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HIGH TEMPERATURE HEAT PIPES FOR WASTE HEAT RECOVERY

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Abstract

Operation of heat pipes in air at temperatures above 1200 K has been accomplished using SiC as a shell material and a chemical vapor deposit (CVD) tungsten inner liner for protection of the ceramic from the sodium working fluid. The CVD tungsten has been used as a distribution wick for the gravity assisted heat pipe through the development of a columnar tungsten surface structure, achieved by control of the metal vapor deposition rate. Wick performance has been demonstrated in tests at approximately 2 kW throughput with a 19-mm-i.d. SiC heat pipe. Operation of ceramic heat pipes in repeated start cycle tests has demonstrated their ability to withstand temperature rise rates of greater than 1.2 K/s.

Introduction

High temperature industrial processing furnaces use nine quad of fossil fuel energy per year with a high proportion lost to the atmosphere in the form of high temperature exhaust gases. Effective high temperature recuperators could recover one to two quad per year of this lost energy. However, high effectiveness recuperation of heat from high temperature furnaces is restricted by the operating temperature limitations of heat exchanger materials. Typically, loss of strength restricts the air preheat temperature capability of recuperators constructed from high temperature steels to about 1000 K. Because of the temperature limits imposed by metallic materials ceramic recuperators have in the past been widely used in industry. However, in recent years they have been largely replaced with metallic units, using a variety of iron-, nickel-, and chromium-based alloys, because of inherent design problems. Conventional ceramic recuperator construction employs a large quantity of ceramic blocks cemented together in a checker-work array providing separate passages for air and waste gas streams. These units are massive, expensive, and invariably develop high leakage rates under the effects of vibration and thermal cycling. The metallic units tend to be cheaper and much less leakage prone. However, their effectiveness in preheat of incoming air is limited to about 0.4 when the temperature of the exhaust gas is over 1500 K. Radiation recuperators will accept flue gases at temperatures of up to 1800 K but are similarly limited in air preheat capability and suffer the additional disadvantage of high pumping power requirements.

The increasing cost of fossil fuels has generated renewed interest in the development of ceramic recuperator elements in order to eliminate their traditional shortcomings and exploit their temperature capability. Much of the ceramic recuperator development resulting from this renewed interest has been concentrated on attempts to eliminate leakage paths and to reduce cost through the use of more compact designs. The result typically is a variety of tube and shell exchanger with a minimum of ceramic tube joints. The problem common to these designs is stressing of the comparatively fragile ceramic tubes by differential thermal expansion. This problem is exacerbated by the use of rigid ceramic joints and by increased tube lengths between joints; both design features normally used to reduce leakage.

The development of a recuperator using heat pipes made of ceramic materials could eliminate the leakage and stress problems caused by thermal expansion. Mounting of individual heat pipes at a single point as indicated in Fig. 1 would permit free expansion under temperature changes, reducing thermal stresses and eliminating the need for sliding seals. The effectiveness and design advantages of heat pipe recuperators have been demonstrated at more moderate temperatures and include redundancy of operation; a particular advantage when employing brittle materials such as ceramics; ease of cleaning and tube replacement, and high overall heat transfer capability; limited primarily by the surface heat transfer coefficients on the gas side of the tubes. Only the lack of heat pipe designs capable of operating in combustion atmospheres at temperatures above 1200 K has restricted the use of heat pipe based recuperators in high temperature process heat applications.[1]

The basic element required for higher temperature recuperator designs, a ceramic heat pipe capable of withstanding the corrosive waste gas environment is under development at the Los Alamos Scientific Laboratory (LASL).

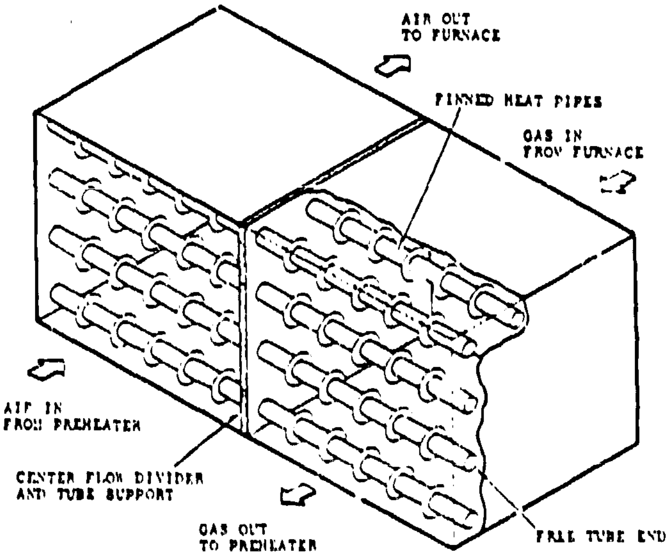

Fig. 1. Heat pipe recuperator arrangement.

Heat Pipe Design Requirements

Before development of ceramic heat pipes was started the potential industrial applications were reviewed in order to ensure that the development program was tailored to realistic requirements. Of the many industrial applications for high temperature recuperators, such as closed cycle gas turbines, fluidized bed fossil fuel combustors, process heat recovery, etc., a few specific applications were chosen for determining the design goals of the basic research program. These specific cases consisted of steel reheat furnaces, glass melting furnaces, and aluminum remelt furnaces. Requirements for recuperators for these three applications are similar, with the highest temperatures encountered in the glass furnace case.[2] Average values for the recuperator design parameters for these applications are summarized in Table I.

A requirement for most of these industrial applications is that the recuperator, in order to be added to an existing installation, must be reasonably compact. This is taken to mean a surface compactness on the order 165 m^2/m^3.

TABLE I

AVERAGE VALUES FOR CERAMIC HEAT PIPE RECUPERATOR DESIGN

Gas/air mass flow rate:	2 to 3 kg/s
Fuel:	Natural gas or oil. Combustion ratio aprox. 20:1 with air.
Furnace inlet temperature:	Limited by burner/furnace design to 1150/1250 K
Operating pressure:	Approximately 1 atmosphere
Allowable pressure drop air:	2500 to 3000 N/m^2
Allowable pressure drop gas:	2700 to 5000 N/m^2
Operating cycle:	Continuous.

The influence of these requirements on the heat pipe design is a function of the gas side heat transfer coefficients to be expected, the operating temperature range, and the operating atmosphere. Recuperators having the desired characteristics can be expected to have surface heat transfer coefficients in the range of 50 to 150 w/m^2 K, with gas velocities of 6 to 10 m/s. The surface compactness requirements will probably necessitate the use of some extended surface. The oxidizing atmosphere will limit the use of most refractory metals.

It is assumed that ceramic heat pipe heat transfer elements will be more expensive than conventional metallic heat exchange surfaces in stainless steel alloys. The recuperator design for the applications considered therefore will consist of a high temperature, ceramic heat pipe section, and a lower temperature metallic section operating as a pre-heater. The transition point for change to a metallic surface will be limited to about 1000 K by material considerations. The operating range of the heat pipes will span the range from 1650 K to 950 K. Appropriate working fluids for this temperature range are sodium and lithium.

Assuming that augmentation of the external heat transfer area of the heat pipes is limited to < 4:1 by the characteristics of the materials, and that gas-to-surface temperature differences are ~150 K, based on the required overall heat exchanger effectiveness of 0.7, the radial flux density for the heat pipes will be approximately 10 W/cm^2. The affect of L/D ratio for the heat pipes on the peak axial heat flux is indicated in Fig. 2. The figure also gives the axial heat flux limit due to onset of sonic flow as a function of temperature. The desireable limit for the heat pipe operating temperature will be below the peak limit for metallic heat exchange material or ~ 920 K, corresponding to a stainless steel material limit of approximately 1000 K. As is indicated on the figure the corresponding L/D ratio will be ~ 50 for sodium working fluid.

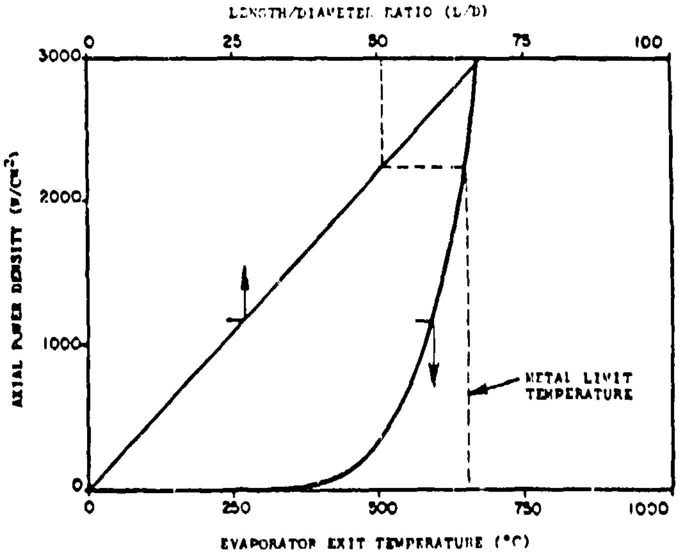

Fig. 2. L/D Ratio vs temperature.

The alkali metal working fluids which are appropriate to the operating temperature range of the ceramic heat pipes are not generally compatible with ceramic materials. The ceramic material must be protected from contact with the working fluid. This can be accomplished by using chemical vapor deposition (CVD) to line the interior of the ceramic tube with a layer of refractory metal known to have good long-term high temperature chemical compatibility with the alkali metals. The refractory metals: tungsten, molybdenum, and niobium are good containers for the alkali metals, if protected from oxidation. Therefore, the ceramic heat pipe is designed with an internal surface of refractory metal and an external surface layer or tube of ceramic.

The general configuration of the ceramic heat pipe as determined from these requirements is shown in Fig. 3. It consists of a ceramic tube with an integral closure at the evaporator end. The tube is lined with a protective layer of refractory matched in coefficient of expansion to the envelope. The tube is closed with a plug at the condenser end. Closure is accomplished by welding or brazing the metal liner material or by bonding the ceramic envelope or both. A circumferential distribution wick is incorporated as a separate structure or as part of the metallic liner. The exterior of the ceramic tube is finned to provide heat transfer area enhancement. Working fluid for the heat pipe in its primary application is sodium or lithium. The prototype heat pipes might also include a central flange to aid in mounting and sealing in the recuperator.

Material Development

Factors in the selection of specific ceramic materials for use on the heat pipe program are: cost, strength, thermal shock resistance, imperviousness, and chemical stability. In addition, the material should be commercially available as tubing, or at least be capable of commercial production. Based on these requirements silicon carbide and alumina were selected for experimental investigation. Among the other materials considered were silicon nitride and mullite ($3\ Al_2O \cdot 2\ SiO^2$). Silicon nitride was not selected for experiment because of availability problems in tubing shapes and because of the materials volatility at temperature. Mullite was eliminated because of concern over its thermal shock resistance. Silicon carbide was considered the primary candidate due to its excellent thermal shock resistance, good thermal conductivity, low volatility, and resistance to both oxidizing and reducing atmospheres. In addition, silicon carbide has an extremely low hydrogen permeability and is well matched to tungsten for expansion coefficient. Alumina was chosen as a second material for experimental investigation despite its comparatively low thermal shock resistance because of its availability and cost, and because it is an excellent coefficient of expansion match to niobium. Alumina was also of interest because of data suggesting that in high purity form it might be compatible with sodium over the lower part of the temperature range of interest.

The development of wick structures for the heat pipes was concentrated on texturing of the metallic liner, where one was to be used, and on the development of an alumina particulate wick for the unlined alumina tubes. Control of the deposition rates in CVD application of tungsten and niobium proved capable of generating surfaces with sufficient surface texture to serve as circumferential distribution wicks for the gravity assisted heat pipes. Figure 4 is a scanning electron microscope photograph of a niobium CVD coating on alumina, showing the surface texture, which is characterized by an average inter-crest dimension of $\sim 25\ \mu m$. Predictions based on a pyramidal model of this type of wick structure indicated that adequate capillary forces could be developed both by the CVD structure and by an alumina particulate wick.

The most difficult problem in the ceramic heat pipe development has been the development of reliable sealing methods for closing the tubes. A variety of high temperature joining techniques have been evaluated for closure, including metal brazes joining the liner to a similar metal layer on the closure plug, active metal brazes joining the ceramic materials directly, and oxide fusion seals.[3] In addition some experimental effort has been directed to development of electron beam weld joining of the metal liners. A listing of braze techniques and materials tested is given in Table II. Development work in this area is continuing.

The chemical compatibility of the metal liner materials with the ceramic tube material has been investigated for the tungsten-silicon carbide combination.[4] The investigations showed that the tungsten-silicon carbide reaction layer growth followed a parabolic law of the form $x^2 = 2\ Kt$, where x is the layer thickness, K the reaction rate constant, and t is time. Predictions based on the measurements give a heat

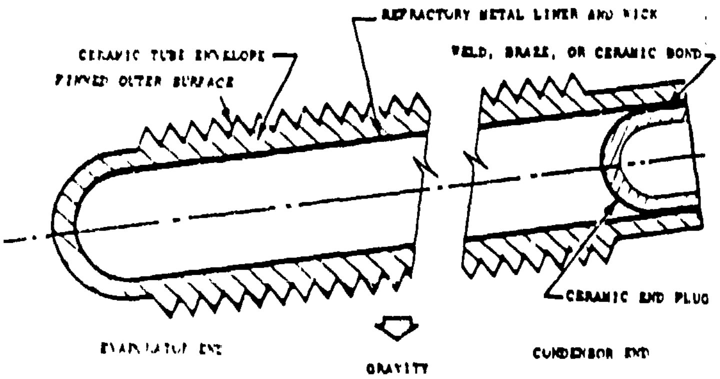

Fig. 3. Ceramic heat pipe configuration.

Fig. 4. Niobium CVD coating on alumina.

TABLE II

BRAZE MATERIAL EVALUATION

Braze Material	Base Material	Braze Temp.	Results
V	Al_2O_3/Nb	2180 K	Leaked, radial microcracks in Nb alloy
V - 39 Nb	Al_2O_3/Nb Al_2O_3/W	2150 K	Leaked, poor braze flow
Ti	SiC/W Al_2O_3/W	1980 K 1980 K	Leaked, but some good bond areas
Ti - 65 V	SiC/W	1940 K	Leaked, but most good bond
Ti - 21V - 25 Cr	Al_2O_3	1875 K	Leaked, braze material bubbled
Ti - 23V - 20 Cr	Al_2O_3	1875 K	Leaked, poor braze flow
Ni	SiC/W	1730 K 1850 K	Leaked, W-Ni alloy formed, alloyed with SiC
Nb - 17 Fe	Al_2O_3	1875 K	Leaked, did not wet Al_2O_3
Y_2O_3	Al_2O_3	2100 K	Leaked, poor bonding
Al_2O_3 - 21 Y_2O_3	Al_2O_3	2100 K	Leaked, but some filleting
Al_2O_3 - 25 Y_2O_3	Al_2O_3	2100 K	Sealed, good fillet
65 Pd - 35 Co	SiC/W	1510 K	Sealed CVD SiC, Pd alloyed with Si, in Si-SiC material

pipe life of more than 20 years at an operating temperature of 1600 K with a tungsten layer thickness of 0.25 mm.

As an alternative to the use of a CVD refractory metal liner fabrication of ceramic heat pipes by vapor deposition of ceramic over free standing molybdenum tube is being investigated. This fabrication method would allow the use of welded tube closures and complete ceramic encapsulation, both advantages permitting an increase in operating temperature over the present configuration, which is limited by the reflow temperature of the braze material and by oxidation of exposed refractory and braze materials. Thermal shock tests have been conducted on tube samples with various layer thicknesses and interface treatments over the temperature range from ambient to 1500 K. These preliminary tests indicate that with the use of an intermediate layer of tungsten high strength amorphous CVD silicon carbide will withstand the thermal stresses induced by the mismatch in expansion coefficients. No evidence of layer separation was disclosed in metallurgical examination after more than 40 thermal test cycles. A photomicrograph of the tube wall section after test is given as Fig. 5. Continuing investigation of this fabrication technique will involve the fabrication of a test heat pipe of molybdenum for coating as a complete assembly.

Experimental Heat Pipe Assembly

Experimental development of SiC and Al_2O_3 heat pipes was conducted in parallel through compatibility tests, wick, and seal development. The Al_2O_3 pipes used CVD niobium liners and lithium as a working fluid. Various braze materials were evaluated and final alumina heat pipe assembly was accomplished using 25% Y_2O_3 - 75% Al_2O_3 reaction bonding of a re-entrant plug closure. Operational test of the Al_2O_3 showed that the alumina material could not withstand the severe thermal stresses developed in startup. Development of the Al_2O_3 pipes was suspended and effort concentrated on the SiC heat pipes.[5]

Heat pipes were fabricated of CVD SiC using a columnar tungsten liner and wick with sodium working fluid. The CVD SiC tubes were nominally 610 mm long by 25 mm in diameter with a wall thickness of 3.0 mm. The tubes were fabricated with an integral closure at one end and lined with CVD tungsten (0.1 to 0.3 mm thick) textured to provide the wick structure. An untapered end closure plug with a radial clearance of 0.15 mm was made from CVD SiC with an external layer of CVD tungsten. The assembly procedure for these pipes began with a vacuum bakeout at 1873 K. The heat pipe was then positioned in a quartz tube vacuum envelope within an RF susceptor. The envelope was evacuated and purged with argon and a capped container holding the sodium charge under a cover gas of argon was positioned on top of the heat pipe within the heat zone.

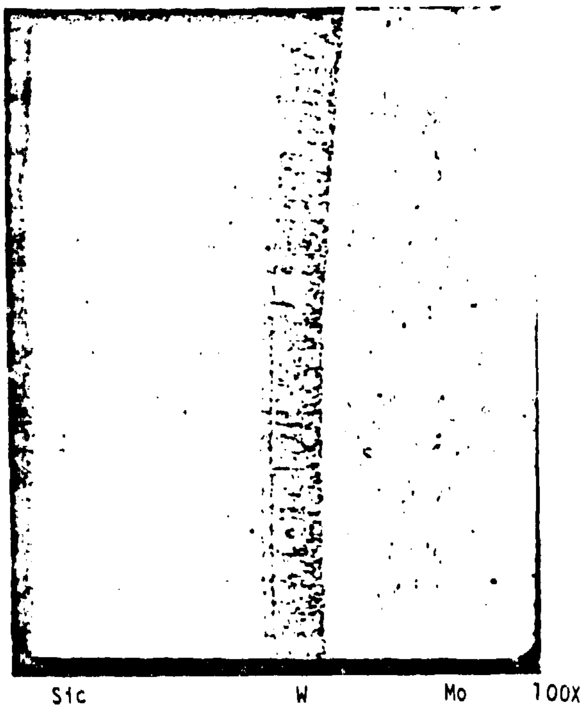

Fig. 5. Cross sectional view of thermal shock test molybdenum tube coated with tungsten and silicon carbide.

The protective caps were then removed from the container and the quartz envelope sealed and evacuated to 10^{-5} torr. The charge container was heated by the r.f. susceptor and the melt allowed to flow into the pipe by gravity. After vacuum degassing of the melt and cooling to ambient temperature, the chamber was backfilled with argon and continuously purged while the working-fluid container was replaced by the ceramic end plug and braze material. Multiple wraps of 0.092 mm tungsten foil were placed around the end plug prior to insertion into the end of the heat pipe. The braze used for the seal was a palladium-cobalt eutectic composition (65% Pd - 35% Co) with a melting point of 1510 K. The braze alloy was in wire form, 0.5 mm diam, placed above the root opening of the joint. After insertion of the end plug, a W-25% Re thermocouple was positioned inside the end plug adjacent to the seal area to monitor the joint temperature. The system was evacuated and the seal area of the heat pipe was heated to 1525 K to melt the braze and form a final closure seal.

To check for leak-tightness and for initial wet-in of the working fluid, the heat pipe was repositioned to place the top of the sodium pool in the heat input zone and operated as a reflux boiler.

Before the ceramic heat pipe is removed from the assembly facility, small leaks in the joint area can be sealed by overcoating or rebrazing with a lower melting point material. Once the heat pipe has been wet-in, the leak is filled with working fluid which forms a vacuum-tight seal when the heat pipe is cooled to ambient. This allows the system to be backfilled with argon and additional braze material placed for resealing at a lower melt temperature.

Prototype Heat Pipe Tests

Thermal testing of ceramic heat pipes is complicated by the nature of the pipe material which prohibits rigid attachment of heaters or calorimeters. In addition, the heat sources and sinks to be used with the pipes must induce no more severe thermal transients than the intended operational environment in order to avoid unrealistic thermal shock failures. Test heat sources must be coupled to the heat pipe through a non-rigid medium such as gas or a fluidized bed or by radiation. In the tests conducted to date this requirement has limited the loading imposed on the heat pipes and consequently the determination of absolute heat pipe throughput limits.

All ceramic heat pipes are initially tested by radiant heating from a r.f. susceptor within the ceramic heat pipe assembly facility. For initial test the heat input is controlled to produce a temperature rise at an average rate of 0.5 K/s until a heat pipe temperature of 1200 K is attained. Maximum heat throughput is 2 kW with loading of the heat pipe by radiation to the surroundings. Temperature measurements of the pipes in the vacuum assembly facility are taken with an optical pyrometer.

The majority of the testing in air has been conducted in a simple gas-fired furnace, shown schematically in Fig. 6. This test set-up has the advantage of providing simulation of the gas environment as well as the temperature at the evaporator end of the pipe. For initial tests in the facility the heat pipe thermal load consists of radiation from the condensor area of the pipe to the laboratory background. The gas gap calorimeter is used subsequently to increase the heat pipe thermal load and provide direct measurement of the pipe output.

In startup and thermal cycling tests, one burner is fired to bring the exhaust gas temperature to approximately 1000 K. The corresponding heat pipe temperature under normal test load is about 800 K. Addition of the second burner provides gas temperatures in excess of 1300 K and heat pipe temperatures to 1900 K. The average rate of temperature rise of the SiC heat pipes during startup is 1.2 K/s up to 800 K and 0.5 K/s thereafter. Heat transport by the heat pipes measured with the calorimeter has been in excess of 1100 W. Test results indicate that the CVD tungsten wick structure provides good distribution of the sodium fluid throughout the heat pipe.

Experimental Data

Heat throughput data has been obtained through the use of a water calorimeter on the condenser section of the heat pipes. The heat pipes are radially centered in the calorimeter to provide a 1 mm gap between the heat pipe and the inner surface of the calorimeter. The primary mode of heat rejection to the calorimeter is by conduction through a gas medium in the gap. Heat rejection rate is controlled by adjusting the

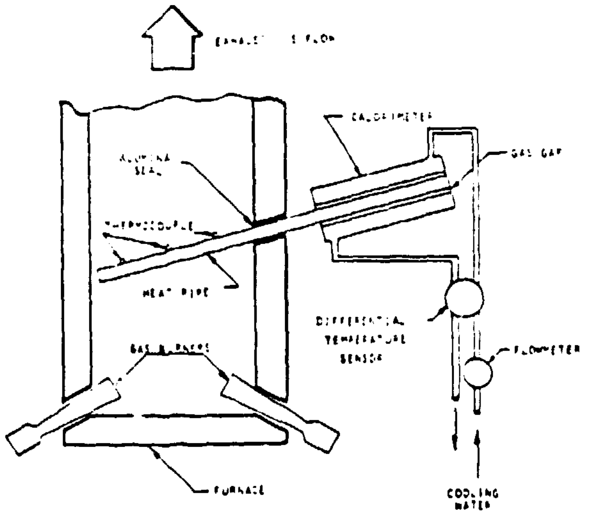

Fig. 6. Ceramic heat pipe test configuration. Tilt angle of the pipe is 11.6°.

ratio of helium to argon gas within the gap, thus varying the thermal conductivity of the gas mixture.

A temperature difference meter and rotameter are used to measure the energy change of the water flowing through calorimeter. The heat pipe temperature is measured by thermocouples attached to the outside wall of the pipe in the heat input region and in the adiabatic region. Thermocouple measurements during test have shown a decrease in temperature at the lower end of the pipe of ~50 K due to the presence of the excess of the 16 gm sodium charge as a pool. This has been true throughout the tests, indicating that a dryout limit for the pipes has not been approached. A thermocouple located the alumina fiber packed gland region in the furnace wall has consistently measured ~10 K below the evaporator temperature, in agreement with the anticipated radial temperature gradients in the pipe wall.

Initial performance testing of the SiC heat pipes has been carried out using a 200 mm heat input zone. Results to date, have demonstrated a maximum heat transfer of 1.96 kW at 1010 K as shown in Fig. 7. The corresponding axial heat flux is 692.0 w/cm^2. The radiation transport limits of the pipes operated without the calorimeter are also shown in the figure. Present performance limitations are established by the temperature and gas velocity limits of the furnace together with the relatively short heat input zone of the heat pipe. Ultimate performance limits of the SiC prototype heat pipe have not been attained as yet.

An SiC heat pipe in test underwent 34 starts from room temperature to temperatures >1200 K and was operated for more than 200 h at temperature. Absolute performance limits for the pipe were not established due to the limited thermal loading capacity of the test set-up. However, test data taken and indicated in Fig. 7 are above the calculated entrainment limit for a wickless, vertical heat pipe. This is most readily explained by the existence of puddle flow of the liquid in the gravity assisted, near horizontal mode of operation. A fluidized bed gas calorimeter is now being assembled in order to achieve higher heat pipe thermal loads for further tests to determine intrinsic limits of the SiC ceramic heat pipes.

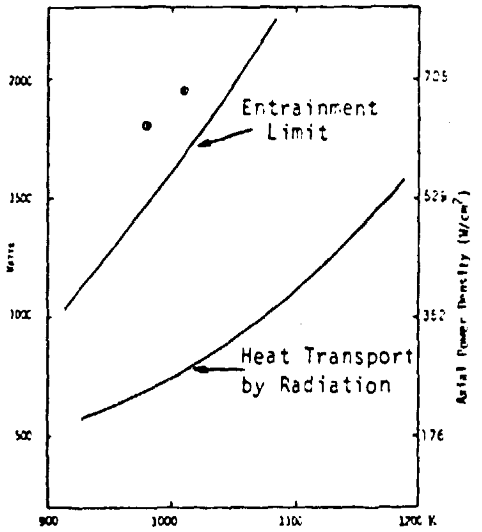

Fig. 7. Performance curves for 25-mm-o.d. SiC heat pipe. Data points represent performance of heat pipe with calorimeter.

Conclusion

A variety of materials and assembly techniques for high temperature ceramic heat pipes have been evaluated. Successful assembly and operation of heat pipes at temperatures over 1200 K has been accomplished using SiC as a shell material and a CVD tungsten inner liner to protect the ceramic from the sodium working fluid. The CVD tungsten has been used as a distribution wick for the heat pipe through the development of a columnar tungsten surface structure, achieved by controlling the deposition rate. The adequacy of this wick structure has been demonstrated in tests at ~ 2 kW throughput with a 25-mm-o.d SiC heat pipe. Operation of ceramic heat pipes of this configuration in repeated start cycle tests have demonstrated their ability to withstand the thermal transient stresses of startup at temperature rise rates ~ 1.2 K/s.

These demonstrated temperature and heat flux rates are in the range of the values predicted for operational use in high temperature industrial recuperators. Further effort on the program will be directed toward the demonstration of ceramic heat pipes in a subscale recuperator as well as to the continuing development of ceramic heat pipe technology.

Acknowledgements

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