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FLAME HEATED THERMIONIC
CONVERTER RESEARCH

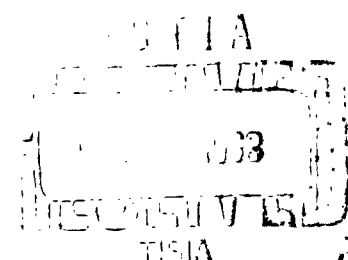
Report No. 5
Fifth Quarterly Report
(1 July 1962 to 30 September 1962)
Contract No. DA-36-039 SC-88982

Department of the Army Task No. 3A99-09-002-04
U. S. Army Electronics Research and Development Laboratory
Fort Monmouth, New Jersey



ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.



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**FLAME HEATED THERMIONIC
CONVERTER RESEARCH**

**Report No. 5
Fifth Quarterly Report
(1 July 1962 to 30 September 1962)**

By

**W. R. MARTINI
E. V. CLARK**

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

Department of the Army Task No. 3A99-09-002-04

**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
ISSUED:**

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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 w 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 w.
- 4) The construction of a sample generator, rated at 100 w output, to demonstrate the feasibility of the design approach.

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, U.S. Air Force
- 5) Company-sponsored research on new emitter and collector materials.

ABSTRACT

TASK A - THERMIONIC CONVERTER DEVELOPMENT

The flame-heated diodes being made for this project are undergoing constant improvement in order to eliminate construction difficulties and operating limitations.

Ten diodes have been constructed so far on the project.

TASK B - HEAT SOURCE DEVELOPMENT

The vacuum-insulated heating experiment was activated to test emitter thimble assemblies in an environment very close to that of an actual diode. The effect of ignition delay and longitudinal heat conduction on burner operation was considered.

TASK C - MATERIALS DEVELOPMENT AND EVALUATION

Materials in addition to Durak-B coated molybdenum are being procured for evaluation. Additional measurements of gas permeation through Durak-B coated molybdenum agree with the previous measurement of 1 std. cc/hr cm² at 1400°C. But under certain conditions lower leak rates are observed.

TASK D - PROTOTYPE DEVELOPMENT

Five flame-heated converters were tested during this report period. Some power was generated. Failure of the Durak-B coating is a continuing problem.

PUBLICATIONS, LECTURES, REPORTS, AND CONFERENCES

1. On July 31, 1962, Dr. W. R. Martini delivered and demonstrated a sample product at USELRDL, Ft. Monmouth. Progress on the contract was also discussed.

2. Dr. Emil Kittl visited Atomics International on August 9, 1962, for the purpose of discussing the contract. He met with Drs. R. C. Allen, D. E. McKenzie, R. L. McKisson, and W. R. Martini.

3. On August 13, 1962, Dr. W. R. Martini went to San Francisco to attend part of the Energy Conversion Conference at the Fairmont Hotel.

TECHNICAL PROGRESS

TASK A - THERMIONIC CONVERTER DEVELOPMENT

Phase 1 - Construction Techniques

Work during this report period has produced further detail changes in the flame-heated diodes described in previous quarterly reports. These changes and results are presented here.

Emitter thimbles have now been obtained by machining, end cap welding, and deep drawing. The first two of these methods have been well evaluated. The improved end cap welding methods have resulted in thimbles that are much cheaper and more reliable than the machined product. Evaluation of deep drawn units is proceeding.

The expansion diaphragm is now made from a new material yielding greater reliability and an improved spring rate.

The emitter thimble is now welded rather than brazed to the center of the diaphragm assembly. Elimination of this braze joint allows higher temperature operation in this region which may occur during burner malfunction.

A simple jack-screw unit has been incorporated into diodes 5, 6, 7, and 8. This jack permits variation of the interelectrode space during operation. Although this feature is not desirable on a production unit, it allows important information to be obtained on the prototypes.

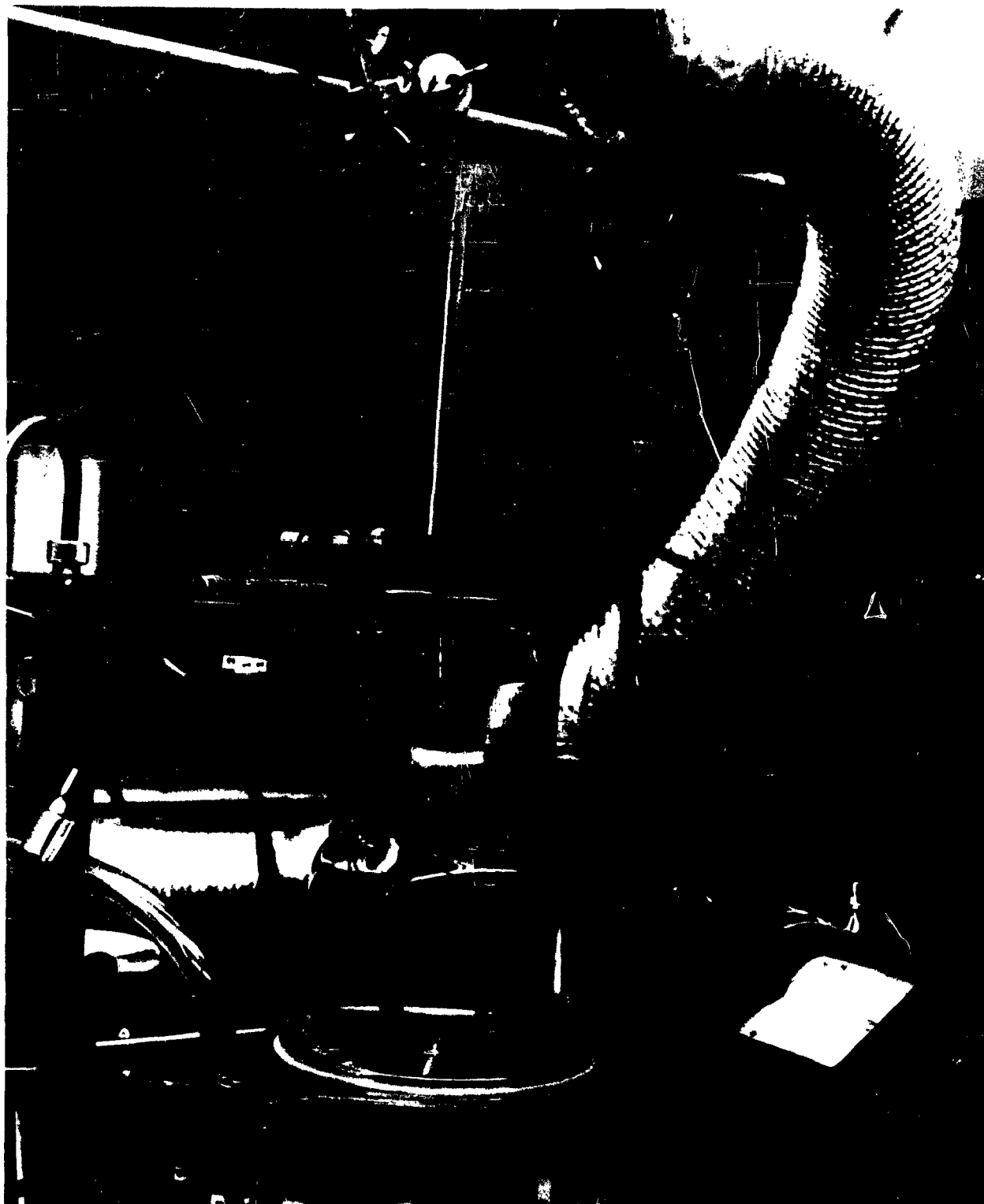
Phase 2 - Diode Construction

Ten internal flame-heated diodes have been constructed so far. Testing of these diodes is described under Task D - Phase 1.

TASK B - HEAT SOURCE DEVELOPMENT

Phase 1 - Premixed Burner Development

Availability of suitable molybdenum thimbles made it possible to activate the experiment which is shown in Figure 1. In the vacuum-insulated heater experiment, the heater and the emitter thimble assembly is identical to that used in the thermionic converter. In fact, the same heater assembly can be tested out in this experiment and then built into a converter if desired. In this



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Figure 1. Vacuum Insulated Heater Experiment

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experiment a water-cooled copper block is in the position normally occupied by the collector. The flowrate and the temperature rise of the water flowing through the block are used to determine the heat transfer from the emitter to the collector. Since simple radiation from the emitter to the collector is usually not sufficient to simulate the high heat flux experienced between the emitter and collector in a thermionic converter, provision is made to allow a measured amount of helium gas into the vacuum space so that proper heat flux can be obtained. One important advantage of this burner test rig is that the combustion chamber can be observed through the outlet tube in the heat exchanger. The emitter surface can be observed through a hole in the heat sink and a window at the bottom of the vacuum envelope. In this way the brightness temperature of the combustion chamber and of the emitter surface can be observed and measured with a pyrometer. The mirrors and the windows were calibrated. The emissivity of the molybdenum emitter surface can be estimated from the spectral emissivity of molybdenum found in the literature. It is apparent, however, from the preliminary data which will be presented shortly that a black body hole is necessary in order to eliminate the emissivity correction. The burner used in the vacuum-insulated heater experiment is shown in Figure 9 of the Fourth Quarterly Report.² This burner has operated for a total of over 14 hours at temperatures up to 1425°C read at the bottom of the combustion chamber. There appears to be a 100 to 200°C difference between the emitter temperature and the combustion chamber temperature depending upon how one makes the emissivity and window corrections. The heat flux between emitter and collector during these experiments was found to be about 10 w/cm². The barrier between the surface of the combustion chamber and the surface of the emitter consists of 1/8 in. of KT-SiC, 1/8 in. of molybdenum, and an indeterminate effective thickness of air between the two layers. At a heat flux of 10 w/cm² the temperature drop through the silicon carbide is expected to be about 10° and through the molybdenum about 3°. Any air blanket between the two can be serious, however, since a 1-mil thickness of air would have a temperature difference of 23°C if only gas conduction were considered and radiation interchange neglected.

Phase 3 - Theoretical Studies on Heat Economy and Combustion

When the internally flame-heated diode was planned (see Figure 2 of the Fourth Quarterly Report²), it was always realized that the heating efficiency could not be very high with such a rudimentary type of heat exchanger between

the outgoing combustion gases and the incoming fuel and air mixture. It was also realized that the fuel and air mixture preheat must be limited or preignition would take place, and that this preignition limit was somewhat dependent upon how fast the fuel-air mixture was heated. It is further apparent from the present diode heater design that longitudinal heat conduction is an important effect which is usually overlooked in most heat transfer calculations.

Heat Exchanger Analysis

The case of a regenerative heat exchanger, shown diagrammatically in Figure 2, was considered. The following assumptions are made:

- 1) Heat transfer between the annulus and the heat exchanger tube and the hot shell is very rapid so that at any longitudinal location (x) the temperatures of the hot shell, heat exchanger tube, and the gas mixture in the annulus are the same.
- 2) Heat transfer between exhaust gas and incoming mixture is adequately represented if an average is assumed of the stack gas temperatures entering and leaving the heat exchanger. Actually, the assumption of a varying stack gas temperature still leads to an analytical solution, but for the problem that will be presented here the extra complexity is not warranted.

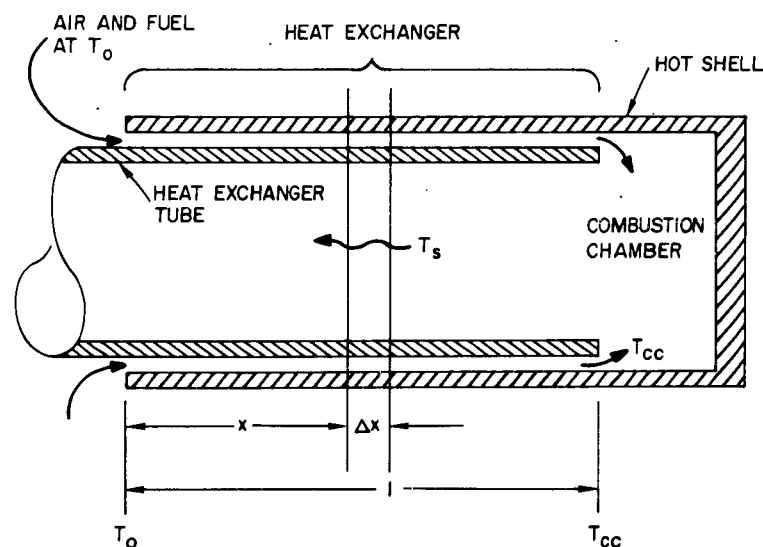


Figure 2. Nomenclature for Heat Exchanger Analysis

The heat increase in the gas mixture passing from point x to point $x + \Delta x$ is given by

$$\Delta Q_M = w C_p \Delta T \quad \dots(1)$$

where

w = mass rate of flow, g/sec

C_p = mixture heat capacity, j/g°C

ΔT = gas mixture temperature rise in space Δx , °C.

This heat is supplied in part by longitudinal conduction in the heat exchanger tube and the hot shell according to the expression

$$\Delta Q_c = K \Delta x \frac{d^2 T}{dx^2} \quad \dots(2)$$

where

$$K = k_{HS} A_{HS} + k_{HE} A_{HE}$$

k_{HS} , k_{HE} = thermal conductivity of hot shell and heat exchanger material respectively, w/cm°C

A_{HS} , A_{HE} = heat conduction area for hot shell and heat exchanger respectively, cm².

The rest of the heat comes from the stack gas according to the expression

$$\Delta Q_{HT} = U \pi D \Delta x (T_s - T) \quad \dots(3)$$

where

U = overall heat transfer coefficient based upon the inside diameter of the heat exchanger, w/cm²°C

D = inside diameter of heat exchanger, cm

T_s = average stack gas temperature, °C.

Now by a heat balance

$$\Delta Q_M = \Delta Q_c + \Delta Q_{HT} \quad \dots(4)$$

Substituting Equation 1, 2, and 3 into Equation 4 one obtains

$$wC_p \Delta T = K \Delta x \frac{d^2 T}{dx^2} + U \pi D \Delta x (T_s - T) \quad \dots(5)$$

This reduces to the differential form

$$wC_p \frac{dT}{dx} = K \frac{d^2 T}{dx^2} + U \pi D (T_s - T) \quad \dots(6)$$

The solution to Equation 6 is:

$$T = C_1 \exp\left[\frac{A+B}{2} x\right] + C_2 \exp\left[\frac{A-B}{2} x\right] + T_s \quad \dots(7)$$

where C_1 , C_2 = constants determined by boundary conditions

$$A = wC_p / K$$

$$B = \left[A^2 + \frac{4U\pi D}{K} \right]^{1/2}$$

With Equation 7, the temperature profile for any specific heat exchanger configuration and set of boundary conditions can be computed. As an example, the temperature profile in a heat exchanger similar to that shown in Figure 2 was computed. The parameters were:

$$K = 0.527 \text{ w/cm}^\circ\text{C}$$

$$U = 6.1 \text{ Btu/hr ft}^2\text{ }^\circ\text{C} = 0.00346 \text{ w/cm}^2\text{ }^\circ\text{C}$$

$$D = 2.42 \text{ cm}$$

$$w = 0.513 \text{ g/sec (1200 w heat input)}$$

$$C_p = 1.00 \text{ j/g}^\circ\text{C}$$

$$T_s = 1525^\circ\text{C}$$

$$l = 6.35 \text{ cm}$$

From these parameters the solution shown in Figure 3 was computed for the boundary condition $x = 0$, $T = 400^{\circ}\text{C}$, and $x = 6.35$, $T = 1400^{\circ}\text{C}$.

Note that for the sample solution shown in Figure 3, very little heat is conducted to the surroundings from the entrance to the heat exchanger because the temperature gradient at this point is small. It also is apparent that under these conditions most of the air mixture preheat is accomplished by heat

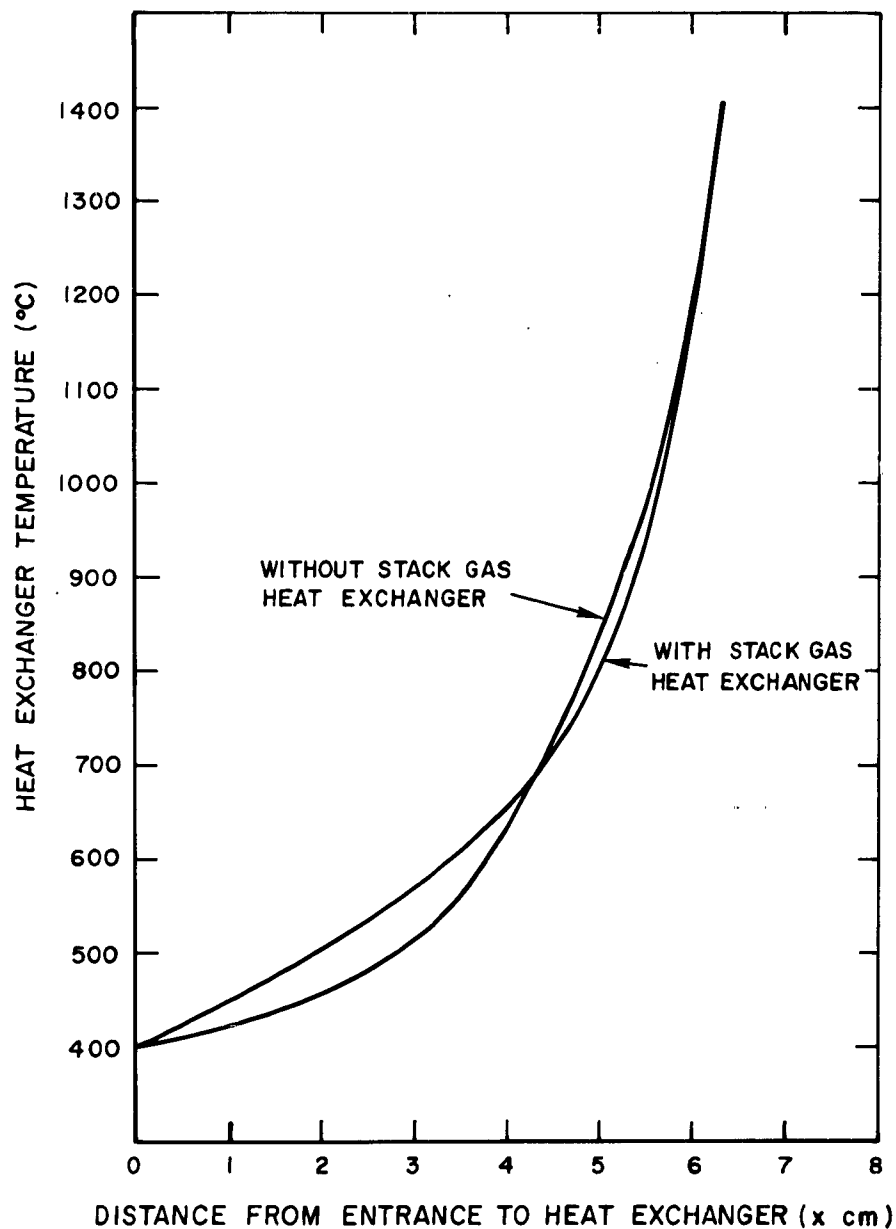


Figure 3. Calculated Heat Exchanger Temperature Profile
(Sample Problem)

conduction from the combustion chamber along the walls and along the heat exchanger tube. In Figure 3 the results of a calculation in which heat transfer from the stack gases was ignored is compared with the sample solution. The similarity of the results is shown. At any rate such a situation leads to a very rapid increase in gas mixture temperature in the range where ignition of the gas can take place spontaneously.

Ignition Delay

During this report period some studies were made concerning the phenomenon of ignition delay in propane-air and gasoline-air systems. A survey of the recent literature was made and pertinent information was applied to the present system. Measurements by various investigators of the ignition delay for various fuels are compared in Figure 4. It is shown that methane has the longest ignition delay and hydrogen the shortest. For some other materials like commercial butane, benzene, and propane, ignition delay has been measured by two different investigators, and the results do not agree well.

The experimental techniques were not identical, however, so that some disagreement would be expected. Mullins^{3,4,5} used a pre-burner to obtain the required air temperature before injection of the test fuel. The higher the air temperature, therefore, the lower the percentage of oxygen in the air and the higher the percentage of combustion products. Salooja⁶ mixed the fuel with nitrogen and then mixed the nitrogen-fuel mixture with oxygen in a very small mixing chamber. The mixture then passed down a Kovar tube to a vacuum pump. The pressure of the tube was raised until the flame passed down the tube. Brokaw⁷ used a similar experimental apparatus but used heated air to bring the apparatus to thermal equilibrium and then injected propane and measured the time from fuel injection to ignition in the Kovar tube downstream of the mixer. Chang and others⁸ used a preheated air stream and fuel injection at the beginning of the combustion tube. The injector was designed so that mixing between fuel and air occurred instantaneously in comparison with the time required for ignition due to ignition delay. The distance between the fuel injector and the flame, combined with a flowrate and temperature of the mixture, was used to compute the ignition delay.

The ignition delay for propane as measured by Chang was approximately the same as that for kerosene vapor as measured by Mullins, benzene

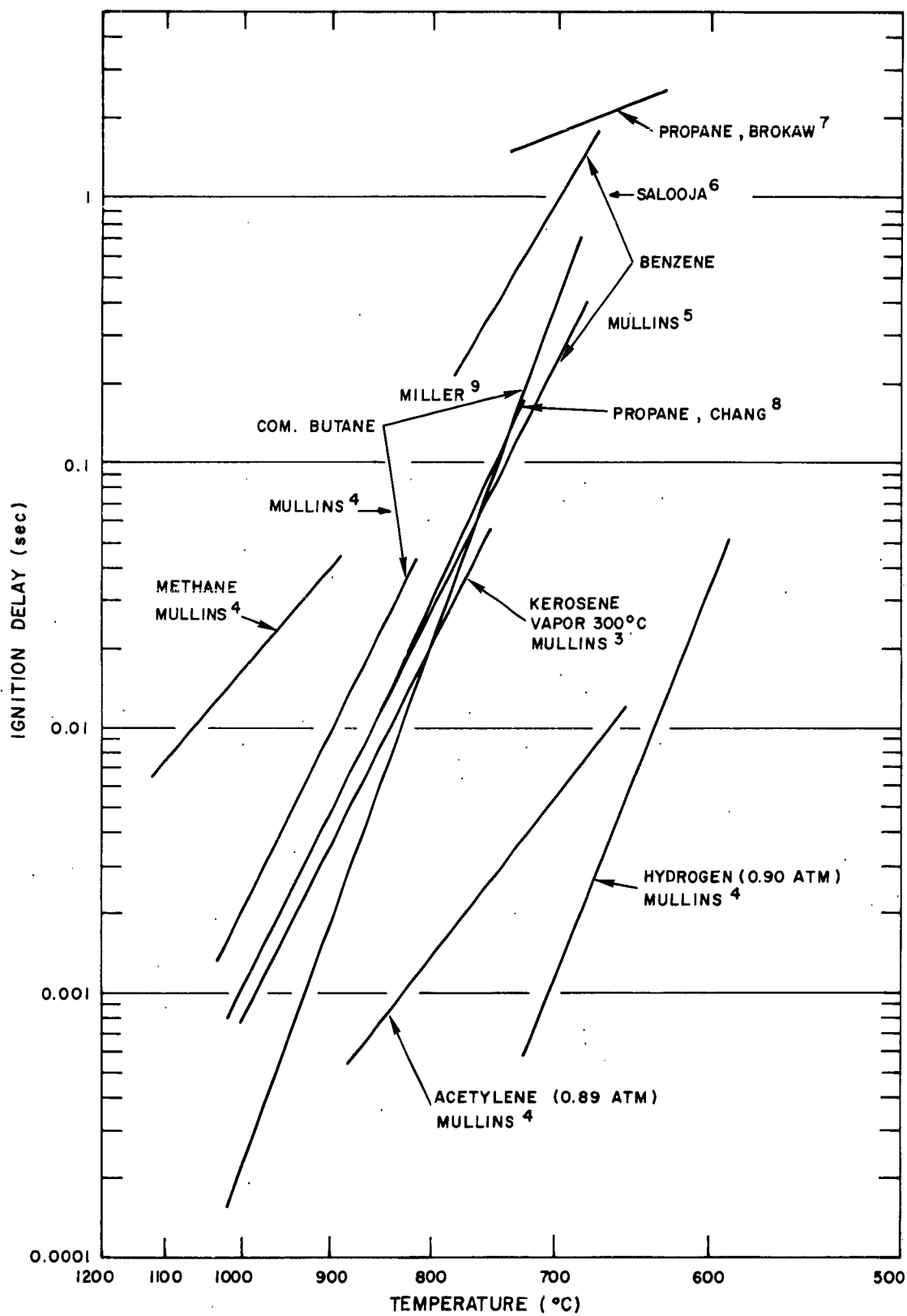


Figure 4. Ignition Delay for Various Fuels Burning in 0% Excess Air

measured by Mullins, and commercial butane measured by Miller. One would expect that the ignition delay for gasoline would also lie near this same grouping.

For the example presented in the previous section the relation of heat exchanger temperature to the distance from the cold end is given by the equation

$$T' = 648.6 + 24.4 e^{0.56x} \quad \dots(8)$$

where T' = temperature, °K.

The volumetric flowrate, v of the gas mixture is:

$$v = 0.513 \text{ (g/sec)} \frac{(22,400)}{29} \frac{T'}{273} = 1.45 T' \text{ ml/sec} \quad \dots(9)$$

The flow area of the annulus is 0.42 cm^2 . Thus a differential volume of gas in the annulus $dV = 0.42 dx$, and the residence time of gas in volume dV is

$$\frac{dV}{v} = \frac{0.42dx}{1.45T'} = 0.289 \frac{dx}{T'} \quad \dots(10)$$

When the data of Chang⁸ is used as representative, the time to ignition as a function of temperature is given by the following equation.

$$\tau = 0.845 \times 10^{-11} e^{23,600/T'} \text{ sec} \quad \dots(11)$$

Now, if F is the fraction of ignition time accumulated along the exchanger, then

$$dF = dV/v\tau \quad \dots(12)$$

By substituting Equations 8, 10, and 11 into Equation 12, one obtains

$$dF = \frac{0.289}{(648.6+24.4e^{0.59x})(0.845 \times 10^{-11} \exp \left[\frac{23,600}{648.6+24.4e^{0.59x}} \right])} dx \quad \dots(13)$$

$$dF = G(x) dx$$

The function $G(x)$ was evaluated and $G(x)dx$ was integrated graphically to obtain F as a function of x in Figure 5. When $F = 1$, ignition is assumed to take place. This assumption ignores the important wall effect. The sample problem assumes that the mixture is being heated in an annulus 0.020 in. thick whereby the data presented in Figure 4 were taken for cylinders 1 to 2 in. in diameter. In addition to the effect from the presence of the walls, the coating on the wall sometimes has a large effect. Bardwell¹⁰ found that lead oxide (PbO) coating the surface of a silica container suppressed cool flames and low temperature ignitions over a wide range of experimental conditions. Nevertheless, if no wall effect is assumed, a comparison of Figures 3 and 5 shows that for the sample problem, ignition would take place at 1240°C.

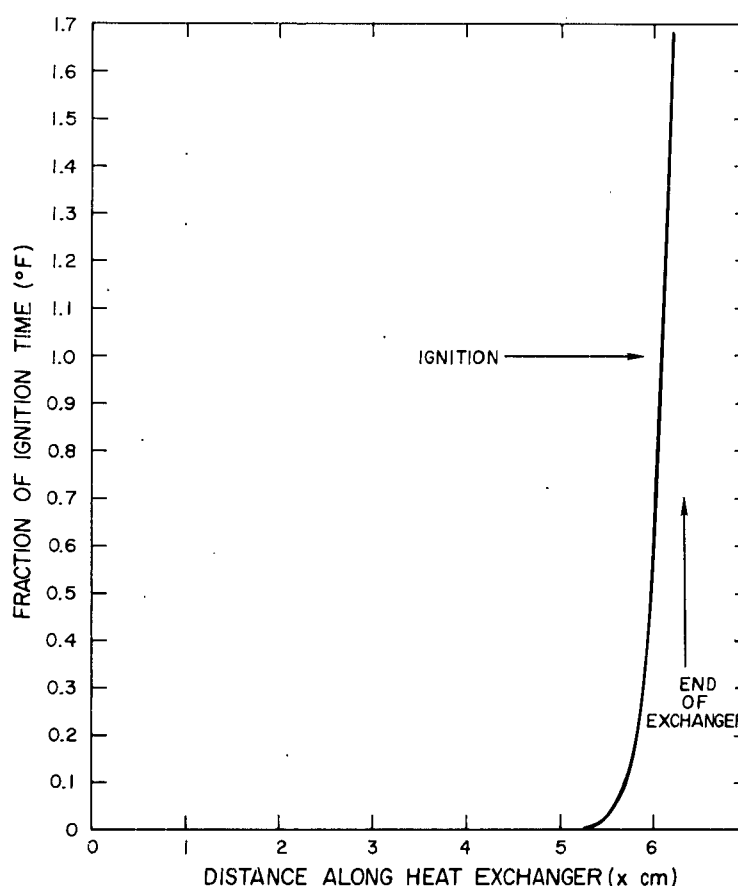


Figure 5. Calculated Ignition Point
(Sample Problem)

TASK C - MATERIALS DEVELOPMENT AND EVALUATION

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

Although a number of suppliers have been contacted in order to develop alternate sources for refractory metals and anti-oxidation coatings, no testing of anti-oxidation coatings was performed.

Phase 2 - Gas Permeation Measurements

The gas permeation experiment described in Task C, Phase 3 of the Fourth Quarterly Report² was repeated during this report period with a duplicate test capsule. The data from this second experiment are shown in Figure 6 compared with the line drawn through the previous data. The results in some cases are considerably lower than those obtained in the first test capsule and in

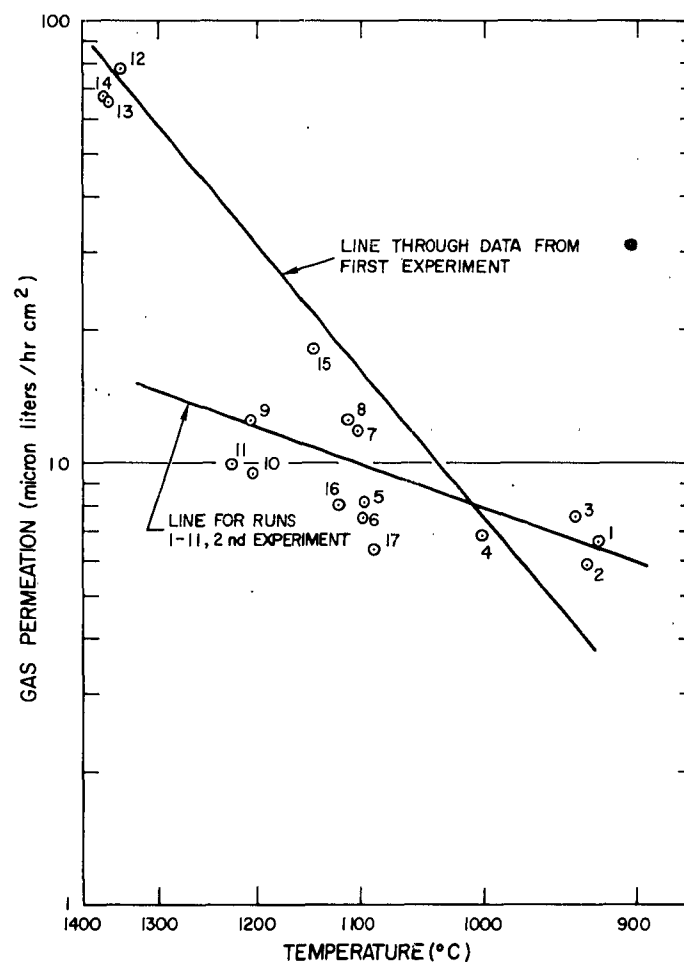


Figure 6. Second Gas Permeation Experiment

some cases agree substantially with the results of the first test capsule. The numbers next to the points in Figure 6 indicate the order in which the determinations were made. R. L. McKisson, who performed this experiment, suspects that many hours may be required for the true permeation rate at any particular temperature to become apparent. That is, on heating to a high temperature the permeation rate through the Durak-B coating increases. However, the increased solubility of hydrogen in the molybdenum allows the molybdenum to absorb the increased gas permeation for several hours. A considerable number of hours is required, therefore, before the molybdenum is saturated with hydrogen and hydrogen is being given off into the vacuum space at the same rate that it is entering the molybdenum metal. Conversely, during cooling, the molybdenum metal is loaded with hydrogen which comes out relatively slowly and time is required before a low permeation rate is again observed. This hypothesis is substantiated somewhat by the second gas permeation experiment.

TASK D - PROTOTYPE DEVELOPMENT

Phase 1 - Testing Flame Heated Converters

Diodes 1, 2, and 3 were tested and described in the previous report.² Testing of diodes 4 through 8 has been completed and is described here.

Diode No. 4 was operated as a vacuum diode in order to test burner configurations under operating conditions. Several runs were completed, then the diode was disassembled for examination.

In Diode No. 5 a leak developed in the diaphragm-cathode braze after the second run. A number of flashbacks had badly overheated this braze, indicating need for burner modifications. Some changes in the radiation shields were also required to trim the temperature distributions.

Diode No. 6 incorporated all of the changes indicated by the No. 5 runs and was delivered to USARDL and demonstrated. One run was made with a 10-amp output. Difficulty with Cs reservoir temperature control prevented full power generation.

Diode No. 7 was a duplicate of No. 6, but the emitter protective coating failed immediately upon startup.

Diode No. 8 was operated intermittently for six hours at various power levels. Currents as high as 15 amps were observed even though additional trouble with cesium reservoir temperatures precluded optimum operation. Volt-ampere curves were run which gave promising results. A failure in the emitter coating stopped the operation.

Based upon visual examination of the coating failures and incipient failures in Diode No. 8, there is a good possibility that metallic silicon from the KT-silicon carbide may be contributing to premature failure.

CONCLUSIONS

1. The phenomenon of ignition delay theoretically allows premixed fuel-air to be heated to a high temperature before combustion begins.
2. Failure of flame-heated diodes by corrosion through the Durak-B is a serious problem.

PROGRAM FOR NEXT QUARTER

•TASK A - DIODE DEVELOPMENT

Evolutionary improvement of the present diode design will continue.

TASK B - HEAT SOURCE DEVELOPMENT

Higher efficiency burner designs will be developed by employing more efficient air preheaters. The vacuum insulated heating experiment will be used to simulate closely an actual diode. Some burner designs for leaded gasoline will be tried.

TASK C - MATERIALS DEVELOPMENT

New types of coatings for Mo and other refractory metals will be evaluated first as a coating for an electrically heated wire and second as a coating for the inside of the emitter thimble.

TASK D - PROTOTYPE DEVELOPMENT

A multi-diode power source will be designed to guide development work.

Series connection and push-pull connection studies will be conducted if diodes are available.

KEY PERSONNEL ASSIGNED TO PROJECT

		Hours Worked During Fifth Quarter
E. V. Clark	Research Engineer	345.5
R. G. Cole	Mechanic-Engineering Laboratory, Junior	359.0
W. R. Martini	Project Engineer	470
R. L. McKisson	Project Engineer and Supervisor	159.5
A. J. Riccio	Advanced Technical Personnel	297.5
J. D. Nogý	Senior Mechanic	357.4

ADDITIONAL BIOGRAPHY

A. J. Riccio Advanced Technical Personnel

Education

<u>College</u>	<u>Year</u>
Massachusetts Radio School	1953-1954
Fullerton Evening J.C.	1954-1958

Experience

<u>Place</u>	<u>Year</u>	<u>Field</u>
Beckman Instr. Fullerton, California	1954-1959	Fabrication, processing, and testing of phototubes, mercury and hydrogen discharge tubes.
Hughes Aircraft, Microwave Tube Lab., Los Angeles, California	1959-1961	Research and development of metal and ceramic microwave tubes.
Atomics International	1961-	Development of fabrication techniques for experimental and prototype thermionic converters.

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AD Atomics International, Canoga Park, Calif. FLAME HEATED THERMIONIC CONVERTER RESEARCH by W. R. Martini, E. V. Clark 5th Quarterly Report, 1 July - 30 September 1962 37 pp, 6 illus., 10 refs. (Report No. AI-7841) (Contract DA 36-039 SC-88982) Unclassified Report The purpose of this research is to develop the technology required for portable, flame-heated thermionic power sources. Active development in thermionic diodes, heat sources and materials is underway. The flame-heated diodes being made for this project are undergoing constant improvement in order to eliminate construction difficulties and operating limitations. Ten diodes have so far been constructed on the project. (over)	UNCLASSIFIED 1) Combustion Chambers 2) Combustion Chamber Liners 3) Molybdenum Compounds and Silicides 4) Silicon Compounds and Carbides 5) Inverter Rectifiers 6) Diodes (Electronic Tube Devices) - Cesium Vapor 7) Power Supplies (Power Equipment) I Thermionic Converter Research II Martini, W. R. III Clark, E. V. IV U. S. Army Electronics Research and Development Laboratory, Ft. Monmouth, New Jersey V Contract No. DA 36-039 SC-88982 UNCLASSIFIED
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AD Atomics International, Canoga Park, Calif. FLAME HEATED THERMIONIC CONVERTER RESEARCH by W. R. Martini, E. V. Clark 5th Quarterly Report, 1 July - 30 September 1962 37 pp, 6 illus., 10 refs. (Report No. AI-7841) (Contract DA 36-039 SC-88982) Unclassified Report The purpose of this research is to develop the technology required for portable, flame-heated thermionic power sources. Active development in thermionic diodes, heat sources and materials is underway. The flame-heated diodes being made for this project are undergoing constant improvement in order to eliminate construction difficulties and operating limitations. Ten diodes have so far been constructed on the project. (over)	UNCLASSIFIED 1) Combustion Chambers 2) Combustion Chamber Liners 3) Molybdenum Compounds and Silicides 4) Silicon Compounds and Carbides 5) Inverter Rectifiers 6) Diodes (Electronic Tube Devices) - Cesium Vapor 7) Power Supplies (Power Equipment) I Thermionic Converter Research II Martini, W. R. III Clark, E. V. IV U. S. Army Electronics Research and Development Laboratory, Ft. Monmouth, New Jersey V Contract No. DA 36-039 SC-88982 UNCLASSIFIED
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AD The vacuum insulated heating experiment was activated to test emitter thimble assemblies in an environment very close to that of an actual diode. The effect of ignition delay and longitudinal heat conduction on burner operation was considered. Materials in addition to Durak-B coated molybdenum are being procured for evaluation. Additional measurements of gas permeation through Durak-B coated molybdenum agree with the previous measurement of 1 std. cc/hr cm² at 1400°C. But under certain conditions lower leak rates are observed. Five flame-heated converters were tested during this report period. Some power was generated. Failure of the Durak-B coating is a continuing problem.	UNCLASSIFIED UNITERMS Thermionics Converters Diodes Molybdenum Molybdenum Disilicide Protective Coatings Combustion Durak-B Propane Gasoline Silicon Carbide UNCLASSIFIED
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