pressure on inlet and outlet of barrier (atmosphere)

E = activation energy for process (cal/gm-mole)

R = gas constant (cal/gm-mole-°K)

T = absolute temperature (°K)

For permeation through a ceramic body Equation 1 is used with the square root signs removed. This denotes the fact that the hydrogen must disassociate into atomic hydrogen to pass through metal, while hydrogen will pass through the ceramic as hydrogen molecules. However, all the hot shells to be tested are a composite barrier made up of a metal and a nonmental. Also, the flame is a complex mixture of many chemical constituents including hydrogen, nitrogen, and carbon monoxide. The activity of these species in the flame is not easily determined. Therefore, one has two courses of action. One may devise an experiment which will heat a well-defined area of hot shell material to an accurate temperature, put a specific gas on one side of the hot shell, and determine permeation rate. These rates can then be compared with other rates published in the literature. Or one may build a hot shell intended for use in a flame heated thermionic converter and heat it with the burner intended for this use. The permeation rate determined from such an experiment could not be compared with any literature values but would be directly usable to determine the amount of getter material required to maintain the vacuum for a given period of time.

Both courses have been followed to some extent on this project. A study of the permeation of flame components through a DURAK-B coated molybdenum wall was carried out. A block diagram of the apparatus used is shown in Figure 12. The test capsule used was made by welding a 0.010-in.-wall, 3/4-in.-diameter molybdenum tube to 1/4-in.-thick end pieces. One end piece had a thick walled molybdenum tube welded into it, to serve as a lead tube through

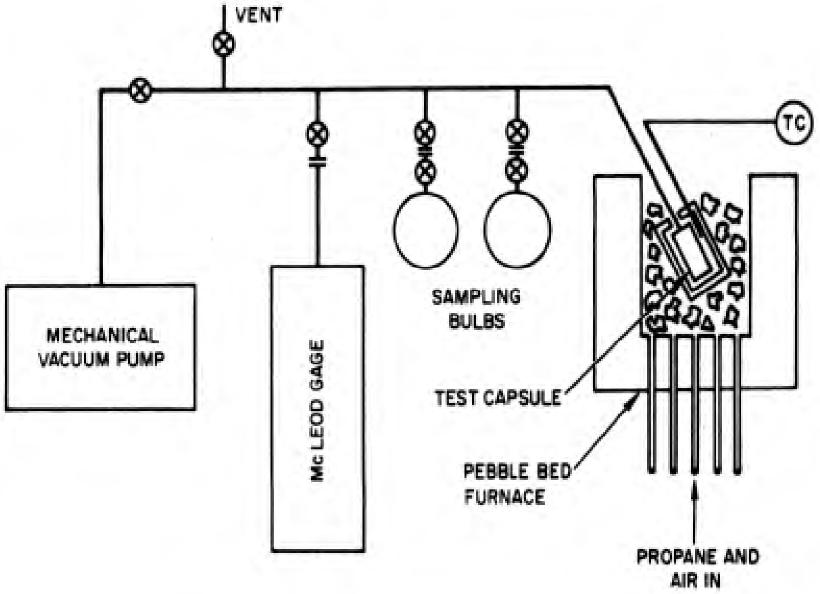

Figure 12. Apparatus for Gas Permeation Measurements

which the sample was taken. The effective area of the thin walled section was 12.8 cm^2. The caps on each tube were coated externally with a 2-mil-thick DURAK-B coating. The coated capsule was brazed to a copper tube which was affixed by means of a kovar-glass seal to the vacuum system manifold. In addition to glass sample bulbs, a McLeod gauge, a vent valve, and a mechanical vacuum pump are connected to the manifold. The capsule was inserted to a KT silicon carbide cup as an auxiliary flame protector. Both were embedded in the pebble bed furnace for heating. A chromel-alumel thermocouple was inserted inside the cup to provide temperature measurement.

The measurements of gas permeation rate were performed by pumping the system down to about 0.001 Torr pressure and determining the time rate of increase of pressure. Two samples of the permeating gas were analyzed. One sample was 99.5% hydrogen and the other was 90% hydrogen. Permeation rate of this material is compared with literature values on the permeation rate of other materials for hydrogen in Figure 13. Notice that materials have extremely wide range of permeability. Also notice that the coated molybdenum samples subjected to a flame have approximately the same permeability as have been reported from pure molybdenum exposed to hydrogen or molybdenum coated with Pfaudler's PFR-6, a molybdenum disilicide coating. In fact, it has been reported[15] that the molybdenum disilicide coating offers little or no resistance to the passage of hydrogen and that the coated molybdenum has the same permeability as the uncoated. DURAK-B and PFR-6 are both molybdenum disilicide coatings. These coatings depend upon a thin layer of silica glass for their primary protection. However, from Figure 13 it appears that silica glass is not much more impermeable to hydrogen than is the coated molybdenum. Pure alumina has been found to be almost completely impermeable to hydrogen.[15] In fact, the method of detection was not adequate to determine the relationship of gas permeability to temperature.

Figure 14 shows that the stainless steel collector cup of the thermionic converter may also pass appreciable quantities of hydrogen. The cup is not exposed to any hydrogen per se but obtains its hydrogen by decomposition of water vapor on the surface and passage of the hydrogen thus produced through the metal into the vacuum space. Note that it makes a considerable difference whether the hydrogen is supplied as a pure gas, or as contained in room air at normal relative humidity, or as contained in especially dry air. Note that in dry air the permeating gas is still hydrogen. Also, it is important to note from Figure 14 that the vitreous coating, Solaramic, on stainless steel greatly reduces the effective permeability and that 347 stainless steel in contrast with 304 or 410 stainless steel develops an oxide coating which is quite impervious to hydrogen permeation. In the flame heated thermionic converter being developed by Atomics International, the collector cup runs in air at 600 to 800°C. If a thin, deep-drawn cup of stainless steel were made of the wrong material, hydrogen permeation through this cup from atmospheric air would be as great as hydrogen permeation through a DURAK-B coated molybdenum hot shell heated to 1400°C.

Hydrogen is not the only gas which permeates molybdenum. Nitrogen and carbon dioxide also have measurable permeabilities. However, nitrogen permeation is down by 2 orders of magnitude under hydrogen and the use of even a very thin molybdenum disilicide coating reduces nitrogen permeation by another 2 orders of magnitude.[15]

In determining whether a given gas permeation rate is tolerable or not, one must consider the capacity of practical getter materials to adsorb hydrogen. For instance, titanium has a theoretical capacity for adsorbing hydrogen of 388 micron-liters per milligram and zirconium has a theoretical capacity of 204.[13] Titanium-zirconium alloy is often used as bulk getter because of the

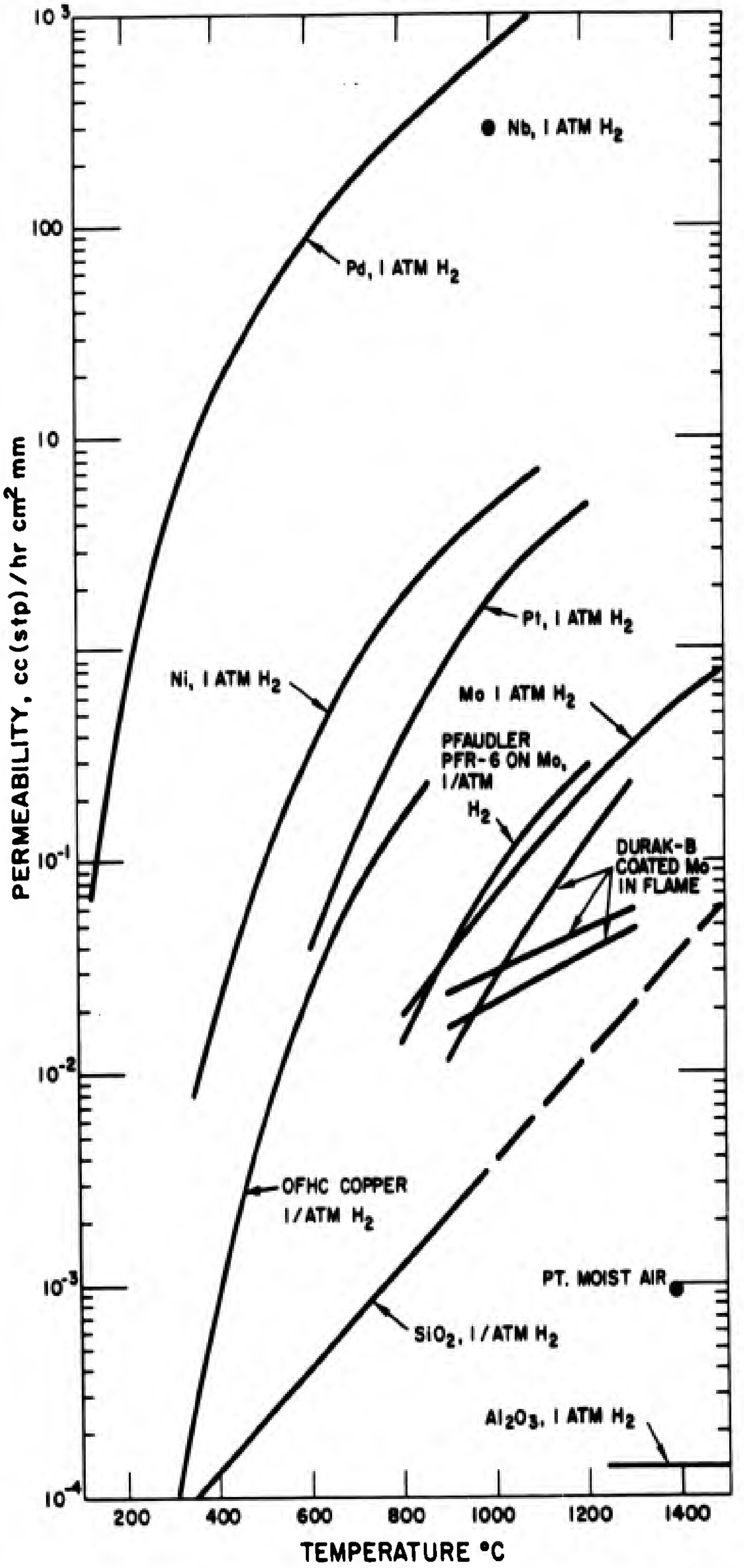

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Figure 13. Permeability of Hydrogen Through Materials (References 2, 3, 6, 12, 13, 14, 15, 16)

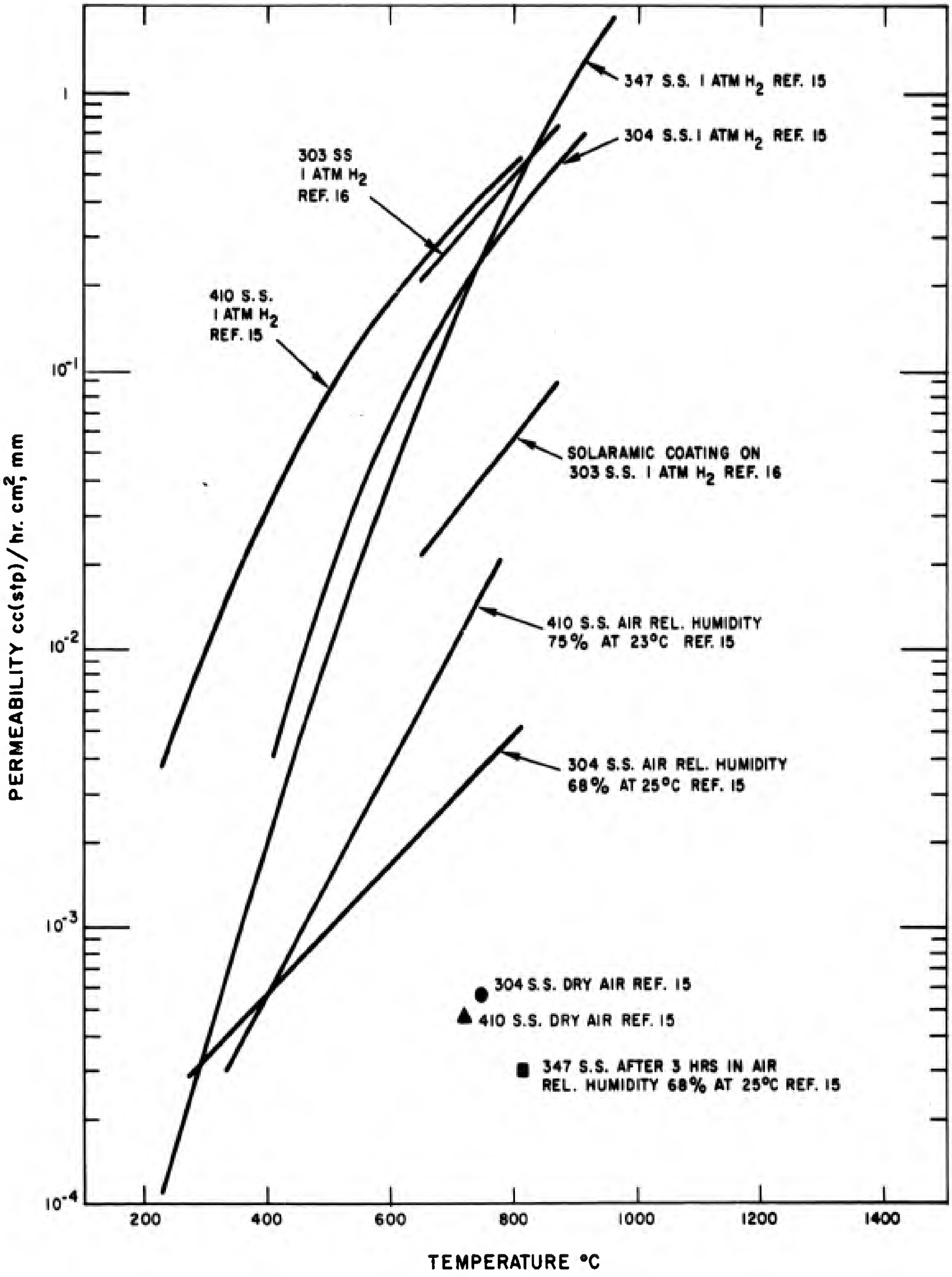

Figure 14. Permeability of Hydrogen Through Stainless Steels

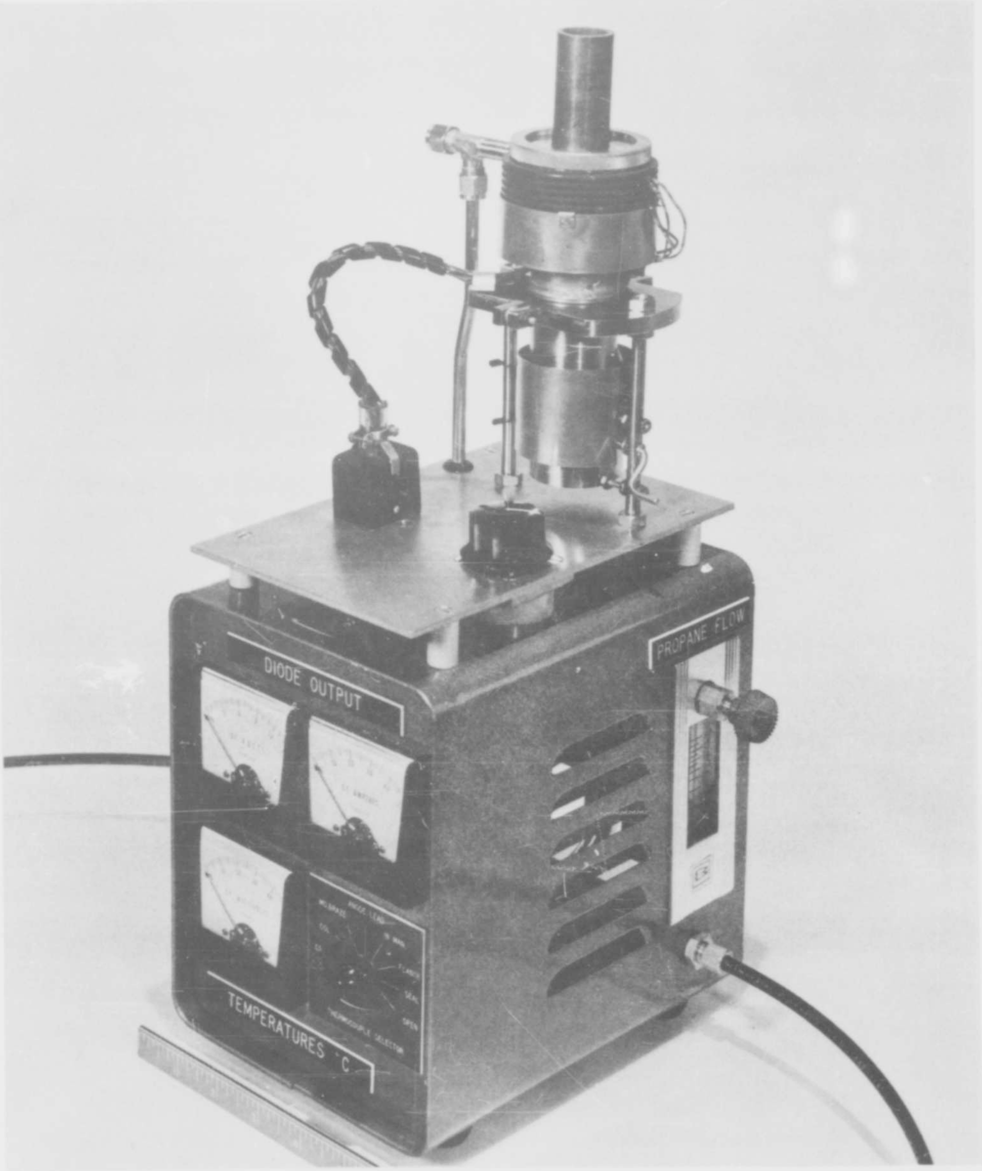

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Figure 15. First Year Sample Product

compounds formed readily diffuse into the body of the metal. Assume that this alloy has a practical capacity of 20 micron-liters/milligram which is about 10% of the theoretical capacity.

Since the new type hot shell testers have been in operation, six hot shells have been tested for an appreciable length of time so that the degassing process is over and the steady-state gas permeation rate can be observed. The results of these tests are summarized in Table X. Notice that the thin DURAK-B and the thin silicon carbide have the highest rate of permeation at thermionic temperatures. Notice also that the lowest permeation rates are usually exhibited by hot shells having a dual protection of silicon carbide and molybdenum disilicide. This rate is converted into grams of titanium-zirconium getter required for 1000 hr of operation providing the titanium-zirconium is assumed to getter only 10% of its theoretical capacity. Several grams of getter can be introduced into the thermionic converter conveniently and is already included as a routine precaution. Care must be taken, however, to make sure that the getter still has adequate capacity after outgassing of the converter.

TASK D – PROTOTYPE DEVELOPMENT

Phase 1 – Thermionic Converter Testing

The 150-watt diode shown in Figure 1 had a maximum electrical output power density of 1.72 watts/cm^2 with an overall efficiency of 7.8% and optimum output voltage of 0.7 volts at an emitter temperature of 1540°C. At 1400°C emitter temperature the power density was 0.9 watts/cm^2. The overall efficiency was 4.3% and the optimum output voltage 0.45 volts. The converter operated for a total running time of 34 hr with an emitter temperature above 1300°C. Of these hours, 10 were above 1400°C. At the end of this time the cesium was completely exhausted from the converter through a leak to the vacuum system. Subsequent examination showed that the converter could not be repaired. At 1540°C about 5 watts/cm^2 should have been obtained instead of 1.72 watts/cm^2. However, on such a large converter it is difficult to obtain the even heating and even spacing required to obtain the 5 watts/cm^2 for molybdenum.

Ten internal flame heated thermionic converters were tested using the portable test stand shown in Figure 15. One of these test stands along with

TABLE X

SUMMARY OF GAS PERMEATION MEASUREMENTS ON HOT SHELLS

Test Number	Hot Shell Description			Emitter Temperature (°C)	Permeation Rate (std* cm^3/hr)	Grams of Ti-Zr Getter Required for 1000 hr†
	Shape	Method of Fabricating Molybdenum	Coating			
VB 28	F. B.	Deep drawn	DURAK-B + KT-SiC (cemented)	1300	0.01	0.38
VB 29	R. B.	Deep drawn	DURAK-B 2-mil thick	1300	0.05	19
VB 31	F. B.	Vapor deposit	Vapor deposit TI-Kote >15-mil thick	1325	0.005	0.19
VB 33	F. B.	Deep drawn	DURAK-B + TI-Kote (cemented)	1350	0.0035	0.13
VB 35	R. B.	Deep drawn	DURAK-B vapor deposit + TI-Kote	1275	<0.035	<1.3
VB 36	R. B.	Deep drawn	Silicon pyrocarbide	1325	0.6	23
VB 38	R. B.	Deep drawn	DURAK-B and vapor deposit TI-Kote	1325	0.003	0.11

*1 atm pressure and 0°C temperature

†Assuming 10% of theoretical capacity

Diode 6 was submitted as the sample product at the end of the first year of the contract. As it has already been explained, flame heated thermionic converters were plagued with hot shell problems and lifetime was too short to obtain any good data. However, a maximum power of 2 watts ($\sim$0.33 watts/cm^2, 10 amp at 0.2 volt) was obtained at a combustion chamber temperature of 1560°C. A cesium temperature of 285°C and a collector temperature of 600°C were observed. Diode 8 obtained a slightly higher power of 3.3 watts (19 amp at 0.17 volt) when the combustion chamber was observed to read 1440°C. The cesium reservoir temperature was 315°C and the collector temperature was 530°C.

On the company sponsored program, a duplicate of the flame heated thermionic converter was constructed and tested using an electron bombardment heater to circumvent the hot shell problem. The highest output power density experienced with this converter was 1 watt/cm^2 (9.13 amp, 0.72 volt) at an observed emitter temperature of 1510°C, an apparent collector temperature of 439°C, and an apparent cesium reservoir temperature of 223°C. One possible explanation for this relatively poor output (as compared to other devices tested in this laboratory) is that the converter is cocked; that is, the emitter and collector are not parallel resulting in an uncertainty of the inter-electrode spacing. Another strong possibility for explaining the poor performance is that the collector temperature is much too hot. Unfortunately, the collector temperature cannot be measured on this device since the collector consists of a molybdenum plate which is held by screws to the inside of the collector cup. Thus the thermal contact may be insufficient to allow the collector surface to attain optimum temperature,while maintaining the collector cup above cesium reservoir temperature. Poor electrical contact may also decrease the output voltage.

Phase 3 — Push-Pull Connection Studies

Some information was gathered using the large electrically heated converter concerning the energy required to turn the diode on and to turn it off again.[3]

Phase 4 — System Design and Construction

One must have some idea where the present line of development would lead to in terms of a final product. For this purpose some design work has been completed on an advanced, internal flame heated thermionic converter. Figure 16 shows this converter as it is presently conceived. Note that the combustion

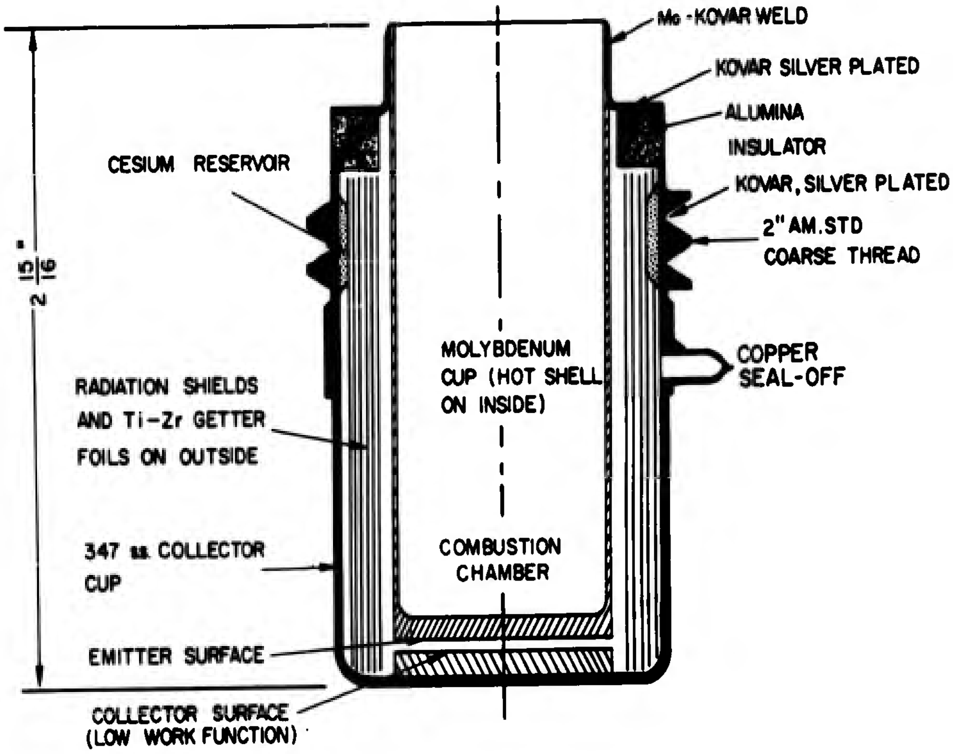

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Figure 16. Internal Flame Heated Thermionic Converter Design

chamber is at the bottom of the molybdenum cup which is protected from oxidation on the inside surface. The outside bottom of the molybdenum cup acts as an emitter. This can be opposed by a specially prepared low work function collector surface. The converter is a fixed space device fitted with threads so it can be screwed into a mounting something like a light bulb. The threaded area also functions as a cesium reservoir. Some of the radiation shields inside the converter are made with titanium-zirconium alloy to act as a getter material.

This type of screw-in diode is then screwed into a diode heater module where it makes contact electrically and thermally (see Figure 17). Note that the copper disc into which the converter screws is fitted with adjustable fins so that under operating conditions the cesium reservoir temperature is correct. Note also that an internal heat exchanger and an external heat exchanger are fitted into an overall module. The first test of this module was accomplished in Run VB 27. This test heater module can be duplicated as many times as desired to give sufficient voltage for power conversion. Figure 17 shows how two modules are hooked together. Figure 18 shows how four modules can be arranged

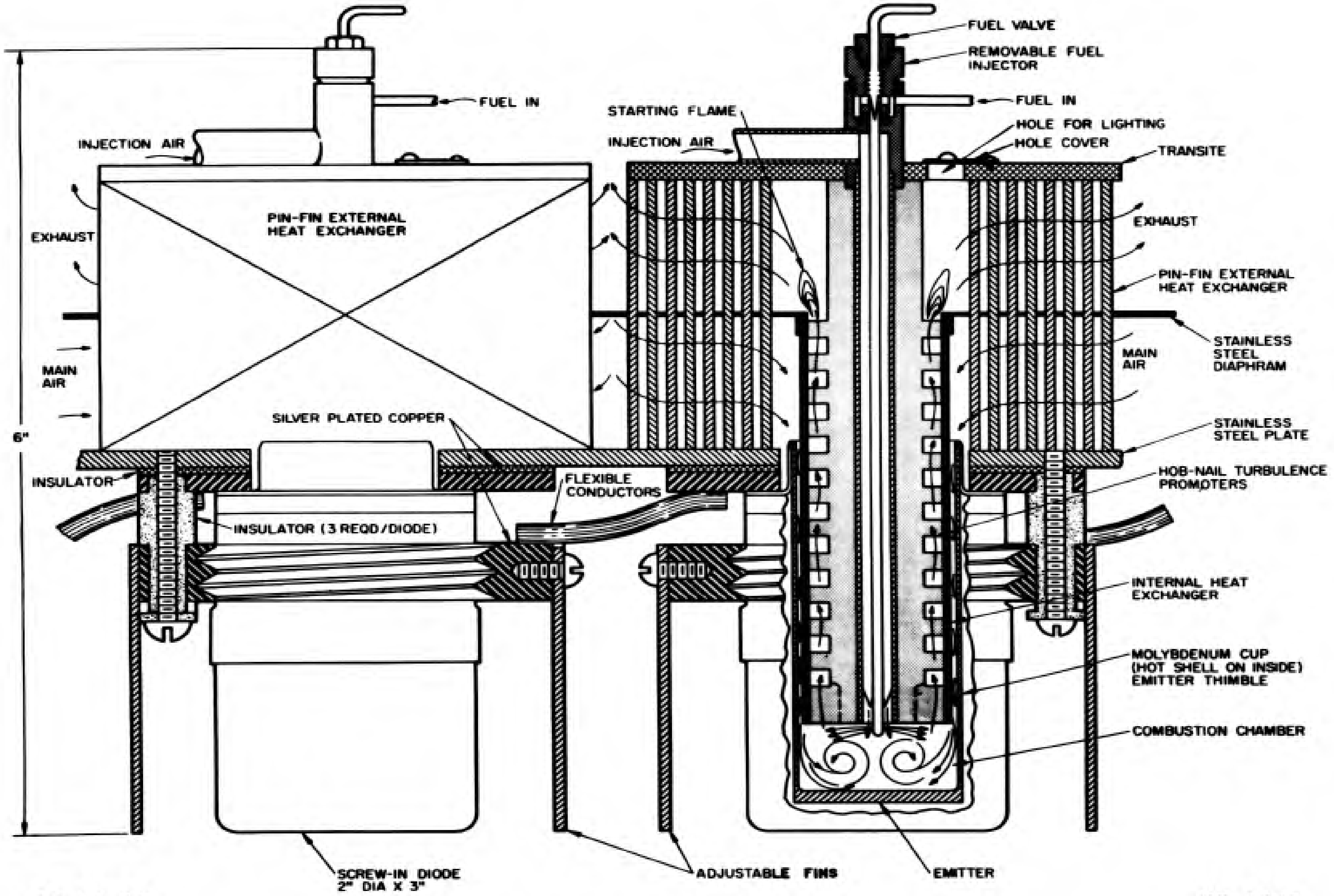

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Figure 17. Flame Heated Converter Modules

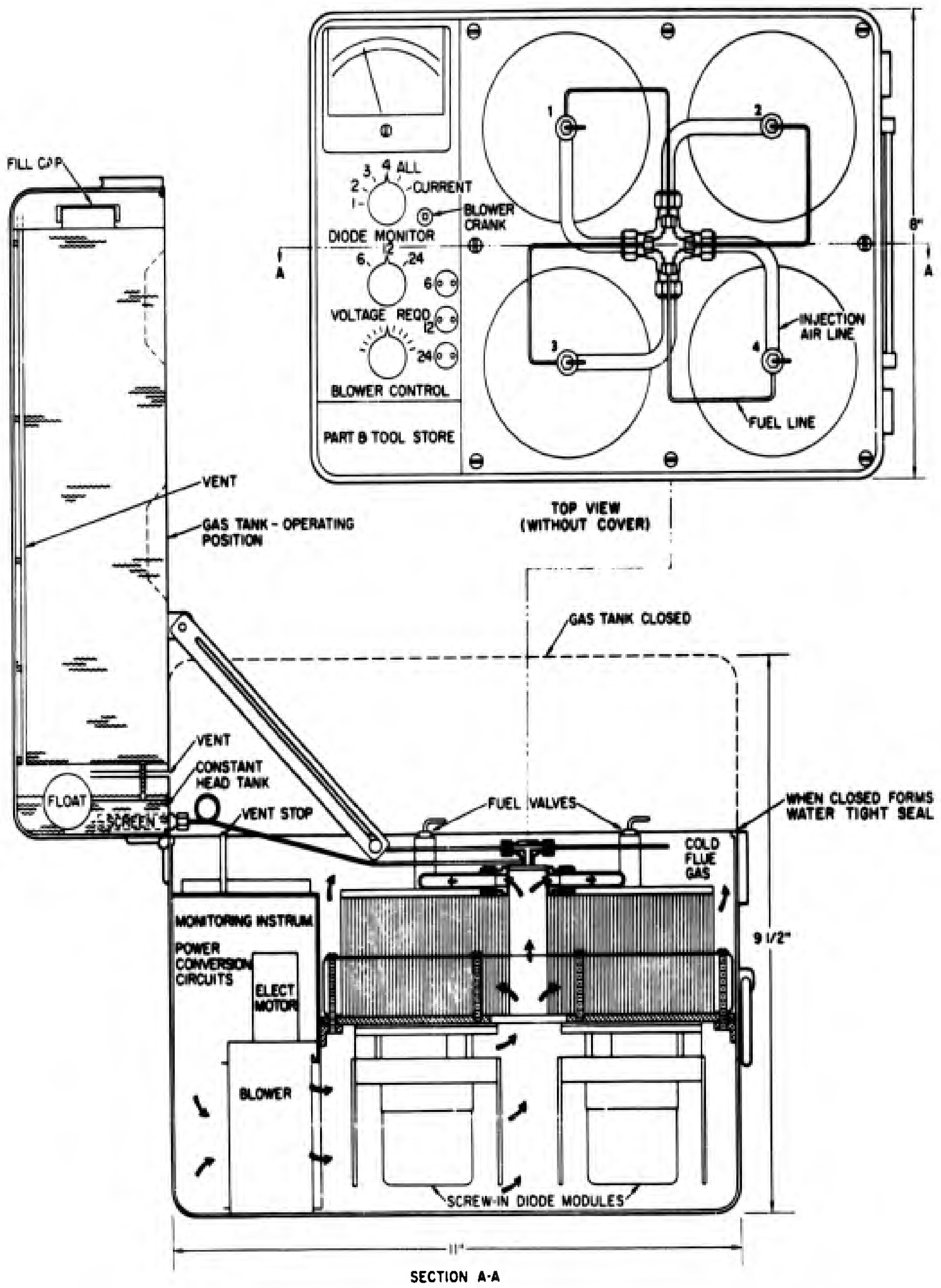

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Figure 18. 100-Watt Thermionic Field Generator

in an overall power source. Each of the four converters has an emitter area of 6 cm^2. If a 5 watts/cm^2 low temperature thermionic converter can indeed be constructed as has already been reported,[17] then a gross power output of 120 watts is possible providing the diodes are perfectly matched. From this, a net power output of 100 watts is possible. A small positive displacement blower is used to supply the combustion and injection air. Gasoline is fed into individually controlled fuel injectors by gravity from a fuel tank in the lid. During operation the positive displacement blower would be powered by an electric motor operated off of the output. During startup batteries may be employed, or the low speed blower may be cranked by hand. The power source can produce either 6, 12, or 24 volts by the flip of the switch. A transistorized low voltage input electrical converter would be included to give the proper voltage outputs. Figure 19 is a better idea of how the conceptual power source would appear in operation.

By increasing the diameter of the hot shell from 1 in. to 2 in., the required electrical energy can be generated in a single thermionic converter operated as an unsymmetrical wave form generator as described by J. Angello of USAELRDL.[18] In the unsymmetrical wave form generator type of electrical converter being developed by W. Dudley of the US Army Electronics R&D Laboratory,[18] the converter is approximately as efficient at 1/2-volt input from one converter as it is from the 2-volt* input expected from four converters connected in series, and the problems associated with series operation of thermionic converters can therefore be eliminated. Also, the module for a 2-in. thermionic converter would be much smaller than the array of four modules for the 1-in. diode. Possibly then, an even smaller and more convenient power source could be developed using a single diode concept. However, the design must be optimized. True, the electrical conversion efficiency is between 70 to 80% for an input voltage of 0.5 to 2 volts, but for a given electrical converter the power converted at maximum efficiency is approximately proportional to the input voltage. Therefore, the electrical converter might be four times as large for 0.5 volt input as it would be for 2 volts input.

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*W. Dudley's paper indicated measurement up to 1.5 volts input. It is assumed that the same observations will hold at least up to 2 volts.

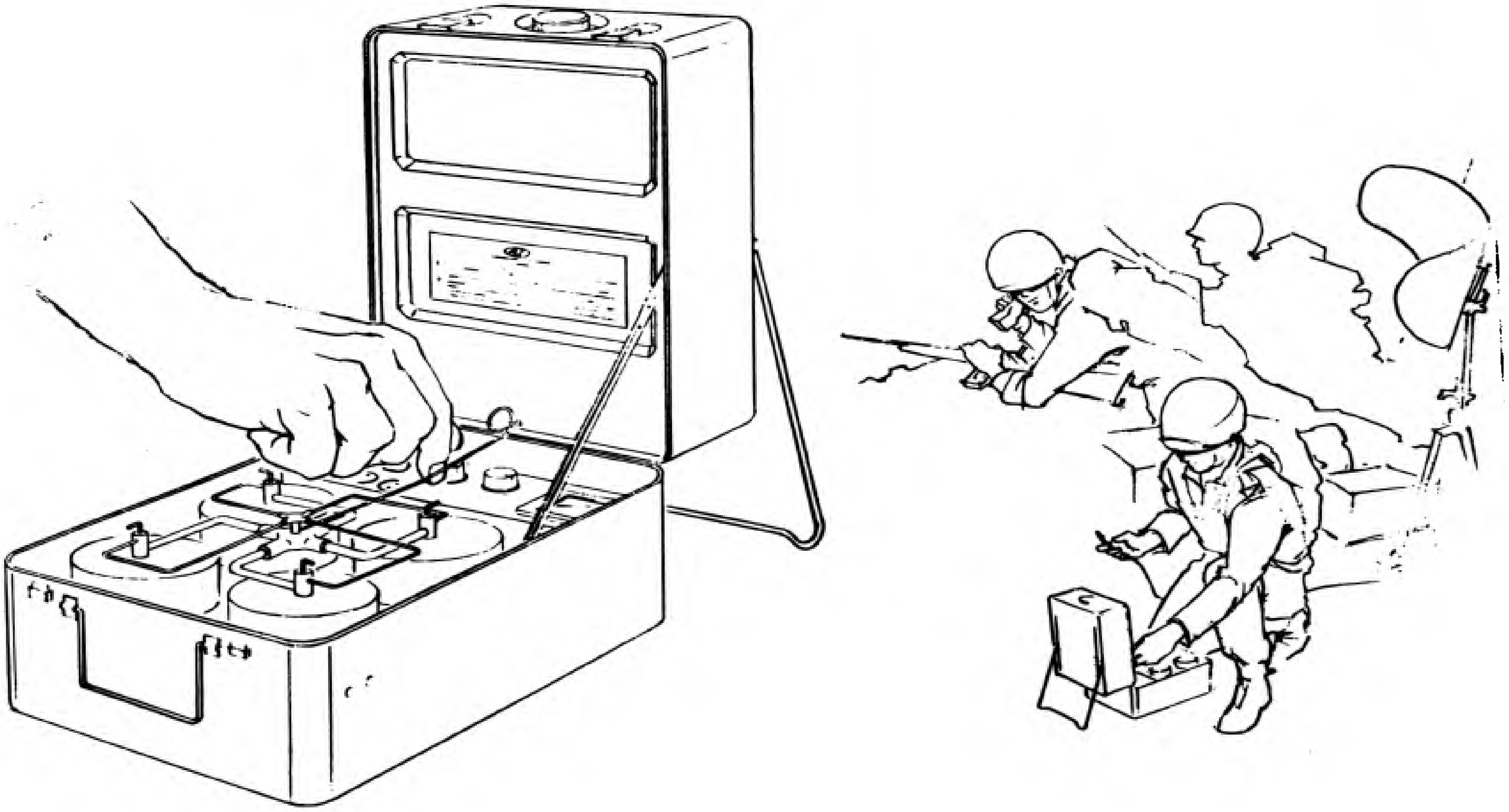

Figure 19. 100-Watt Field Generator in Operation

CONCLUSIONS

1. A hot shell made from deep drawn molybdenum coated on the inside first with molybdenum disilicide and then with pyrolytic silicon carbide is a satisfactory hot shell material. It has endured a temperature of 1300°C for greater than 600 hr while maintaining a satisfactorily low leak rate.
2. A composite of pyrolytic silicon carbide and vapor deposited molybdenum may also prove to be satisfactory hot shell material for an internal flame heated thermionic converter.
3. A fuel injection burner in which the flame is stabilized by converging vortex flow is a satisfactory burner design since it gives complete combustion in an extremely small combustion volume.
4. Pyrolytic silicon carbide and foam silicon carbide are satisfactory materials for which to make the burner an internal heat exchanger. Such materials have endured over 600 hr of operation without requiring repair or cleaning maintenance.
5. Gas permeation measurements of actual hot shell thimbles are conveniently and accurately measured by the pressure-rise method. They give measurements which are immediately applicable to the design of the necessary getters for the sealed-off thermionic converter.
6. The magnitude of gas permeation for a hot shell made from the satisfactory hot shell material is 0.003 cm^3 (stp)/hr at 1325°C. This would conservatively require 110 milligrams of Ti-Zr getter for 1000 hr of operation.

RECOMMENDATIONS

At least two materials are now available for the construction of flame heated thermionic converter hot shells. One of these materials is disclosed in this report and the other one has been reported by F. J. Lyczko.[18] Both of these materials will endure at least 500 hr at thermionic temperatures. The internal flame heated thermionic converter system is best used when a single diode is employed. However, a small number of diodes are a possible solution as shown in Figure 18. With a single or a small number of diodes an electrical converter is required to attain usable voltages. Electrical conversion devices are now being developed for 1/2-volt inputs which have an efficiency in the neighborhood of 70 to 80%. Thus the use of a single converter now becomes feasible and possibly even advantageous. These two very recent developments made the development of a flame heated thermionic power source much more attractive than they previously have been. The steps of development planned by the author are as follows:

1. Complete the construction and test a hot shell test apparatus which includes a heat sink designed to simulate the heat fluxes required in a thermionic converter. With this apparatus, measurements of heating efficiency will be obtained over a range of emitter temperatures (1200 to 1500°C) and over a range of emitter heat fluxes (10 to 40 watts/cm^2).
2. In some of the hot shell tests where the hot shell is a duplicate of a previously tested hot shell, and the gas permeation is known within reason, the getter of action of titanium-zirconium foils on the outside of the radiation shields will be determined during hot shell testing. If the vacuum space around the hot shell can be kept at a low pressure simply by use of getter material, then the getter material will be presumed to function. When the getter material stops functioning then the pressure would rise in this space.
3. After the heater and the hot shell are perfected a variable spaced internal flame heated thermionic converter will be built, possibly with sapphire observation ports so that the interelectrode space and the temperatures of emitter and collector can be observed visually. In this type apparatus the best type of emitter and collector surfaces available will be employed and a respectable power density of >2 watts/cm^2 at 1400°C will be attained. In addition, data

on the thermal expansion of the emitter and collector cups will be taken so that a workable fixed spaced flame heated thermionic converter would be constructed.

4. Using the data obtained from the variable spaced flame heated thermionic converter, a number of fixed spaced thermionic converters would be prepared for evaluation in completed flame heated thermionic power supplies.
5. A prototype thermionic power supply will be designed and constructed for evaluation.

KEY PERSONNEL ASSIGNED TO PROJECT

		Hours Worked During Eighth Quarter
D. H. Adair	Advanced Technical Personnel	462.3
W. R. Martini	Project Engineer	192

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- Thermionics
- Converters
- Diodes
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- Molybdenum Disilicide
- Protective Coatings
- Combustion
- DURAK-B
- Propane
- Gasoline
- Silicon Carbide

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54 pp, 19 illus.
(Report No. AI-8681)
(Contract DA 36-039 SC-88982)

Unclassified Report

The internal flame heated thermionic converter concept has been shown to be feasible. A hot shell to be used on this type of converter has been tested at a temperature of 1300°C for 607 hr and then at 1400°C for a total of 640 hr. The shell is still leak-tight. The permeation rate of gas through this type of hot shell is insignificant, being only 0.003 cm^3 (stp)/hr. The successful hot shell is molybdenum coated first with molybdenum disilicide and then with pyrolytically deposited silicon carbide.

A burner has also been perfected for the internal flame heated thermionic converter. It employs an internal and an external heat exchanger to salvage flue gas heat. Propane is injected into the combustion chamber where mixing and burning are promoted by a converging flow vortex. Heating efficiencies of up to 56% and burner lifetimes greater than 600 hr at 1300°C emitter temperature have been obtained.

A conceptual design showing the advantage of using the internal flame heated thermionic converter is presented.

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Ford Instrument Co. Div. of Sperry Rand Corp. 31-10 Thomson Ave. Long Island City 1, N.Y. ATTN: Mr. T. Jarvis	(1)
The Bendix Corp. Red Bank Division Eatontown, N.J. ATTN: Mr. Joseph E. Sidoti	(1)
General Dynamic Corp. General Atomic Division 10955 John Jay Hopkins Drive San Diego 21, California	(1)
General Motors Corporation GM Defense Research Labs. 6767 Hollister Ave. Goleta, California ATTN: Mr. John Werth	(1)