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THERMIONIC CONVERTERS
FOR
COMPACT NUCLEAR POWER PLANTS

AEC Research and Development Report

ATOMICS INTERNATIONAL

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FOR
COMPACT NUCLEAR POWER PLANTS

By

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ATOMICS INTERNATIONAL

A DIVISION OF NORTH AMERICAN AVIATION, INC.
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ABSTRACT

A brief review of several thermionic nuclear power plants leads to the conclusion that the in-core concept is the most practical and useful system for space applications. Fundamental considerations indicate that emitter temperatures from 1500 to 1900°C are required for this concept, and that the high work function refractory metals are the best electrode materials for achieving the required performance and lifetime at these temperatures. The analytical and experimental technology developed for systematically defining the optimum materials and operating conditions is described, along with the significance of the experimental results obtained. These results have shown that the required performance can be obtained with the elementary cesium vapor diode converter. It has also been shown that impurities cause pronounced effects on both performance and lifetime, and that their suppression or controlled utilization is an important aspect of the present approach.

I. INTRODUCTION

The thermionic converter is a device for the conversion of heat to electrical power by thermionic emission.¹ As part of a general program in thermionic energy conversion, Atomics International is investigating experimental thermionic converters in the region of performance appropriate for their use in compact nuclear power systems. This work is supported by the U.S. Atomic Energy Commission,^{*} to define and project basic converter characteristics and capabilities for a realistic analysis of advanced systems incorporating these devices.

The converter studies are complemented by investigations of basic processes in thermionic conversion for the Office of Naval Research,[†] and by advanced reactor system analyses. This report deals with the significance of the results obtained to date in the converter studies, and is an interim review of the status of this work. Results of the basic processes² and system^{3,4} studies are reported elsewhere.

The report is developed in four principal parts. In the first, the converter performance requirements imposed by compact thermionic reactor systems are summarized, to define the most appropriate region of emphasis in the converter studies. In the second, the basic physics of the conversion process is used to define the general approach which has characterized converter studies at AI over the past 2-1/2 years. The third part describes the experimental techniques and devices developed in the experimental investigations. Finally, in the fourth part, the experimental results are reviewed, to illustrate the extent of progress toward the program objectives and to define the direction which future converter studies should take.

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II. CONVERTER PERFORMANCE REQUIRED FOR REACTOR APPLICATION

A. GENERAL PURPOSE

The essence of an applied research program is the systematic development of a central theme, leading to efficient achievement of a practical and useful objective. It is of prime importance that this practical objective be well defined, and that it will be useful when it is reached. In this section, the potential usefulness and practicality of thermionic reactor systems are briefly reviewed, and the objectives of the present research program are defined as those which can be expected to meet the requirements of the most practical and useful system. The central theme of the research leading to those objectives is described in Sections III and IV.

B. INCENTIVES FOR DEVELOPMENT OF THERMIONIC REACTOR SYSTEMS

The major potential advantages of thermionic reactor systems are:

- 1) Silent and static operation.
- 2) Compactness, resulting in lower specific weight for the system.
- 3) High temperature operation, for efficient heat rejection, or for increasing the output of conventional systems (i. e., for "topping").⁵

Most attention has been directed to the use of such a power plant in space-power applications, where system weight and heat rejection considerations are often crucial to the feasibility of a system. Interest has also been shown in its possible use in submarine or military applications, where silence is an important consideration. However, it should be recognized that the novelty of the thermionic approach does not in itself ensure that it will be superior to more conventional systems which have been modified to incorporate the above advantages. Thus, to be practical and useful, a thermionic system must possess sufficiently outstanding potential advantages over competing conventional systems to merit undertaking advances substantially beyond existing technology. It is especially important that the growth potential of the thermionic system concept selected for development be sufficiently great that its superiority is easily maintained during concurrent evolution of other systems. Several thermionic reactor system concepts are reviewed in the following sections, with emphasis on these criteria.

C. SYNOPSIS AND COMPARISON OF SYSTEM CONCEPTS

There are a number of nuclear thermionic system concepts which can be considered, each of which possesses potential advantages relative to the others. In general, these concepts can be categorized as follows:

- 1) Out-of-core converters, conduction heat source;
- 2) Out-of-core converters, convection heat source;
- 3) In-core converters, conduction heat source.

These systems are illustrated schematically in Figure 1.

In the discussion which follows, these concepts are examined and compared on the basis of their potential for space applications. However, adaptations for heat rejection to terrestrial environments would not materially alter the relative merits of each concept.

1. Out-of-Core Converters, Conduction Heat Source

A schematic of this concept is shown in Figure 1a. Heat is generated within the reactor core, and conducted to thermionic converters located either between the core and the radial neutron reflector or outside the reflector. Part of the heat arriving at the converter is converted to electricity, and the remainder is conducted to the outer surface, from which it is radiated to space. It is possible to mount the converters on the outer surface of the radial reflector if the temperature drop across the radial reflector and the area available for heat rejection lead to usable emitter and collector temperatures.

By electrically segmenting the core and reflector, or by electrically isolating the converters from other conducting components, a number of cells can be operated in electrical series to provide voltages high enough for static inversion and currents low enough for minimal I^2R losses. Such systems are described in the literature (e. g., Reference 6).

The exclusive use of conduction heat transfer within this system gives it a unique advantage of completely static operation; but, at the same time, gives rise to severe limitations on its practicality and usefulness. In most cases, the emitter area and the surface area available for heat rejection by radiation are incompatible with the emitter and collector temperatures required for efficient performance. Because the very limited area available for heat

a. Out-of-Core Conduction-Cooled Concept

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b. Convection Out-of-Core Concept

7590-A0325

c. Convection In-Core Concept

7590-A0324

Figure 1. Schematics of Thermionic Concepts

rejection requires a high rejection temperature, very high fuel temperatures are required in this concept. Even if fuels technology is extended to about 2400°K (current technology is less than 1500°K), a compact system design is limited to about 20 kwe or less output at an unshielded system weight of about 65 lb/kwe (See "prototype" design in Reference 6). If a less compact system could be tolerated, a more dilute nuclear fuel could be used and additional surface area could be made available. However, for reactor shadow shielding reasons, it is highly desirable to increase reactor length, rather than diameter; in which case, the system weight would increase almost linearly with power output, to again yield an unshielded specific weight of about 65 lb/kwe. For comparison, the unshielded specific weight of the 30-kwe SNAP 8 system, now being developed, is about 57 lb/kwe, including startup and safety equipment.

2. Out-of-Core Converters, Convection Heat Source

Figure 1b is a schematic of this concept. The conventional process of fluid convection is used to transfer the heat generated within the reactor to the emitters of independent converter assemblies, mounted on heat exchange tubes. Part of the heat is converted to electricity, and the remainder is rejected by thermal radiation from the outer surfaces of the converter collectors.

If it were possible to construct such a system with only a fuel temperature limitation, assuming as before that fuels technology is extended to 2400°K within the next half decade, the results would appear attractive; because separability of the reactor and converter would yield a high degree of flexibility. However, severe limitations on this system arise, not from the fuel temperature capability, but rather from the necessity for the entire heat transfer system (including the coolant and the complete reactor) to operate at temperatures at least equal to those that are needed to operate the converters at even small outputs and efficiencies (> 1400°K). The present state of the art for long-term operation of liquid metal convection loops limits maximum permissible liquid temperatures up to about 1450 to 1475°K. One study, which assumes operation at the latter temperatures, reports⁷ unshielded system specific weights ranging from about 10 lb/kwe at 300 kwe to about 8 lb/kwe at 1000 kwe. However, since output at these power levels must be at fairly high voltage, to provide reasonable current and to yield reasonable I^2R loss, the few mils of electrical insulation assumed in the reference study to prevent the emitters from shorting to the coolant was inadequate (the dielectric strength of BeO at 1450°K is estimated to

be less than 50 v/mil). Since all emitter heat must pass through this insulation, even a small increase in insulation thickness will reduce the permissible emitter temperature, and will lead to a decrease in performance. Therefore, it appears that the unshielded specific weight of a system, operating at these temperatures and at an output of 300 to 1000 kwe, will more probably be at least 12 to 15 lb/kwe.

In view of the past rate of progress in the development of high-temperature liquid metals technology, even with a highly concentrated developmental program it is doubtful that liquid temperatures beyond about 1650°K will be permissible for long-term operation within the next half decade. Even if such a temperature were permissible today, however, it is doubtful that a system employing the thermionic radiator concept and weighing less than about 10 lb/kwe (unshielded) could be constructed, due to the same problem that plagues the conduction out-of-core concept (i. e., it is very difficult to provide heat rejection area compatible with required emitter area and with the emitter and collector temperatures needed for performance approaching optimum efficiency and power density). Indeed, in the thermionic radiator concept, the emitter and radiator areas are approximately equal, unless emitter area is sacrificed. This incompatibility can be obviated if separate reactor, converter, and radiator were used; however, such a system would be unattractive, because it would have two completely separate coolant loops and would be much heavier than the single loop concept.

There is a secondary, but quite important, disadvantage to this concept: a compact electromagnetic pump is not practical for circulation of the reactor coolant at temperatures above the 1400°K Curie point of useful magnet materials. In addition, even if the complexity and weight of a secondary coolant loop (to cool the magnets) could be tolerated, an electromagnetic pump would be relatively inefficient above 1400°K, because the electrical resistivity of a liquid metal increases substantially with temperature. Therefore, it is necessary that a mechanical pump be used in this concept, and the advantages inherent in operation with no continuous mechanical motion in the system are not realized.

3. In-Core Converters, Conduction Heat Source

The in-core thermionic conversion reactor system is shown in Figure 1c. In this concept, the nuclear fuel is contained within the emitter structure of the

converter cells, such that an assembly of cells in electrical series constitutes each fuel element of the reactor core. The collectors of the cells are cooled by convection, in the same way as are more conventional fuel elements. Except for the interior details of the fuel elements themselves, components of this system are essentially identical in size, configuration, and operating temperature with those of compact reactor systems now in an advanced state of hardware development (e. g., SNAP 8). In addition, this in-core system concept differs from all others, both direct and turboelectric conversion types, in that a single coolant loop removes heat from the reactor at the heat rejection temperature (about 1100°K or less), and delivers it directly to the radiator. This obviates the emitter temperature limitation imposed on the out-of-core concepts by electrical insulation and liquid metals technologies. All electrical insulation (except that between adjacent emitters, which insulates against a potential of only 1 v) operates at 1100°K or less. In addition, the separability of converter and radiator removes the basic incompatibility of emitter and heat rejection areas.

It is anticipated that fuel temperatures up to about 2400°K might become permissible within the next half decade. In addition, reasonable extrapolation of present converter performance and rate of improvement, over that same period of time, indicates that conversion efficiencies and power densities in the vicinity of 20% and 20 w/cm² will be possible, at emitter temperatures of 2100°K or less and interelectrode spacings on the order of 20 mils. Design studies³ of this concept, assuming 20% efficiency and 17 w/cm² power density, yield system specific weights of 4 to 5 lb/kwe (unshielded) for power outputs in the range 300 to 1500 kwe. Even when the current state of converter development is applied to the above system (10% efficiency and 7 w/cm² power density), these systems are estimated to weigh less than 10 lb/kwe (unshielded).

Because many of the components and much of the technology for this concept are now being developed under other programs, the primary unknowns related to this concept are the behavior of fuels and the compatibility of fuels and other materials at temperatures beyond the current state of the art. Work in these areas is proceeding under AEC sponsorship,* and it is expected that useful information will start becoming available within the next year.

*Battelle Memorial Institute

D. SUMMARY OF REQUIREMENTS

The foregoing discussion indicates that all thermionic reactor systems characteristically require fuel temperatures (1400°C) which are substantially beyond existing practice for long-lifetime reactor systems. However, it appears that the in-core concept has distinct, or even decisive, practical advantage over the others in the technology of the remaining components of the system. Of greatest significance is that only the in-core system has the potential superiority in performance over turboelectric systems to make its development appear worthwhile. The conclusion drawn from these considerations is that the practicality and usefulness of a compact thermionic reactor system are crucially dependent on obtaining converter performance meeting the requirements of the in-core system. Conversely stated, it is necessary to anticipate converter performance of at least 10 w/cm² and 12% efficiency at about 1 v or more per cell for periods approaching a year to justify the development of a thermionic reactor system for space applications.

It should be recognized that this high performance need not be attained in early systems, which are developed primarily to demonstrate feasibility and which may find use in less demanding terrestrial applications. However, it appears most logical that the system selected for development should be one which is not inherently limited in performance and growth potential (e. g., a system intrinsically capable of high-temperature and high-performance operation can be useful at lower temperatures for development and terrestrial applications). On the other hand, a system limited to low temperatures would not be useful in space applications or possess the growth potential to remain competitive with the expected evolution of conventional systems or systems employing other new methods of energy conversion. Thermionic converters with high performance potential have therefore been emphasized here, with the low-temperature, low-performance phase of their development considered as an obvious special case.

III. THEORETICAL BASIS FOR EXPERIMENTAL APPROACH

A. ELECTRON EMISSION LIMITS ON CONVERTER PERFORMANCE

It is now necessary to define under what conditions the in-core system requirements fall within the assessable theoretical bounds on converter performance. Since a detailed description of the associated theory has been reported,^{9,10} only the assumptions and results pertinent to the present report are abstracted here.

The primary thermionic energy conversion process occurs, as illustrated in Figure 2, by the emission of electrons from a hot electrode (the emitter, or cathode), and their subsequent collection by a colder electrode (the collector, or anode). Thus, the fundamental limitations on this essential electron emission process impose an upper limit on the performance of the converter. In practical devices, processes also occur, during transport of the emitter electrons across the interelectrode space, which reduce the output below this emission-imposed limit.

It is instructive and useful to compute the performance of an idealized thermionic converter (i. e., one limited by the electron emission process, but having no transport effects to inhibit the interelectrode flow of electrons). This computation defines the upper performance limit on all types of thermionic converters, and illustrates the basic interrelation among the variables which is necessary for optimum performance. It is readily shown^{9,11} that the maximum obtainable efficiency, $\eta_{\max}$, of the idealized thermionic converter is given by:

$$\eta_{\max} = \beta \left[1 - \gamma \frac{\phi_c}{V_e} \right] , \quad \dots (1)$$

where ϕ_c is the work function of the collector, and V_e is the optimum height of the potential energy barrier which electrons must surmount to be thermally excited from the emitter to the collector (See Figure 2a). The parameters, β and γ , are so insensitive to all variables that they can be considered as constants for present purposes. Qualitatively, they include the unavoidable effects of irreversible heat conduction by the electron gas and losses in the lead wires. Typical values in the region of operation important to reactor application are $\beta \approx 0.74$ and $\gamma \approx 1.06$. A useful rule of thumb¹² for comprehending the

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b. Optimum Parameters
(1.7 eV Collector
Work Function)

Figure 2. Ideal Thermionic Converter

significance of Equation 1 is that the optimum operating temperature, T_{opt} , of an electrode is approximately proportional to its work function (or barrier height), ϕ ; i. e.,

$$\frac{T_{\text{opt}}}{\phi} = \left[k \ln \frac{AT_{\text{opt}}^2}{J} \right]^{-1} \approx \begin{array}{l} 670^\circ\text{K/ev for the emitter} \\ 580^\circ\text{K/ev for the collector} \end{array}, \quad \dots (2)$$

where k is the Boltzman constant, and A is the Richardson constant. The logarithm is very insensitive to its argument, and thus is approximately constant for temperatures and emission current densities, J , of interest in the converter. Accordingly, Equation 1 may be re-expressed as

$$\eta_{\text{max}} = \beta \left[1 - \gamma' \frac{T_c}{T_e} \right], \quad \dots (2a)$$

where $\gamma' = \gamma \frac{T_e \phi_c}{T_c V_e} \approx 1.22$, and T_e and T_c are the optimum emitter and collector temperatures. Equation 2a may be seen to be a modified form of the Carnot law, and approaches the Carnot law as its limit when reversible operation is approached (i. e., β and γ' approach unity as T_e approaches T_c , to satisfy the second law of thermodynamics).

Equation 1 emphasizes the need for a low collector work function, ϕ_c , for efficient energy conversion. The lowest known true work functions are in the vicinity of 1 ev, but these relatively unstable surfaces have not yet been successfully used as a collector surface for long periods in the high temperature converter environment. Surfaces with adsorbed films in equilibrium with cesium vapor have been shown to be stable in this environment. However, fairly fundamental considerations¹³ imply that the values presently obtained (> 1.6 ev) are near the lowest obtainable by the use of cesium alone. Even if stable work functions lower than 1.6 ev become available, they will not be useful for a thermionic reactor system in space. The rule in Equation 2 shows that use of collector work functions less than 1.6 ev requires optimum collector (heat rejection) temperatures less than 650°C, which results in excessive radiator size and weight for high-power space systems.

Using these basic limitations on presently useful collector temperatures and work function ($T_c > 650^\circ\text{C}$ and $\phi_c > 1.6$ ev), it can be seen from Equation 2a that the only significant independent variable affecting the maximum obtainable efficiency, η_{max} , is the emitter temperature, T_e . In Figure 2b, the analysis leading to Equation 1 is used⁹ to show the dependence of η_{max} on emitter temperature and system thermal emissivity, ϵ , for a readily achievable value of ϕ_c as a reference case. Also shown are the values of output power density, P_o , output emf, V_o , and collector temperature, T_c , required to obtain this maximum converter efficiency.

It should be recognized that it is frequently profitable to operate at values different from P_o , V_o , and T_c when the converter is integrated into a power system. The penalty in converter efficiency thus incurred by departure from these values at a given emitter temperature is compensated by improved performance elsewhere in the system. In the in-core reactor system, for instance, it is advantageous to operate at a power density somewhat higher than P_o to decrease the core volume, and to operate at a collector temperature higher than T_c to reduce the radiator weight. The extent of this compromise naturally depends on the details of application. However, for present purposes, the optimum values for the in-core reactor system are not appreciably different than those giving maximum converter efficiency (i. e., those shown in Figure 2b).

B. TEMPERATURE LIMITS

As may be seen in Figure 2b, the performance required for a useful in-core system cannot be obtained by any type of thermionic converter with an emitter temperature lower than about 1500°C (for $\phi_c > 1.7$ ev). Since ideal performance will probably not be achieved in engineering practice, it is likely that somewhat higher temperatures than this lower limit will be required. However, fuel lifetime problems rapidly become more severe with higher temperatures, imposing an upper practical temperature limit. A useful empirical rule, related to the thermal activation of process-promoting crystal imperfections, is that a material usually becomes mechanically and chemically unstable at temperatures appreciably greater than about 70% of its melting temperature. Since there is no known material of acceptable uranium content which melts appreciably above 2800°C , emitter temperatures less than about 1900°C should be used if possible. Figure 2b indicates that adequate performance is

obtainable below 1900°C, so that the region of emitter temperature of greatest interest is approximately between 1500 and 1900°C. The usefulness and practicality of the system decrease rapidly outside these limits.

C. SUITABLE EMITTER MATERIALS

1. Electron Emission Requirement

From Figure 2, it may be seen that, from the electron emission standpoint alone, optimum operation of a converter is independent of the value of the emitter work function. It is only necessary that the emitter work function be less than

$$\phi_{\max} = V_e = eV_o + \phi_c \quad \dots (3)$$

to obtain the maximum efficiency and the optimum output emf and current density for the idealized converter. However, other considerations can introduce additional constraints on the values of work function acceptable, and will now be considered.

2. Ion Emission Requirement

The emitted electrons give rise to a negative space charge which can inhibit further electron emission unless steps are taken to suppress it. While this effect can be suppressed by using very small spacings (<0.0005 in.), a more practical approach is to supply enough positive ions to neutralize the negative space charge of the large electron currents required. Ion sources for the thermionic converter are conveniently divided into three types:

- a) Ion emission from the emitter surface in a diode.¹
- b) Ion generation in the gas between the diode electrodes.¹⁴
- c) Ion production by an auxiliary ion source.¹⁵

The emitter work functions which permit adequate emission of both ions and electrons (type a) are discussed in this section. The other sources of ions (b and c) are not discussed in detail in this report, since they have been shown to be unnecessary at the emitter temperatures required in a high performance thermionic reactor system.

Enough ions are emitted from the emitter to neutralize the emitted electrons in its vicinity when

$$\frac{J}{q} < \frac{v_e}{v_i} = \alpha \quad , \quad \dots (4)$$

where J is the electron emission current, q is the ion emission current, and v_e and v_i are the respective drift velocities of the electrons and ions. Since the particles are all emitted at the same temperature (i. e. , same average energy), $\alpha = \sqrt{M/m}$ for field-free space, where M and m are the ion and electron masses. If the ions and electrons suffer many collisions in their motion under the influence of an electric field, $\alpha = \mu_e/\mu_i$, where μ_e and μ_i are the respective mobilities of the electron and ion. While the inequality in Equation 4 does not ensure that neutralization will occur throughout the entire space between the electrodes, it is a valid criterion for ensuring that a negative space charge barrier will not exist in the vicinity of the emitter. The condition in Equation 4 is thus necessary, but not sufficient, for ion emission to allow saturation electron current to be drawn from the emitter.

When the emitter is immersed in an easily ionized gas, such as cesium vapor, a fraction of the atoms incident on it are re-emitted as ions. Since the efficiency of ionization increases with increasing emitter work function, obtaining enough ion current by this process to satisfy Equation 4 imposes new restrictions on the acceptable emitter work functions. In Figure 3, the minimum emitter work function, $\phi_{\min}$, which will give an ion current to satisfy Equation 4 is shown for various pressures of cesium vapor.⁹ Since cesium has the lowest known ionization potential, all other gases would require operation at higher temperatures and pressures. Included in Figure 3 is the maximum emitter work function ($\phi_{\max}$ from Equation 3 and Figure 2), which allows efficient operation for several system thermal emissivities, ϵ . Emitters giving acceptable electron and ion emission currents are those for which $\phi_{\min} < \phi_e < \phi_{\max}$ in Figure 3.

3. Acceptable Work Functions

It is necessary to distinguish between two different states of emitter operation in the presence of cesium vapor. In one state, the rate of arrival of

Figure 3. Acceptable Values of Emitter Work Function for Optimum Emitter Current

cesium atoms from the gas (i. e., its pressure) is not high enough to maintain a layer of cesium atoms adsorbed on the emitter surface. For convenience, an emitter will be described as being in the "dry" state of operation, under these conditions. At a sufficiently high pressure, a layer of adsorbed atoms can be maintained on the surface in equilibrium with the vapor, and this will be called the "wet" state of emitter operation. While the work function of a "dry" emitter in cesium vapor is that of its bare surface, the presence of the adsorbed cesium layer causes the work function of the "wet" emitter to be lower than its bare (dry) value. The number of cesium atoms which can be maintained on the surface, and therefore the extent to which its work function is lowered, depends on the temperature and cesium atom affinity of the surface. This affinity has been shown¹³ to be directly related to the bare work function of the surface. Accordingly, Figure 4 shows how the "wet" work function of a surface depends on its temperature, its bare work function, and the pressure of the cesium in which it is immersed. This dependence has been confirmed experimentally. The cesium pressure is expressed in terms of T_R , the temperature of the liquid cesium reservoir supplying the vapor.

Figure 4. Computed Work Function of Surfaces with Adsorbed Cesium (See Reference 13 for Conditions of Validity)

It is now possible to use Figures 3 and 4 to define under what conditions a material of bare work function ϕ_{eo} is suitable for use as the emitter in a thermionic converter. In Figure 5, the regions of adequate and inadequate ion and electron emission are shown¹⁶ for several emitter temperatures, and a region of excessive cesium pressure requirement is indicated. The boundaries, particularly of the latter region, are not uniquely defined, due to their dependence on other system parameters, such as electrode spacing and thermal emissivity. However, the boundaries shown are typical, and are sufficiently insensitive to other parameters for use in general evaluation of emitter materials. Figure 6 summarizes Figure 5 by eliminating pressure as a variable.

Inspection of Figures 5 and 6 shows that materials having bare work functions roughly between 3 and 4 ev do not emit enough electrons for efficient operation, except at excessively high temperatures or high cesium pressures. Materials having lower bare work functions give acceptable electron emission, when operating as "dry" emitters. Materials with higher work functions operate acceptably in the "wet" state without requiring excessively high cesium pressures. As also may be seen, in Figures 5 and 6, dry emitters with work functions less than about 2.6 ev, and wet emitters with bare work functions less than about 4.2 ev, require excessive cesium pressures to emit enough ions to neutralize electron emission. Higher bare work functions reduce the pressure required, but increase the temperatures required for adequate ion and electron emission.

In summary, the acceptability of materials as converter emitters (at practical temperatures and pressures) can be judged according to their bare work functions by recognition of these different but overlapping groups:

- a) Materials giving both adequate ion and electron emission (i. e., potentially best suited for diode operation), having $2.6 < \phi_{eo} < 3.2$ ev or $\phi_{eo} > 4.2$ ev.
- b) Materials giving adequate electron emission only (i. e., requiring other sources of ions, as in triode operation), having $4.2 > \phi_{eo} > 4.0$ ev or $\phi_{eo} < 2.6$ ev.
- c) Materials giving neither adequate ion emission nor adequate electron emission, except at unattractively high temperatures or pressures (i. e., unacceptable for any type of converter), having $3.2 < \phi_{eo} < 4.0$ ev.

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Figure 5. Emission Limitations on Converter Operation

Figure 6. Summary of Figure 5, Showing Parameters of Electron and Ion Emission

Figure 7. Material Vaporization of Dry Emitter Surfaces and in 5 Torr of Cesium Vapor

4. Materials Having Acceptable Work Functions

A number of materials are listed opposite their bare work functions in Figures 5 and 6. While this list is by no means complete, it is representative of materials whose technology is reasonably well known, and which have been seriously considered as practical emitters¹⁷ at these temperatures. It can be seen that most elemental refractory metals (such as Ta, Mo, W, Re, and Ir) and some refractory compounds (such as ThO_2 , LaB_6 , and Gd_2O_3) have work functions which permit these materials to supply both ions and electrons as a diode emitter at practical temperatures and pressures. In addition, the refractory metals are also acceptable emitters at lower temperatures and pressures when an additional source of ions is available (i. e., an auxiliary source or volume ionization). Low work function emitters, such as the Phillips Cathode (Ba-W), are only suitable when such a source is available. The materials UC, thoriated tungsten (Th-W), and Nb operate satisfactorily only at the extremes of practical temperature and pressure, and therefore are typical of materials which are only marginally useful for present purposes. Finally, materials such as Th, Zr, ZrC, and Hf are distinctly unsuitable.

5. Emitter Materials Capable of Acceptable Lifetime

Even if attractively high output performance is obtained, the usefulness of a thermionic reactor system diminishes rapidly if the lifetime of the converter is significantly less than a year, due partly to the expense of the system, but mainly to the logistics of its use. Therefore, in addition to the acceptability of the work function of a material, its ability to operate for an acceptably long time as an efficient converter emitter must be considered. For materials operating in the "dry" state, there is a fundamental correlation between the vaporization and emission processes, such that a high rate of vaporization tends to accompany large electron emission currents.¹³ As a result, there is no known material which can emit currents greater than 10 amp/cm^2 as a dry emitter without the occurrence of significant loss of material from the emitter by vaporization. This fact is illustrated, in Figure 7, for some of the better emitters. As may be seen in Figure 7, known dry emitters (such as UC and UC-ZrC) are at best only marginally acceptable for lifetimes approaching a year. This correlation between emission and vaporization is a fundamental obstacle in the search for more satisfactory dry emitters.

In wet emitters, this obstacle is circumvented by renewing the surface (film) from a reservoir, as fast as it evaporates, and by greatly decreasing the temperature required for adequate electron emission. The great superiority in the lifetime of the wet emitter over that of the dry is apparent in Figure 7. In the type of wet emitter considered thus far (refractory metal "refluxed" with cesium), the adsorbed film is in equilibrium with the vapor in which it is immersed, and thus is not depleted with time. In other types of wet emitters [e. g., the matrix (Phillips) types, and thoriated tungsten], the film is maintained by the migration of atoms from a reservoir within the electrode. The lifetime of this "dispenser" type emitter is terminated when this reservoir of dispensed material (e. g., Th, Ba, La, etc.) becomes depleted. This consideration limits the use of most conventional dispenser emitters to temperatures which are too low for present purposes.

From a mechanical standpoint, the amount of vaporization which can be tolerated depends on the manner in which the vaporization products are deposited on the collector electrode. If the products are deposited as a dense, adherent, metallic layer, a reasonably large amount of vaporization can be tolerated, since the spacing is unchanged. However, if the deposits flake or grow as whiskers, as little as one thousandth of an inch vaporization could progressively degrade performance, by causing leakage currents between the electrodes.

On the other hand, only a very small amount of vaporization (a few atomic layers or less) can increase the work function and/or thermal emissivity of the collector, and thereby significantly degrade the converter performance. This is a particularly important consideration for the dispenser (e. g., Phillips) emitter and for the compounds (e. g., the carbides), since their vapor products probably adsorb cesium only at low collector temperatures. However, the vaporization products of most of the refractory metals adsorb cesium to give stable work functions in the vicinity of 1.7 ev, and have relatively low thermal emissivities.

Similar considerations apply to vapor products and impurities which evolve from other components and deposit on the emitter. Dry emitters are often "poisoned" by such products, or by preferential vaporization of one component of the compound, while wet emitters can be relatively free of these effects.

6. Conclusions Regarding Best Emitter Materials

The foregoing considerations strongly suggest that the high-work-function refractory metals (such as Ta, Mo, W, Re, and Ir) are the best known materials for emitters in thermionic converters, in that:

- a) The ion and electron emission requirements can be satisfied, over a wide range of attractive temperatures and pressures, by choosing appropriately among them.
- b) They have extremely great potential lifetime and stability, with continuous and perpetual renewal of the emitting surface by cesium vapor.
- c) Their vaporization products are compatible with a low collector work function and low thermal emissivity.

There are other less decisive considerations, in which the refractory metals appear equally attractive: strength, creep, ductility, ease of fabrication, availability, and a mature technology from extensive use in other thermionic devices. However, further comparison is not included here, since the foregoing is sufficient to illustrate the reasons for their emphasis.

IV. DEFINITION OF EXPERIMENTAL OBJECTIVES

A. SEPARATION AND LIMITATION OF VARIABLES

Many variables must be considered, in order to define a unique operating point in a thermionic converter:

- 1) Temperature, surface composition, electrical potential, spacing, number, and configuration of electrodes.
- 2) Composition and pressure of the gas filling the converter.
- 3) External constraints associated with the electrical load, and the heat source and sink.
- 4) Time dependence (hysteresis, oscillations, instabilities).

To ensure coherent progress toward definable objectives, several rules have been used to systematically separate and restrict the number of variables. These rules are not rigidly enforced, but are applied whenever insufficient information is available to allow an unambiguous choice in approach to a problem area. Experimental investigations have thereby emphasized:

- 1) Refractory metal electrodes ("wet").
- 2) Cesium vapor diode operation.
- 3) Configurations and materials of construction approximating those suitable for practical application.
- 4) Operation at or near conditions giving maximum efficiency of energy conversion.
- 5) Environmental testing to advance the technology only of a converter having performance characteristics previously determined to be acceptable.

These rules are based on the practical and theoretical considerations discussed in the foregoing sections, and they have remained largely unchanged throughout the program. They still appear to be valid guideposts, based on present knowledge, and are discussed (in the order listed) in the following paragraphs.

The general utility of the refractory metals as practical converter electrodes has already been discussed. Of equal importance to converter research is that their emission properties (electron, ion, atom, and thermal) are relatively well understood, allowing emission-controlled variables to be separated from the poorly understood transport variables. These materials have proved to be quite suitable for both basic and practical purposes.

The cesium vapor diode is of prime interest for reactor application, since it has a more elementary configuration than any converter employing a third electrode. In addition, fundamental considerations (now supported by direct experimental data) indicate that auxiliary components are not necessary to obtain adequate performance at the temperatures required for useful thermionic reactor systems. Adherence to the diode has also permitted elimination of the variables associated with operation of such auxiliary components.

For converter research to have direct application to practical systems, it is highly desirable that experimental devices employ a technology which is consistent with both research and applied interests, wherever possible. All converters constructed in these investigations have been of advanced ceramic and metal construction, having configurations and dimensions which are within a small-scale factor of those appropriate for a typical thermionic reactor converter cell. The data developed, including overall efficiency measurement, permit confident and realistic evaluation of the results, without the necessity of major corrections based on computed estimates. Similarly, the techniques and materials technology, developed in constructing such devices, establish the constraints which will ultimately be imposed on the converter by such practical considerations. The uncertainty associated with "breadboard" systems, such as electrically heated wires in glass envelopes, requires the eventual repetition of such data, in the presence of these constraints, before its usefulness can be realistically evaluated. Fortunately, the constraints introduced by realistic converter construction generally coincide with those which aid fundamental interpretation of the results, as will be discussed later.

A most effective rule for reducing the amount of data to be taken and interpreted has been emphasis on the fully optimized converter. In this approach, each converter is operated initially to determine the values of input and output variables which give maximum efficiency (or maximum power) at each emitter temperature (corresponding to Figure 2b). The experimental variables given

Figure 8. Computed Emission-Imposed Efficiency Limits

first attention for further exploration are those which deviate most from the expected optimum values. Such attention is initially restricted to values of the variable which are in the vicinity of the optimum value. Since the experimental devices approximate practical and useful converters, this procedure ensures that most of the data obtained is pertinent to the improvement and utilization of a practical and useful mode of operation.

Since no device, to date, has been found to have the previously defined acceptable performance and lifetime outside a reactor, it has not yet been deemed appropriate to conduct tests within a reactor. It is presently felt that such tests will be most useful when independent converter research has produced an acceptable converter, and when independent reactor testing has defined acceptable nuclear components (especially the nuclear fuel material).

B. SPECIFIC CONCEPT SELECTED FOR EXPERIMENTAL EVALUATION

1. Ideal Diode Performance

In Section III, the basic physics of the emission processes was used to define the general bounds on converter performance, and to select the class of materials having the greatest potential usefulness in the converter. A specific approach will not be described toward defining the combination of design parameters which leads to the most efficient converter for specified conditions. The exploration of this approach, and the experimental evaluation of the associated unknown quantities, constitute the central theme of the present converter research program.

While the diagrams in Figures 5 and 6 are useful for a general illustration of the regions of acceptable converter operation, more detailed exploration of the boundaries of these regions is necessary for specification and experimental evaluation of optimum design parameters. For this purpose, attention will now be confined to diode operation, with the ad hoc assumption (supported by experimental evidence in Section VI) that emission processes at the emitter are the limiting source of ions for space charge neutralization. Accordingly, it can be shown⁹ that the maximum efficiency of an idealized converter (i. e., in the absence of transport effects) is as given in Figure 8. This figure thus shows how space charge reduces the ideal efficiency, upon crossing the boundary between the light and dark shaded regions at the top of Figure 6, for each bare

work function, ϕ_{eO} , of the emitter surface. This analysis indicates that the threshold temperature for efficient energy conversion increases as the bare work function increases, which has been confirmed experimentally (See Section VI). Accordingly, materials with low bare work functions would be the unqualified choice for the emitter if transport effects were negligible. Since experimental evidence shows that transport effects can severely degrade performance, the factors which govern the degree of degradation must be considered.

2. Suppression of Transport Effects by Diode Optimization

A great deal of effort has been expended by a number of organizations to obtain an understanding of the processes occurring in the gas ("plasma") between the electrodes. It has been hoped that such understanding would permit the defining of the modes of operation wherein deleterious transport effects are suppressed and/or beneficial ones (such as ion production) enhanced. While the individual elementary processes in the cesium vapor converter are fairly well understood, their simultaneous interaction in an efficient diode converter is a complex problem in gaseous electronics, which is still the subject of considerable controversy. To date, there is no generally accepted description which gives even consistent qualitative agreement with experimental data.

In the absence of a satisfactory theoretical description of these problems, the most direct course is to avoid them, by eliminating the medium in which they take place. In the diode, this amounts to reducing the electrode spacing and/or reducing the cesium pressure. Problems associated with lifetime and ease of construction place a lower practical limit on the spacing, and any change in cesium pressure must be compatible with the ion and electron emission requirements. A semiquantitative illustration of these considerations can be obtained with a few simplifying assumptions.

Though often differing substantially in the reason why, most theoretical analyses indicate that performance-degrading transport effects become important when the electrode spacing becomes greater than 10 to 100 times the electron mean free path, λ . Unfortunately, there is considerable difficulty in unambiguous definition of λ in the converter. Nevertheless, for purposes of illustration, it will be assumed that there is a maximum allowable or optimum spacing, d , which is a constant multiple of the mean free path, λ_c , determined

classically from the cesium pressure. The corresponding limitations on electrode spacing for "wet" emitters can be evaluated by inspection of Figure 8. Each dashed curve indicates the optimum temperature and maximum ideal efficiency for a given cesium pressure, and thus also for a given classical mean free path, λ_c . Accordingly, it can be expected that the electrode spacing must be less than 10 to 100 times the indicated value of λ_c , to avoid significant degradation from ideal performance by transport effects. Thus, Figure 8 shows the relationship between emitter temperature, emitter bare work function, and maximum spacing for ideal efficiency.

From these considerations, converter performance similar to that shown in Figure 9 would be expected. Specifically, for a given spacing (as in Figure 9a), there should be an optimum bare work function giving maximum conversion efficiency at each emitter temperature. The envelope of these maxima should also be dependent on spacing, as shown in Figure 9b. As indicated in Figure 9, it is possible that this envelope decreases, for bare work functions above a particular spacing-dependent value, due to degradation from the large current densities required for ideal performance at high temperatures. Accordingly, an optimum material and optimum temperature would exist, for each spacing, to achieve the maximum efficiency obtainable with a wet emitter. Data allowing the optimization process, shown in Figure 9 to be quantitatively performed, are not yet available. Although work is in progress, here and elsewhere, to derive the associated unknowns from basic theory, the complexity of this problem reduces the prospects of a purely analytical approach in the foreseeable future. Therefore, a major objective of the present converter program has been to obtain direct experimental data, of the form shown in Figure 9. Such data is of immediate importance in the design and development of practical systems, and also should provide valuable assistance to the concurrent analytical approach.

In the foregoing illustration, it was tacitly assumed that direct thermal emission of ions from the emitter is responsible for space charge neutralization. However, it has been shown⁹ that thermal emission of excited atoms from the emitter, with subsequent ionization by other processes, is an alternate emission-controlled source of ions. The qualitative conclusions in the foregoing illustration are the same, assuming either process, and data obtained to date do not permit unambiguous distinction between them. During development

a. Anticipated Converter Performance for One Spacing (d_2)

b. Anticipated Spacing Dependence of Maximum Efficiency Envelope

Figure 9. Efficiency vs Emitter Temperature

of this general approach, comparison of the experimental results with quantitative theoretical predictions, based on emission-controlled processes, should indicate which of these processes predominates. Similarly, such comparisons should indicate whether emission-controlled processes or others (e. g., volume excitation and ionization) are dominant.

Details of the experimental work undertaken in this approach, and the results thus far, are discussed in Sections V and VI. Additional converter investigations, undertaken to evaluate configurational perturbations potentially affecting optimum performance, are also discussed.

V. EXPERIMENTAL APPARATUS AND TECHNIQUES

A. INTRODUCTION

Approximately 30 individual thermionic converters, of various sizes and configurations (some with many modifications), have been constructed and tested in this work thus far. These converters are divided into several characteristic types, for general description of purpose and function. Particular models, which typify the most useful form of a particular type, are selected for detailed description in this report. The significance of the results obtained from these devices is described in Section VI.

The fabrication and the testing equipment and procedures are similarly diverse. A brief history of their development, and a description of typical current practice, is used to summarize this work.

B. TEST EQUIPMENT AND TECHNIQUES

1. Heat Source

Converter emitters are indirectly heated by electron bombardment from filaments completely enclosed in the emitter. Therefore, there is no significant electrical interaction between the power supply and the converter output. In addition, nearly all the electrical power supplied to the heater is delivered as heat to the emitter, allowing convenient and unambiguous calorimetry to be performed. In the cylindrical converter, this arrangement is a close thermal approximation to a fission-heated converter cell.

A totally enclosed electron bombardment heater tends to be thermally unstable (i. e., a perturbation in the temperature of the heated cavity changes the filament temperature by back radiation, and the resulting change in bombardment current is such as to amplify the original perturbation). This difficulty is readily overcome by employing the simple circuit shown in Figure 10 to automatically control the bombardment current by adjusting the filament current. For most purposes, the controller is set to give constant bombardment current for a given constant bombardment potential, resulting in constant power input. Alternately, the bombardment current is controlled to give constant emitter temperature, as sensed by an electronic photopyrometer.

Figure 10. Schematic of Electron Bombardment Heater and Controller

The bombardment filament is ordinarily a loop or coil of 0.010-in. tungsten wire, spot welded to 0.030-in. tantalum leads just outside the region to be heated, as shown in Figure 10. Tungsten filaments are satisfactory for emitter temperatures up to about 2200°C; but, at higher temperatures, the controller ceases to function, since back radiation from the emitter is sufficient to heat the filament to emission temperatures, and the required filament current vanishes. For this reason, filaments with lower work function (e. g., tantalum or thoriated tungsten) are satisfactory only at lower temperatures, and higher work function filaments (e. g., rhenium) are appropriate for higher temperatures.

If the length of the cavity and filament is large, compared with the cavity diameter, the thermal instability mentioned previously can occur locally, leading to an amplification of any temperature gradient along the cavity. Such effects have been detected, and are now suppressed by ensuring that the cavity walls are thick enough to remain essentially isothermal in the presence of unequal heating. Furthermore, the position and shape of the filament is adjusted to give an observed isothermal temperature profile. Additional heat generation

is particularly needed, at the junction between the emitter and its supporting lead, to supply the heat that is conducted away by the lead.

2. Temperature Measurement

The emitter temperature is measured by focusing an optical pyrometer or electronic photopyrometer* on the bottom of 0.032-in. diameter by 0.090-in. deep hole in the bottom of the heater cavity. Two corrections are made to the observed readings: one for absorption by the vacuum chamber window, and one for radiation from the bombardment filament reflected from the bottom of the sight hole. The latter correction is negligible above 1500°C (<10°C), is small at 1400°C (~30°C), and becomes significant below 1300°C (>100°C). The correction is evaluated in situ by first measuring the apparent cavity temperature with the filament glowing and then plotting a temperature decay curve after extinguishing the filament. The difference between the apparent temperature during operating conditions and the zero time intercept of the temperature decay curve is the error caused from the filament radiation entering the cavity. The validity of these corrections has been verified by simultaneous observation of the interior and exterior of an isolated emitter.

All other temperatures (e. g., that of the collector, the cesium reservoir, the seal, etc.) are measured, using Chromel-Alumel thermocouples individually spark welded to the test point. These temperatures are recorded on a multiple-point strip-chart recorder. The methods by which these temperatures are controlled are included in the description of the individual converter types.

3. Converter Output Control and Measurement (Static)

The circuit used to control and measure the electrical output of the converters during static operation is shown schematically in Figure 11. For operation at positive output power, a smoothly variable, high-current resistor (carbon pile rheostat) is used as the load. For reverse current or reverse potential measurements, an externally supplied current, controlled by a similar rheostat, is shunted across the load. Output potential is measured between probes that are spark welded directly to the collector and to the integral emitter lead, where it emerges from the converter. Output current is determined from

*Thermodot

Figure 11. Output Control and Measurement Circuits for Static Operation and Optimization

the potential drop across a 50 mv shunt* having seven ranges between 1 and 100 amp. Output current and potential are recorded continuously and simultaneously on a two-pen, strip-chart recording potentiometer.†

By means of the simple circuit shown in Figure 11, one channel of the recorder can be used to compute the electrical power output of the converter, allowing the output power to be continuously recorded as well. This elementary provision is especially useful in experimentally establishing the optimum operating conditions discussed in the previous sections. When the bombardment current controller is set to give constant input power, each of the other experimentally adjustable independent variables (i. e., load resistance, cesium pressure, collector temperature) can be successively changed iteratively until each variable has the value giving maximum indicated power output. The operating point reached by this optimization process is that giving maximum efficiency at the resulting emitter temperature. The maximum overall efficiency at this temperature is computed by dividing the optimized output power by the

*Weston Precision Multiple Range Shunt Type 8

†Output is also monitored continuously for oscillations, using an oscilloscope

a. Low Resistance Load (For Characteristics in Typical Power System)

b. High Resistance Load (For Obtaining True Converter Characteristics at Low Currents)

c. Controlled Current Load (For Infinite Dynamic Resistance Ideal for Obtaining True Characteristics at all Currents)

Figure 12. Circuits for Quasistatic Output Measurements

total input power. This process, repeated for a series of different input powers, constitutes direct experimental determination of the maximum efficiency and the associated optimum values of the other variables, as a function of emitter temperature (corresponding to Figure 2b).

Alternatively, the converter may be optimized for maximum output power by setting the bombardment current controller to maintain a constant emitter temperature, and then repeating the adjustment procedure described previously. Although the optimum operating point for maximum efficiency is generally quite different from that for maximum output power at a given temperature, the two conditions are frequently confused in the description of experimental results. From the foregoing, it may be seen that the point of maximum efficiency for a given emitter temperature is determined by experimentally optimizing at constant power input, with emitter temperature as a dependent variable. On the other hand, the point of maximum output power for a given emitter temperature is determined by optimizing at constant emitter temperature, with input power as a dependent variable.

4. Converter Output Control and Measurement (Quasistatic)

While the described procedures are appropriate for determining the optimum operating points, exploration of the off-optimum characteristics for each variable can become extremely tedious and time-consuming, if data are taken point-by-point. A more satisfactory approach is to cause the independent variable under investigation to change continuously, over the range of interest, while the others remain constant. Changes in the dependent variables (typically the electrical output) are continuously recorded. If the independent variable is swept over the range of interesting values so quickly that the others do not change appreciably, there is no need for precise automatic control of these variables. Data taken by this "quasistatic" method are physically equivalent to static data, but are highly superior in resolution and ease of collection.

Figure 12 shows the arrangements used to quasistatically obtain the electrical output characteristics of converters. In Figure 12a, the potential across the converter is varied periodically by inserting the secondary of a transformer in series with the previously described electrical load, and driving the primary with an audio oscillator. This arrangement is appropriate for obtaining the isothermal volt-ampere characteristics for the high currents

and low load resistance typical of a practical system. However, in two distinct ways this arrangement can seriously distort the true characteristics of the converter itself, thus obscuring features of importance to fundamental interpretation of the conversion process. First, the inductance and saturation of the transformer can grossly distort the input waveform for a nonlinear element, such as the converter. As a result, part of the cycle can be swept over in a time which is short, compared with the relaxation time of the basic processes in the converter. The observed characteristics thereupon depart substantially from the static characteristics. While analysis of the transient response of the converter is a useful diagnostic tool, a method giving an input of prescribed form is more appropriate. The second difficulty with the low impedance circuit in Figure 12a is that the circuit can be unstable under conditions where the converter is not. Thus, if the dynamic resistance, $\partial E / \partial I$, of the converter has a negative value greater than the load resistance, the circuit has negative resistance and is unstable. The resulting discontinuous transitions and oscillations, which are not inherent to the converter itself, can prevent observation of the true converter characteristics in this region. This difficulty can be avoided, and the true characteristics observed, by employing the circuits in Figures 12b and 12c, in which the (positive) load resistance is very large. Although the power requirements for the arrangement in Figure 12b become excessive at large currents, the negative resistance regions, occurring only at low currents, are readily explored. Use of an electronically controlled current supply, as shown in Figure 12c, provides an infinite dynamic resistance load at all currents without excessive power requirements. Examples of data taken with the circuits in Figure 12 are presented in Section VI.

C. CONVERTER CONSTRUCTION AND PREPARATION

1. Component Technology

While the fabrication and assembly technology of most converter components is best discussed with the particular converter type, the technology of the ceramic-to-metal seal (or insulator) and the cesium charge is common to all types, and is of sufficient importance to deserve separate description. In all converters constructed here, the envelope has been composed entirely of ceramic and metal. This choice was dictated by the desire to approximate conditions in practical devices, as previously discussed, and by the requirement

that the cesium pressure should not be limited by the maximum permissible temperature of the envelope. When the first converters were constructed here (early 1959), the availability and variety of commercially manufactured ceramic-to-metal seals was relatively poor, by present standards. For this reason, the ceramic and metal parts comprising the vacuum envelope of the converter were brazed directly together in a laboratory induction furnace. Some successful seals were obtained, using the active metal hydride technique,¹⁸ but more consistent results with less preparation were obtained, using a commercially obtainable titanium-cored brazing wire.* Fortunately, at about that time, several commercial organizations† began marketing ceramic-metal seals, in a wide variety of shapes and sizes, which had a reliability corresponding to that of seals formally found only in mass-produced commercial electron tubes. In addition, minor modifications of standard seals are obtained with relatively short delivery times. All subsequent converters therefore have utilized such commercial seals.

A number of successful converters were constructed, in which the flanges of the commercial seals were brazed to the converter by heating the entire assembly in a vacuum induction furnace. The incidence of faulty seals, as a result of this procedure, was high; since it imposed severe thermal stresses on the seal at temperatures near the melting point of its bonding metal. A technique has since been developed, in which the seal flanges are welded or brazed into place without significantly heating or stressing the ceramic-metal bond. The parts are precisely aligned by jigs in which the metal adjacent to the ceramic-metal bond is in good thermal contact with "chill-bars" which are maintained near room temperature. The protruding joint between the seal flange and the converter assembly is then arc welded or arc brazed, as the assembly is rotated by a machine-driven chuck in an inert gas retort. Precise control of arc power and feed, vacuum purging of the retort, and the use of purified gas§ results in a highly uniform and clean joint. A high degree of seal reliability and reproducibility has been achieved by this technique. The variety of materials and configurations successfully employed can be inferred in the description of individual converters.

*Handy-Harmon BT Solder

†U. S. Stoneware, Ceramaseal, Coors

§Argon bubbled through Na-K eutectic.

The cesium metal available for filling the early converters was specified, by its commercial producer, as being only greater than 99% pure. Alkali metals, particularly rubidium, were the principal impurities. Through systematic inquiry among various chemical companies, and by placing orders for the purest product they estimated could be produced, the quality of the cesium used has steadily improved to the present specified purity of greater than 99.99%.* Work to further improve and verify the quality of the cesium is being performed here, in another program.²

As the quality of the cesium charge improved, the steps necessary to prevent its contamination during insertion and operation in the converter envelope deserved more attention. Early converters were filled with cesium by techniques similar to those used in its production and packaging by the commercial supplier. The required amount of liquid cesium metal was removed from a relatively large Pyrex glass shipping ampule, and injected into the converter by means of a hypodermic needle. A blanket of purified argon was used to prevent reaction with air during this operation. While there is no evidence that this method introduces harmful impurities into the system when carefully performed, more sophisticated procedures are now used to ensure more precise control of the cesium history, and to permit more thorough final outgassing of the converter. Several converters were constructed, in which a glass ampule containing cesium was inserted into the converter under high vacuum after final outgassing, and was crushed after seal-off to release the cesium. While this procedure removed the necessity for the argon blanket, the crushed glass ampule thus remained in the envelope and introduced the uncertainty of possible reaction with the cesium at high cesium temperatures. A procedure has been developed which is now in routine use, and which eliminates the uncertainties introduced by the previous methods. The cesium is delivered by the manufacturer, as specified, in the small inexpensive container shown in Figure 13, which is known here as a "breakoffski." The converter and container are sealed onto a high vacuum system, where they are outgassed by prolonged heating. After outgassing, the tip of the breakoffski is broken by the attached magnetic armature, and the required amount of cesium is measured by distillation into an appendix, sized for that purpose. This amount is then

*Dow Chemical Co. and Kawecki Chemical Co.

Figure 13. Shipping Ampule for High Purity Cesium (Breakoffski)

distilled from the appendix into the converter. Final seal-off is accomplished either by pinch-off of a soft copper tube, or by melting a stainless steel tube by electron bombardment.

In addition to improved cesium purity and outgassing procedures, active metal getters have been incorporated as interior components in later converter models, to maintain a low concentration of reactive impurities in the cesium atmosphere.

2. Initial Models

While the construction techniques and data for the first type of converter constructed have largely been superseded by superior subsequent types, its description is appropriate, to illustrate the incentive for subsequent improvements. Figure 14 illustrates the first converters constructed. The most important phenomena were first observed, in June 1959, in these models: threshold emitter temperature, optimum cesium and collector temperatures, optimum load, ignited and extinguished modes, and oscillations. The highest overall optimized efficiency was observed to be about 5%. The working surfaces of the electrodes were parallel planes, about 0.030 in. apart. These surfaces were reentrant into a 0.010-in. thick molybdenum tube, which served as both the envelope and the emitter lead. Heat was supplied to a cavity in the molybdenum emitter by electron bombardment, and was removed from a cavity in the nickel collector by forced gas convection. The length of the electrodes and envelope were selected to give optimum electrical and heat transfer

Figure 14. Initial Converter Models Having Plane Electrodes
(One model is cut, to expose internal components)

properties at the design operating point. Several layers of dimpled molybdenum foil were interposed between the envelope and the electrodes as radiation shields. The molybdenum emitter was joined to the envelope by carbon eutectic welding in a vacuum induction furnace. The envelope was closed by brazing it and the collector flange to the alumina insulator. The collector flange also served as the cesium reservoir. Reservoir and collector temperature were individually controlled by regulating the flow of coolant gas.

In this design, as in all others constructed here, the temperature of the insulator was only slightly above that of the cesium reservoir. This allowed it to be operated near the lowest temperature of the system, for maximum reliability, without being short circuited by cesium condensation. Two models of this type were constructed and operated: one having a simple plane emitter, and another having a cavity ("heat-shielded") emitter, as discussed in Section VI.

Recently, a model of this type, similar in many respects to that in Figure 14, was constructed and operated here under another program.* The entire converter was operated in air, using a flame as the heat source.

*Signal Corps Contract DA-36-039SC 88982

3. Dismountable Cylindrical Converter

From the operation and analysis of the initial converters, it was concluded that realistic interpretation of the energy conversion efficiency of a converter with parallel plane electrodes would be difficult and ambiguous. Calorimetry was inaccurate, since stray heat losses predominated over heat transferred by essential converter processes. Furthermore, potentially large edge effects and possible stray currents to the envelope and radiation shields, inherent to this configuration, reduced confidence in the generality of the results obtained. For these reasons, it was decided that more rapid progress could be made toward the experimental objectives by using cylindrical (rather than plane) electrodes. In addition, more convenient access to the interior of the converter was also desired, to permit surveying a number of different electrode materials and gas compositions, and to permit inspection and adjustment of the electrodes between experimental tests. Production of a completely new model for each test was not possible, within the resources then available.

The type of converter constructed to meet these requirements is illustrated in Figure 15. The emitter consisted of a rod of refractory metal (molybdenum, tungsten, or tantalum) with a hole drilled along its axis from one end for electron bombardment heating. The electrical lead was an integral extension of the emitter, with a wall thickness and length giving the optimum lead resistance. By means of a jig, the emitter lead and flange were brazed to a metal-ceramic insulator in a single operation, using vacuum induction heating. Such an assembly was constructed for each different emitter material and/or spacing used.

Although one collector assembly thereby was reusable for many tests, two were constructed to reduce downtime of the test equipment during emitter changes. The flange of the emitter assembly was clamped firmly into a recess in the top of the collector, precisely positioning the emitter in the collector cavity. A silver plated, elastic, Inconel-X gasket* vacuum-sealed the joint between the flange and the collector. Leakage of cesium past this gasket was never detected, even though it was occasionally operated in excess of 950°C. However, a new gasket was required with each disassembly. The collector was

*United Aircraft Products, Inc.

Figure 15. Small Dismountable Cylindrical Converter

mounted on an integral stainless steel tube which protruded through the vacuum system base plate. A tungsten rod along the axis of this tube permitted the opening or closing of a small hole in the base of the collector cavity, to admit or remove gas during operation of the converter. After evacuation and outgassing, cesium was injected directly into the converter through this hole with a hypodermic needle. The tungsten needle valve was then closed, and the collector heated to distill the cesium into a cavity in the insulator flange, which thereafter served as the cesium reservoir. The reservoir and collector temperatures were independently controlled by regulating gas flow in the attached cooling coils. A total of about 10 different emitter assemblies were tested in this type of converter.

The success and relatively high performance of models of this type of converter led to the construction of the improved and larger model shown in Figure 16. The larger size was chosen to even further reduce the significance of stray heat losses and electrical edge effects. The emitter assembly was modified to allow use of the insulator flange itself as the mechanical vacuum seal, as shown. This resulted in a very simple emitter assembly which required only one small weld. Truing the silver-plated insulator flange with a lathe, and precise jiggling during welding, yielded precise location of the emitter in the collector cavity and a leak-free joint, as before.

Increased emphasis on impurity control led to an improved method of inserting the cesium charge into the larger model. After outgassing, a glass cesium ampule was crushed in an appendix which was subsequently used as the cesium reservoir. In this model, the reservoir temperature was regulated by an electrical heater. Also, to further reduce impurities, large capacity titanium metal getters were inserted in the passages between the collector cavity, the cesium reservoir, and the insulator.

To ensure strictly equivalent thermal and electrical conditions for comparison, various metals were evaluated as emitters in the large model by plating them onto a standard molybdenum electrode. Dense, uniform, and adherent coatings of tungsten and tantalum, 0.005 in. thick, were formed by standard methods of pyrolytic decomposition of the gaseous chlorides.¹⁹ Although this process did not yield adherent coatings of rhenium, a similarly adherent layer of rhenium was obtained by electrodeposition from aqueous solution. In addition to this standard evaluation of emitters, tests were also performed with uncoated

Figure 16. Large Dismountable Cylindrical Converter

molybdenum and uncoated tantalum emitter electrodes. Uncoated molybdenum electrodes were also used to evaluate performance at different spacings in this model (0.008, 0.015, and 0.100 in.). In all, about 10 different emitter electrodes have been tested in this converter thus far.

4. Permanently Sealed Cylindrical Converter

Upon the achievement of promising electrical output performance in the dismountable models, work was undertaken to investigate the problems of life-time and reproducibility in permanently sealed units more closely approximating practical devices. While versatility of components and flexibility of operation are emphasized in the dismountable (analytical) types, emphasis in the permanently sealed types is on simplicity, high reliability, and high reproducibility. It was expected that corrosion and other uncontrolled impurity effects observed in the dismountable models could be overcome in models designed specifically to accommodate the rigid controls characteristic of commercial electron tube production. As discussed later, this was indeed found to be so. A separate ("clean") room was equipped by Atomics International to permit final inspection of components (e. g. , leak check), final assembly, bakeout, cesium charge, and sealoff in a closely controlled procedure. Personnel assigned to this operation have its improvement as their specific and primary objective.

A typical model being regularly produced in this facility is shown in Figure 17. It should be noticed that it is very similar to the small dismountable model (Figure 15), which considerable direct experience has shown to be a very effective design. Except for the ceramic-metal bonds in the commercial seal, all vacuum joints in the unit are retort arc welded, as discussed in Section V-C-1. Large capacity active metal getters are interposed between the collector cavity, the insulator, and the cesium reservoir. All parts are individually outgassed in high vacuum well above operating temperatures, prior to assembly, and, as a unit, prior to sealoff by electron bombardment welding. The emitter, collector, insulator, and sealoff tube temperatures can be independently controlled. Therefore, either the insulator flange or the sealoff tube can be used as the cesium reservoir.

Approximately 20 models, of the type shown in Figure 17, have been produced and their characteristics tested. This includes models having emitters of molybdenum coated with tantalum, tungsten, and rhenium, as well as uncoated emitters of niobium, tantalum, molybdenum, tungsten, and rhenium.

Figure 17. Permanently Sealed Cylindrical Converter

Efforts to obtain a satisfactory coating of iridium have been unsuccessful thus far, but are continuing. Spacings ranging from 0.005 to 0.080 in. , and a heat shielded (cavity) configuration have been employed. Most models of this type have had about 5 to 7 cm² emitter area; but a model with about 60 cm² emitter area has been constructed here and operated extensively under another program.*

5. Variable Spacing Converter

To properly evaluate interaction of the important variables in the converter, as previously discussed, it is highly desirable to be able to change each variable continuously, while the others are recorded or held constant. Although the cylindrical configuration is preferable for most such measurements, it is mechanically impractical to continuously and uniformly change the spacing between cylindrical electrodes during operation. Therefore, a converter was constructed in which the spacing between plane electrodes could be precisely and continuously changed during operation. The first of this type operated satisfactorily and yielded important data.²⁰ However, the ceramic-metal insulator, into which the stationary collector was rigidly brazed, was subject to frequent failure, as a result of inadvertent application of excessive stress when the electrodes came in contact. Replacing the insulator required the complete disassembly of the converter. Cleaning, after more routine disassembly, also proved to be a significant problem.

These difficulties were overcome in the somewhat simpler and more versatile subsequent models of this type, illustrated in Figure 18. In these models, which are presently in use, the stationary emitter is supported by the insulator on an easily removable flange. This emitter assembly is very similar to that used in the cylindrical dismountable converters previously described. The collector is supported by a stainless steel tube which passes through a sliding vacuum seal to a rugged drive mechanism attached to the outside of the vacuum baseplate. By means of a differential thread, a single turn of the large adjusting screw moves the collector only 0.0125 in. Thus, changes in spacing may be made easily to within 0.0002 in. , and are measured

*"Flame Heated Thermionic Converter Research Program" carried out under Contract DA-36-039 SC 88982 with U. S. Army Signal Research and Development Laboratories, Fort Monmouth, N. J.

Figure 18. Variable Spacing Converter

with the attached precision dial indicator. The remainder of the converter is independently supported by a rigid tripod attached to the baseplate. The converter envelope, or "shell," is bolted to this tripod, and is vacuum sealed at its upper end by the removable emitter flange, using the type of metal gasket previously mentioned. The other end of the shell is permanently closed by a stainless steel bellows which permits free movement of the collector along its axis. Using a precision jig, the emitter surface is adjusted parallel to the collector surface, within 0.0005 in., by adjusting screws in the tripod legs. Ample space is available in the shell for cleaning and for mounting components, such as probes, viewports, and guard rings.

The collector is cooled by forced gas convection in an attached cooling tube. Its temperature is controlled by an attached electrical heater, which permits calorimetry to be performed on the heat reaching the collector. The temperature of the cesium reservoir is also electrically controlled. Cesium is distilled into the converter, prior to a pinch sealoff, as previously described. The interior of the envelope is lined with replaceable sheets of active metal, serving as a high capacity getter.

The area of the plane surfaces of the electrodes is about 2 cm^2 . The remaining surfaces of the emitter are covered with several layers of molybdenum foil. In addition to reducing stray radiant heat losses from the emitter, this foil supposedly prevents significant electron emission from these surfaces. This supposition, and other edge effects inherent to plane electrodes, are subject to evaluation by comparison with data from the cylindrical models for the same operating conditions.

6. Variable Spacing Triode Model

As previously mentioned, the topology of plane electrodes is such that it is difficult to prevent short circuit discharges from occurring between the hot emitter and cold portions of the emitter lead, which serve as spurious collectors.

Figure 19. Dual Diode Models

Differences in the observed performance of cylindrical and plane models, particularly a higher power density at low temperature in the plane models, have suggested that such spurious discharges may be responsible. Since these discharges can have both beneficial and detrimental effects, to be discussed later, one of the variable spacing (plane) converters has been modified to permit their evaluation. Except for the modified collector shown in Figure 18, this model is identical to the variable spacing diode. As may be seen, a third electrode is mounted through the center of the collector, in thermal contact but electrically insulated from it. The current to this electrode and its potential can be independently measured and adjusted, but its temperature is approximately that of the collector.

7. Dual Diode Model

In the in-core thermionic reactor, it is highly desirable to operate a number of cylindrical converters stacked in electrical series with a common cesium reservoir, i. e., within the same envelope.^{3,4} Two problems arise in this arrangement which are not amenable to analytical resolution. First, the significance of end effects in isolated cells must be known, to determine the number and length-to-diameter ratio of the cells in an optimum design. Secondly, to permit realistic design of such a converter assembly, it is necessary to know the degree of isolation required to prevent detrimental interaction of adjacent series-connected cells through the cesium vapor.

To evaluate these effects experimentally, the "dual diode" converter assembly, shown in Figure 19, has been constructed. In essence, this device is a combination of the variable spacing and cylindrical types previously discussed. A drive mechanism, similar to that for the variable spacing converters, allows the adjusting of the distance between the open ends of two independent coaxially-mounted cylindrical diodes. A flexible metal diaphragm seals the envelope, but allows axial movement of one of the diodes. When the ends of the diodes are in contact, the unit is equivalent to a single diode. Thus, end effects may be directly evaluated as the ends are separated and each diode is operated individually. Furthermore, interaction of the diodes, when operated in series, can be directly determined for various isolating structures interposed between them. The device therefore permits a realistic mockup of the conditions occurring between adjacent cells in an in-core thermionic reactor. Other components and procedures in this device are similar to those in converter types described previously.

7590-5519A

a. Small Dismountable Cylindrical Converter

7590-5501C

b. Large Dismountable Cylindrical Converter

7590-1805E

c. Variable Spaced Parallel Plane Converter

7590-1807A

d. Multicell Converter

7590-5519F

e. Sealed Cylindrical Converter

f. Sealed Cylindrical Converter (Plug In)

Figure 20. Converters Mounted for Operation

8. Experimental Installation of Converters

Converters are typically mounted on the baseplate of a high vacuum system, as illustrated in Figure 20. Operation of the converter entirely within a vacuum system permits convenient use of electron bombardment heating, thermally insulates and protects high temperature components from attack by air, and allows operation to be continued, despite a small leak in the envelope, until the liquid cesium charge is exhausted. By passing all electrical leads, etc., through a standard baseplate, and by using standard connectors, a complete converter assembly is easily removed and another assembly mounted in its place. Thus, a single test installation (i. e., vacuum system, power supply, recording and controlling equipment, and load) serves a number of converter models with a minimum of interference.

7590-1830

Figure 21. Typical Optima in Cesium Reservoir and Collector Temperatures

Figure 22. Typical Observed Maximum Power and Efficiency, with Associated Optimum Currents and EMF's

VI. REVIEW OF EXPERIMENTAL RESULTS AND THEIR SIGNIFICANCE

A. GENERAL

The large amount of experimental data which has been obtained thus far may be divided conveniently into three general categories:

- 1) The first category includes purely exploratory data which either yielded a negative result, have been superceded by improved subsequent data, or have been rejected as invalid, due to errors recognized after it was obtained. While such data have been valuable in defining the course of subsequent work, they at most deserve only qualitative description.
- 2) The second category is a large body of presumably valid data which, although obtained incidental to the primary purpose of the experiment, are fertile ground for stimulating and testing new hypotheses. Except where such an existing hypothesis allows their interpretation or their correlation with other results, such data will not be reported in detail, but may appear in later reports, as their usefulness is recognized.
- 3) The remaining category includes data representing the most advanced progress toward the specific objectives previously described. While these data are of interim nature, subject to confirmation or revision by further work, they illustrate the significance and present status of the work, and so will be presented in detail.

B. PERFORMANCE OPTIMIZATION

1. Basis for Comparison

As previously described, all of the converters thus far (including the initial models) were constructed to allow the input and output variables of the device to be changed over wide limits. It was therefore easy to experimentally determine the relationship among the variables giving maximum efficiency or maximum power. Figures 21 and 22 show typical observed optima in collector temperature, cesium pressure (in terms of reservoir temperature), output emf, and current. The conditions under which an optimum spacing is observed will be discussed later. By the iterative adjustment process described in

Figure 23. Typical Observed Optimized Performance Characteristics, Compared with Computed Ideal Values

Section V-B-3, the values of these optima, corresponding to maximum efficiency or maximum power at each emitter temperature, can be obtained experimentally. Such fully optimized performance data, exemplified in Figure 23, are obtained for each modification of all converters tested here, and are used as the primary means of comparing their relative merits. Investigation of the off-optimum characteristics is usually centered about these optimum values. Early recognition and continued emphasis of fully optimized operation has permitted the observed performance achieved here to be among the most advanced in thermionic conversion during the existence of this program.

As an aid in intuitive comprehension, it is convenient to choose a single optimized variable in comparing the various converter modifications. Although not always the most important quantity, the maximum (optimized) efficiency is significant in most practical applications, and reflects any significant changes occurring in the other important thermal and electrical quantities in the converter. Changes thereby detected can be identified and explored by detailed attention to the individual quantities. However, such an approach is appropriate only when spurious heat transfer in the converter is small, compared to necessary heat transfer. Since the cylindrical converters employed satisfy this requirement, observed overall efficiency will be used, for present purposes, as the basis for comparing their various modifications.

2. Emitter Evaluation at Constant Spacing

As may be seen in Figure 23, the temperature dependence of the optimum quantities is in general qualitative agreement with that indicated by the analysis of the emission limitations (Section III). The observed optimum collector and cesium temperatures, which primarily affect the emission characteristics of the electrodes, are in fair quantitative agreement with computed values. However, the efficiency and other observed optimum output variables are substantially degraded from the values obtainable in the absence of transport effects. As previously discussed, it should be possible to reduce this degradation by decreasing the electrode spacing and/or by perspicacious choice of electrode materials. Figure 24 shows that the emission-imposed limit is indeed more closely approached as the spacing is decreased. Detailed experimental exploration of the spacing dependence is discussed in Section VI-B-3.

A series of tests, to evaluate electrode materials by direct comparison in standard converters, has been a central theme unifying most of the experimental work. The first such series was performed in the small dismountable cylindrical converter. The fully optimized performance of this converter, using tantalum, molybdenum, and tungsten electrodes, spaced 0.032 in. apart, was determined under as nearly identical conditions as could be achieved at the time. In these tests, the performance of molybdenum was observed to be clearly better than that of tungsten or tantalum; efficiency and power density for molybdenum were nearly twice those for the other materials at most emitter temperatures. For this reason, molybdenum was chosen as a reference emitter for investigating the effect of spacing and diluent gases. However, in the course of these further tests, it was found difficult to reproduce results, from one test to another, under supposedly identical conditions. Furthermore, during extended operation, changes in output with time, and eventual failure by short

Figure 24. Effect of Spacing on Observed Optimized Efficiency, Using Molybdenum Diodes

circuiting, were encountered. These effects, and the corrosion which accompanied them, are discussed in Section VI-C.

The larger model of the dismountable cylindrical converter was then constructed, to achieve better control of the variables suspected of being responsible for the lack of reproducibility in the smaller models. As previously discussed, changes were instituted to improve control over uniformity of electrode spacing, temperature uniformity, temperature measurement, and cesium purity. To ensure that temperature and potential gradients along the emitter would be the same for all materials, a standard molybdenum electrode was employed which was coated with the test materials, as previously described. Results of the emitter evaluation test series, performed in this model at 0.015-in. spacing, are shown in Figure 25. As may be seen, molybdenum again gave performance superior to the other metals tested. The results were somewhat disappointing, in that no satisfactory correlation of the "saturation" efficiencies with bare work function or thermal emissivity was obtained. However, the observed value of the threshold temperature for efficient conversion and its dependence on bare work function agreed with that expected from the emission analysis. Diffusion of molybdenum through the coating might have affected the results at the higher temperatures. Again, extended tests with molybdenum emitters showed that reproducibility was still a problem, and thus indicated that satisfactory control had not yet been achieved over an important variable. Since it was felt that all other factors were under adequate control in this model, and since corrosion was still observed during extended operation at high temperatures, the uncontrolled variable was tentatively identified as an impurity effect. Further emitter evaluation testing was therefore discontinued, and a major effort mounted to achieve the degree of impurity control necessary to obtain reproducible results and eliminate corrosion.

As a result of this effort, permanently sealed cylindrical converters, employing highly controlled production techniques and high purity cesium, as previously described, are now being employed for emitter evaluation testing. Operation appears to be highly reproducible and free of corrosion in these converters. However, the results thus far (Figure 26) indicate that the suppression of impurity effects has resulted in a substantial reduction in the performance, especially for molybdenum. It should now be possible to define, unobscured by impurity effects, the optimum emitter for each spacing and temperature, by the

Figure 25. Emitter Performance Evaluation Series in Large Cylindrical Converter Model

7590-1843

Figure 26. Emitter Performance Evaluation Series in Permanently Sealed Cylindrical Converter Models

Figure 27. Spacing Dependence of Maximum Power Output Observed with First Variable Spacing Converter Model

Figure 28. Spacing Dependence of Maximum Power Output Observed with Improved Variable Spacing Converter Model

approach described in Section IV-B. It is possible that this optimum emitter was unintentionally approached in previous converters by the action of impurities on the molybdenum surface. If so, it should be possible to recover the high performance previously obtained, without corrosion and other undesirable impurity effects, by employing a material which does not depend on impurities to achieve the optimum surface properties. On the other hand, it is possible that converter performance was improved by an impurity effect not directly involving the emitter, as considered in Section IV-C. However, the importance of defining the optimum emitter material is not diminished by such a possibility.

3. Effect of Spacing

Concurrent with the above-described work on cylindrical converters, the variable spacing converter was employed to obtain continuous data on the effect of spacing more rapidly than by constructing a series of individual converters with fixed electrodes. Results obtained, using molybdenum electrodes in the first model of this type, are shown in Figure 27. Since this type of converter is inherently inefficient, due to thermal losses from nonemitting surfaces, the data represent the maximum power output obtainable at each spacing and temperature (i. e., the cesium pressure and collector temperature were adjusted to give maximum power output at each spacing, with the emitter temperature held constant). As may be seen, the performance rapidly increases as the spacing is decreased. In fact, it may be seen that a power density of about 20 w/cm^2 was obtained at 1800°C for a spacing of about 0.005 in., which approaches the performance desired for a useful in-core thermionic reactor system. However, as with the early dismountable cylindrical converters, difficulty with reproducibility of the data was encountered, accompanied by significant corrosion at the higher temperatures.

The improved model of the variable spacing converter was then constructed, as previously described, to obtain the high degree of impurity control achieved in the sealed cylindrical models. The spacing dependence of optimized power output for this model, using molybdenum electrodes, is shown in Figure 28. As in the cylindrical models, suppression of impurity effects greatly improved the reproducibility of the data and eliminated corrosion; but, at the same time, it resulted in a substantial reduction in performance. As previously discussed, it should now be possible to proceed with the approach described in Section IV-B to define the optimum emitting material for each spacing and

temperature. In this approach, the variable spacing model is used for interpolating the results obtained in the cylindrical models to intermediate spacings.

It should be noted, in Figure 28, that an optimum spacing, giving a maximum in the optimized power, is observed for emitter temperatures in the ion-generation-limited region defined in Figure 8 (i. e. , $<1750^{\circ}\text{K}$). At higher temperatures, no such maximum is observed up to contact of the electrodes. At extremely small spacing (<0.001 in.), the nonoptimized power density increases rapidly as the spacing is decreased, as shown in the inset in Figure 28. The converter thus approaches close-spaced vacuum diode operation at those spacings (i. e. , electron space charge becomes so small that ion generation is unnecessary).

While quantitative basic understanding of the effect of spacing on performance has not yet been obtained, qualitative hypotheses have been made which are being investigated experimentally. The existing data can be interpreted in two basically different ways. In one interpretation, the ions for space charge neutralization are assumed to be produced thermally. At high temperatures (i. e. , in the electron-emission-limited region), an adequate number of ions are generated, but detrimental transport effects reduce the output power from the emission limit as the spacing is increased. This reduction in power occurs as a result of the potential drop arising from the internal impedance to current flow between the electrodes. As spacing is increased, the optimum cesium pressure is reduced from the emission-imposed values to minimize this internal impedance. Also, according to this interpretation, the output initially rises as the spacing is increased at low temperatures (i. e. , in the ion-generation-limited region), due to the improvement in ion lifetime by multiple scattering. No such initial increase occurs at higher temperatures, since a surplus of ions is presumably available there at all spacings.

An alternate interpretation of the data is that ions are assumed to be produced in the space between the electrodes by electron impact, as in ordinary gas discharges. Under this circumstance, the spacing must be greater than a certain number of mean free paths for most efficient ion production. The observed maximum in optimized power is thus identified with this spacing. As electrode spacing is decreased, the pressure must be increased proportionally to maintain the optimum number of mean free paths between them. At high

temperatures, the pressure required by emission considerations is so high that the power maximum presumably occurs at too small a spacing to be observed.

Since both of these interpretations are consistent with present experimental data, an unqualified choice between them is not yet possible. It seems likely that the true situation is a combination of these effects, with one or the other being dominant under different circumstances. Resolution of this question should occur in the course of determining the relationship between optimum pressure and spacing with different materials, as previously described.

C. ANOMALOUS (IMPURITY) EFFECTS

1. Reproducibility and Stability of Performance

The difficulty in obtaining reproducibility in the series of electrode evaluation tests and its identification as an impurity effect have already been discussed. However, this and associated considerations are of sufficient importance to merit individual description. In one sense, these effects were detrimental, in that they impeded progress toward the specified objectives. In another sense, they were fortuitous, in that they permitted early direct experimental demonstration that the performance required for an attractive thermionic reactor system could be achieved in a diode at reasonable spacings and temperatures. One of several such cases in point is illustrated by the optimized performance once observed for the large cylindrical converter, shown in Figure 29. An overall efficiency of 16.8%, with an output of 8.3 w/cm^2 at 1.0 v, was observed at 1650°C, using a spacing of 0.008 in. This performance was consistent with the results obtained in the first variable spacing model at this temperature (Figure 27), which thereby permitted extrapolation to about 20% efficiency and 18 w/cm^2 at 1800°C. The test was terminated, after about an hour at the higher temperatures, by failure of the heater filament. Upon disassembly for inspection, substantial corrosion was observed to have occurred.

To explore the reproducibility and stability of this operating point, a permanently sealed cylindrical converter with the same spacing was constructed. More attention was given to outgassing and general cleanliness of the sealed unit, but cesium from the same source was used in both converters. The performance of the sealed converter was found to be similar to that in Figure 29, with a slightly lower efficiency, as expected, due to its smaller size. The unit

Figure 29. Anomalous High Performance Observed with Large Cylindrical Converter Model

was then put on life test, where it delivered about 7 w/cm^2 at between 14 and 15% overall efficiency for 130 hr before gradual short circuiting by corrosion products began. It should be recalled that the efficiencies quoted as "overall" are the ratio of observed electrical output power to total input power. Computed efficiencies, in which nonessential (stray) heat losses are subtracted from the total input power, would be somewhat higher.

An effect presumably related to irreproducibility and corrosion effects was the pronounced "activation" observed in the early model converters. Upon establishing thermal equilibrium after an increase in emitter temperature, the optimized output power was usually observed to increase steadily, for 15 min or more, to an equilibrium value of as much as 25% above the initial value. Similarly, if the collector temperature were increased substantially above its optimum temperature, the output would decrease; but, upon returning to the optimum temperature, the optimized output would be found to be considerably higher than before this "activation" cycle. However, over the period of an hour or so, the output would steadily return to its initial value. It was thought, at the time, that the increased performance resulted from decontamination (outgassing) of the electrodes at the higher temperatures. However, the virtual absence of these effects, and the lower performance in the present (clean) models, indicate that contamination of initially clean electrodes gave rise to these activation effects.

Although the mechanism by which impurities improved the performance has not yet been firmly identified, some clues from observed behavior can be used to estimate their probable origin. At first thought, it might appear that the effect of the impurities was to substantially lower the work function of the collector; since cesiated surfaces, contaminated with oxygen and the halogens, have long been known and employed as low work function surfaces.²¹ However, it thereby would be expected that the optimum collector temperature would be proportionately lowered (See Equation 2) during anomalous high performance. Since the optimum collector temperature was observed to be the same, for both high and low performance operation, it seems unlikely that only changes in the collector were involved in the observed effects. While it may be possible to employ an impurity to lower the collector work function, this method of improving performance is unattractive for space application, since it requires decreasing the heat rejection (radiator) temperature, as discussed in Section III-B.

On the other hand, impurities such as oxygen, nitrogen, and the halogens are known to substantially increase the work functions of high temperature emitters, and to change their cesium adsorption characteristics.²² Thus, an alternate hypothesis is that such impurities change the bare work functions of the converter emitters, such as to more closely approach an optimum surface. As previously discussed, such changes in emitter bare work function would result in large changes in the optimum reservoir temperature (See Figure 4). Although a consistent correlation has not yet been obtained, such changes in optimum cesium pressure have been observed to be associated with "activation" and other anomalous changes in performance. Therefore, the implication is that the observed effects primarily involve contamination of the emitter. The probable nature and source of the contaminating impurities is discussed in the following section.

2. Corrosion and Lifetime

As has been repeatedly implied, anomalous high performance was almost always accompanied by a high rate of corrosion of the emitter. This corrosion resulted in the transport of molybdenum or tungsten across the electrode interspace at a rate far in excess of that possible by ordinary vaporization. The other metals tested exhibited neither the corrosion nor the anomalous high performance. The corrosion products were deposited on the collector as a metallic (molybdenum) layer which apparently did not adversely affect performance until it flaked off and shorted the electrodes. Since the amount of corrosion (typically, 0.0005 in.) was far greater than could have occurred if the impurity were consumed in the process, a cyclical mechanism appeared to be involved. A study of likely chemical processes²³ indicated that active impurities such as oxygen, which could readily account for the corrosion under other circumstances, would react with the cesium vapor and be quickly eliminated from the converter atmosphere by condensation on the collector. However, the study further indicated that only a small amount of hydrogen could act as a carrier to cyclically return the oxygen to the emitter. As previously described, active metal getters (of outgassed titanium or zirconium) were enclosed within the envelope to eliminate hydrogen. The first use of such a getter did not result in an appreciable reduction of the corrosion. However, a post-operation spectrochemical analysis of the getter showed it to be saturated with hydrogen. On this particular occasion, the cold trap for the oil-diffusion-pumped high

vacuum system was not functioning, although one is usually employed. It was therefore concluded that pump oil vapor was thermally decomposed on the hot exterior surface of the envelope, and that the atomic hydrogen thus liberated diffused through the converter envelope into the cesium vapor. Although the cesium reservoir can act as a getter for hydrogen, it is quickly saturated in the presence of even a small source of hydrogen. Corrosion has not been detectable in any subsequent converters, which now all include getters and are only operated with thoroughly trapped vacuum systems (e. g., one permanently sealed cylindrical converter was operated continuously, with an emitter temperature of about 1700°C, for 170 hr without failure or detectable corrosion).

As previously described, reduction in performance accompanied the elimination of hydrogen and the associated corrosion from the converter. Although recent preliminary results observed elsewhere²⁴ indicate that hydrogen can substantially lower the work function of a low-temperature cesiated surface, the considerations cited in the immediately preceding section indicate that the primary effect observed here involves the high temperature emitter, rather than the low temperature collector. A reasonable assumption, which correlates the corrosion and performance effects, is illustrated in Figure 30. An active impurity (e. g., oxygen) is evolved from the interior of the electrodes, and is deposited as an oxide on the collector. There it is reduced by any free hydrogen present to form H_2O , which is subsequently decomposed at the hot emitter surface. Free hydrogen is thereupon liberated to continue the cycle. The oxygen impurity, continuously supplied to the emitter surface by this process, is assumed to account for the correlated performance and corrosion effects. While on the surface, the impurity alters the emitter work function, as previously described. Upon leaving, it carries away part of the surface as the gaseous oxide, which in turn deposits the corroded emitter material on the collector when reduced by free hydrogen.

It is perhaps worthwhile to estimate why these impurity effects were not observed in all converters operated here, or were not generally observed by other groups. First of all, it is likely that only molybdenum and tungsten participate with oxygen in the cycle shown in Figure 30, since the oxides of other materials generally employed as emitters are not sufficiently volatile. Also, some emitter materials (e. g., tantalum) actually serve as hydrogen getters, if a portion of the emitter (e. g., the lead) is operated at lower temperatures.

Figure 30. Conservative Hydrogen and Oxygen Cycles Causing Emitter Corrosion and Supplying Emitter-Modifying Oxygen Layer (a similar cycle can occur with a halogen, instead of oxygen)

Secondly, the cycle proceeds rapidly, only if the collector is operated at the higher (optimum) temperatures. This dependence of the cycle on collector temperature presumably gives rise to the collector "activation" effect described previously. Finally, the cycle proceeds vigorously, only when the converter is operated at relatively high emitter temperatures ($>1600^{\circ}\text{C}$), with the high optimum cesium reservoir temperatures ($\approx 350^{\circ}\text{C}$) associated with molybdenum and tungsten at these temperatures. In addition to a greatly increased reaction rate at these emitter temperatures, the equilibrium pressure of hydrogen over cesium is also quite high at these reservoir temperatures. In summary, therefore, it appears that the conditions for occurrence of a significant impurity cycle coincided with the region of emphasis in the converter studies here [i. e., metal electrodes (including Mo and W), optimum collector operation, and relatively high emitter and reservoir temperatures appropriate to a useful thermionic reactor system]. It follows that groups omitting any one of these conditions would not have observed appreciable corrosion or anomalous high performance effects.

D. OUTPUT CHARACTERISTICS

Typical volt-ampere characteristics obtained for high and low load resistance, as discussed in Section V-B-4, are shown in Figures 31a and 31b. A number of features, and the nomenclature used here to identify them, are indicated in the figures. The dependence of the existence, location, and magnitude of these features on emitter temperature and cesium pressure, obtained using the circuit of Figure 12a, are illustrated in the series of oscillograph curves in Figure 32. In addition, the distortion of the curves which occurs at high frequencies is exemplified in Figure 33, and the effect of spacing in Figure 34. It is apparent, from these figures, that the interaction of all the variables is quite complex. This complexity is annoying, from a system design standpoint, but its analysis provides valuable quantitative insight into the basic processes occurring. Although a sizable amount of analysis of these characteristics has been performed here and elsewhere,²⁵ their interpretation is still the subject of considerable controversy, since no theory consistently describes the observed data, except under highly specialized conditions. However, a review of the various theories and their status will not be included in this report. One approach, particularly pertinent to the objectives of the experimental converter work here, involves a semiempirical relation of the observed interelectrode power dissipation to general classes of transport processes. Many features of the true diode characteristics (Figure 31b) can be semiquantitatively described by this approach and related to the basic processes. However, the greatest

Figure 31. Typical Volt-Ampere Characteristics ($T_e = 1310^\circ\text{C}$,
 $T_{Cs} = 176^\circ\text{C}$, $T_c = 450^\circ\text{C}$, Spacing = 0.080 in.)

utility of the approach is expected to be its use in extending the analysis of ideal diode performance (Section IV-B-1) to include transport effects, and thus permit analytical description of the overall optimization process described qualitatively in Section IV-B-2.

E. PERFORMANCE IMPROVEMENT OF DIODES

1. Cavity ("Heat-Shielded") Emitter

At the onset of the program and since, several schemes have been conceived here for improving performance by removing the basic limitations on the elementary cesium vapor diode. While many of these are in the purely speculative stage, awaiting future evaluation, and as such will not be mentioned here, a few which have received at least preliminary evaluation will be described.

As mentioned in Section V-C-2, the first converter models constructed were designed to allow use of cavity, or "heat-shielded," emitter configurations. When the electron mean free path and/or Debye length is sufficiently small to allow the electric field (or plasma) to penetrate into cavities in the emitter, the electron-emitting area can, in principle, be made very large, as compared to the projected area exposed to the collector. In the past, such "heat-shielded cathodes" have been used extensively²⁶ to improve the efficiency of high power rectifiers and thyratrons by achieving an electron-emitting area more than 100 times the effective heat-radiating area. If this principle could be similarly applied to the thermionic converter,^{10,12} the emission-imposed limit on efficiency could be extended to approach the Carnot limit (i. e., ϵ in Figure 2b could be made arbitrarily small).

Other potential advantages accrue with this approach, in addition to the reduction in radiant heat transfer. For instance, a much higher emitter vaporization rate can be tolerated, since only a small fraction of the atoms leaving the surface in the emitter cavity reach the collector surface. In addition, at ion-generation-limited temperatures, present data indicate that there is an optimum electrode spacing which presumably favors the production or conservation of ions. This suggests that the size of the cavities in the emitter could be chosen to optimize ion generation in their interiors; while a much closer inter-electrode spacing, adjacent to the cavity openings, would be employed for

Figure 32. Dependence of the Volt-Ampere Characteristics on Emitter and Cesium Reservoir Temperatures for Low-Resistance Load (Figure 12a)

BLANK

a. 45 cps

b. 60 cps

c. 90 cps

d. 120 cps

e. 180 cps

f. 240 cps

Figure 33. Effects of Cycle Frequency on Volt-Ampere Characteristics
(taken with circuit of Figure 12a) ($T_c = 1350^\circ\text{C}$, $T_{Cs} = 310^\circ\text{C}$,
Spacing = 80 mils. Zero amps is one grid below, and
zero volts is two grids to right, of grid center.)

	SPACING
1	10 MILS
2	20 MILS
3	40 MILS
4	60 MILS
5	80 MILS

EMITTER TEMPERATURE 1685°C
CESIUM RESERVOIR TEMPERATURE
343°C

a. Cesium Reservoir Temperature, 343°C; Optimum for 0.010 in. Spacing

	SPACING	Cs TEMP
1	10 MILS	343°C
2	20 MILS	337°C
3	40 MILS	327°C
4	60 MILS	318°C
5	80 MILS	318°C
6	100 MILS	318°C

EMITTER TEMPERATURE 1685°C

b. Cesium Reservoir Temperature Optimized for Each Spacing

Figure 34. Effect of Spacing on Volt-Ampere Characteristics

efficient electron current transport. Furthermore, if the cavities were to extend deep into the interior of the fuel element in an in-core thermionic reactor, electron cooling could potentially aid in transferring heat from the interior, and ions (or excited atoms) formed in the hotter interior might allow the outer surface of the element to operate efficiently below the ion-emission temperature limit.

The major question in the application of the cavity emitter to thermionic conversion is whether adverse transport effects, introduced by the presence of the cavity, outweigh all potential associated improvements. Due to the present imperfect knowledge of the transport effects, this question can be answered only by direct experimental observations in a converter. Comparison of plain and cavity emitters, in otherwise identical converter models, has failed to show any significant difference in performance. Lack of reproducibility of early data impugned the validity of this result in the initial models. Work therefore was concentrated on achieving reproducible data and more complete understanding in the elementary diode, before attempting to evaluate the more complex configurations. Since considerable progress has been made toward satisfying these criteria, it is now appropriate to proceed with the evaluation of concepts, such as the cavity emitter. Further work should lead to configurations which will obtain a net advantage over the plain emitter. It is possible that the use of inert gas additives, as described in the next section, will greatly benefit this approach.

2. Inert Gas Additive

One means of improving performance in the ion-generation-limited region of temperatures (i.e., low temperatures) is to provide an auxiliary ion source, as in the various triode schemes.¹⁵ An inherently more efficient and less complex approach is to improve the utilization of spontaneously produced ions in the diode (whether by surface or volume ionization), to the point where they can still provide neutralization at these lower temperatures. Equation 4 suggests that the effectiveness of a given ion emission current, q , toward neutralizing an electron emission current, J , can be improved by increasing α , the ratio of the respective mobilities of the electrons and ions. It would appear that the threshold temperature for efficient operation could be lowered by introducing a gas having an electron-scattering cross section which is small, as compared with its gas-kinetic cross section.¹⁰ The most favorable gas appears

to be argon. Due to the Ramsauer effect, the electron-scattering cross section of argon is less than 1/1000 that of cesium at thermal energies, even though their gas-kinetic cross sections are comparable. Therefore, a 10/1 mixture of argon and cesium vapor should pass electrons essentially as easily as pure cesium vapor. However, for a given ion current, the cesium ion density would potentially be a hundred-fold higher in the mixture, due to the corresponding reduction in mobility of the cesium ion.

As described in Section V-C-3, gas at a specified pressure could be added to or withdrawn from the cesium vapor during operation of the small dismountable cylindrical converter with 0.030 in. spacing. With the converter operating in an optimized condition, at an emitter temperature in the ion-generation-limited region, addition of argon up to 20 mm pressure produced no significant change in performance, other than that attributable to increased thermal conduction. Again, this negative result led to a decision to postpone further work on this approach until more advanced understanding of pure cesium operation was obtained. Based on present knowledge, several postulates can be advanced to explain the negative results obtained under the conditions employed. Further tests should therefore be undertaken, to identify the correct interpretation. If the existing interpretations are valid, it should be possible to obtain the advantages sought, when the proper combination of emitter material and spacing are employed.

3. Active Gas Additives

The possible effect of active gases on converter operation were discussed in Section VI-C as impurity problems, rather than as desirable effects obtainable by controlled addition of active gases. However, it seems quite likely that such beneficial effects might be utilized, now that uncontrolled impurities have been suppressed and a degree of understanding of the elementary emission processes achieved.

In the tests described in the immediately preceding section, controlled bursts of active gases (O_2 , N_2 , H_2 , air, natural gas) were admitted to the diode during operation. In each case, an abrupt initial decrease in output was observed, with full recovery occurring within less than a minute. While these qualitative tests demonstrated a high degree of inherent stability of the converter against such bursts, a systematic approach, seeking optimum operating conditions for each additive, is necessary to achieve a beneficial effect.

Two approaches are recognized as having potential use. In one, an active additive would be sought which would provide optimum electrode surfaces and be compatible with presently employed electrode materials. Recently reported results²⁷ indicate that halogen additives may be quite useful. In addition to potentially having beneficial effects on the electrode work functions, as previously discussed, the halogens offer the possibility of actually transporting emitter material that has deposited on the collector back to the emitter. Conditions under which this could occur have been explored elsewhere.²⁸ However, a net beneficial performance in a converter has not yet been demonstrated. An alternate approach is to seek electrode materials which are compatible with the previously discussed oxygen cycle, so that the beneficial effects observed could be utilized without corrosion.

VII. SUMMARY AND CONCLUSIONS

In terms of known technology, the in-core thermionic reactor concept appears to have distinct practical advantage over the other types. Furthermore, it appears to be the only type of thermionic reactor system with sufficient potential superiority over competing systems to make its development worthwhile, especially for space application. At temperatures required by any thermionic converter to achieve the performance necessary for such a useful in-core system, the elementary cesium vapor diode is basically capable of achieving this performance (i. e., no auxiliary ion source is necessary). Fundamental and practical considerations indicate that elementary refractory metals with high work functions are the best known materials for emitters in high performance thermionic diodes. Therefore, this program seeks to define, by direct experimental and analytical evaluation, the refractory metals and associated optimum operating conditions which give the highest performance at the lowest emitter temperature for cesium vapor diodes of realistic practical design.

The analytical basis and experimental technology for systematically accomplishing this objective have been established. Converter performance has been experimentally observed, at moderate temperatures, which approaches that required for a useful in-core system. However, these results were obtained in the presence of impurity effects which adversely affected lifetime and reproducibility. The impurity effects are now under control, and the systematic approach to define the conditions for obtaining high performance, in their absence or by their controlled utilization, is in progress. Based on the results obtained, it appears that this approach has a high probability of experimentally demonstrating the performance required for a realistic and useful thermionic reactor system.

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