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HEAT PIPE DEVELOPMENT FOR HIGH TEMPERATURE RECUPERATOR APPLICATION

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ABSTRACT

Heat pipes have been developed for operation in oxidizing atmospheres at temperatures above 1100 K. The heat pipes comprise a metallic liner and wick structure with a protective outer shell of an oxidation resistant material. The working fluids used in the heat pipes are alkali metals. A number of configurations have been evaluated, ranging from pipes using a metallic inner liner of a chemically vapor deposited (CVD) refractory metal applied to ceramic tubing, to one utilizing ferrous materials with an outer layer of a developed oxide. A promising intermediate configuration consisting of free-standing refractory tubing covered with a layered structure of fine grain, equi-axed CVD silicon carbide has also been evaluated. The test heat pipe was fabricated using low-carbon, arc-cast molybdenum tubing and a wick composed of 150 mesh molybdenum screen. Hafnium gettering was used with sodium working fluid. Assembly of the pipe was by electron beam welding. Following closure and capping of the fill tube the assembly was operated in a vacuum for several hours prior to the chemical vapor deposition of the exterior ceramic coating. After coating, the pipe was operated in air and in combustion gases for performance evaluation.

The use of iron-chromium-aluminum alloys as container materials for operating in high temperature oxidizing and sulfiding gas streams has been investigated. Alloys of this type develop heavy, protective oxide surface layers when exposed to high temperature oxidizing atmospheres, and are commonly used in electrical heating elements because of their exceptional oxidation resistance. A heat pipe was assembled from Kanthal A-1 seam welded tubing in order to establish limits of operation for these materials. The heat pipe employed a 150 by 150 mesh 316 stainless steel (SS) screen wick and hafnium foils for oxygen gettering with sodium used as the working fluid. This heat pipe has been tested in a fossil fired flame above 1200 K to determine its operating characteristics and the durability of the material.

KEYWORDS

Heat pipes, recuperation, ceramic heat exchangers, molybdenum fabrication, alumina, silicon carbide.

INTRODUCTION

Increased interest in energy conversion through high temperature recuperation has stimulated investigation of heat pipes capable of operating at temperatures of 1100 to 1500 K. Los Alamos National Laboratory is conducting a program for the Pittsburgh Energy Technology Center of the Department of Energy which has as its objective the development and demonstration of heat pipes capable of operating at these temperatures and conditions. The principal focus of the effort is the development of heat pipes based on ceramic materials for use in the upper end of this temperature range, however lower temperature application of ceramics and intermediate temperature use of oxide-protected metal surfaces has also been investigated.

BACKGROUND

The development of heat pipes for a specific temperature range entails the selection of appropriate envelope materials, wick structures, and operating fluids. For flue gas heat recuperation at temperatures above 1100 K the principal external envelope materials of interest are the full density, high strength ceramics having reasonable thermal conductivity and good temperature shock characteristics. Table 1 gives the pertinent physical characteristics for some of the ceramic materials considered for this application.

Table 1 Ceramic Materials for High Temperature Recuperators

	SiC Si Bonded	SiC Solid Alpha	SiC CVD	Si_3N_4 Reaction Bonded	Al_2O_3	SiC C Composite
Failure strength Kgf/cm² at 1360 K	3164	3516	3164	2460	[illegible]	3164
Thermal expansion, cm/cm/K	4.7	4.5	5.7	3.1	1.3	5.4 to 9.0
Thermal conductivity at 1360 K cal/s-cm-K	8.77×10^{-2}	4.55×10^{-2}	4.11×10^{-2}	2.48×10^{-2}	9.01×10^{-3}	[illegible]
Max. use temp. for extended time, K	1600	1600	2000	1800	2000	1600 to 2000
Fabricability	Good	Good	Good	Good	Good	Good
Density, g/cm³	3.1	3.2	3.2	2.8	3.9	2.5 to 3.5

These ceramics may be used as basic structural materials or as coatings. In addition oxide protected metallic materials may be useable for some high temperature applications, although the nonrefractory metallics will be limited by high temperature strength and creep resistance even when protected by ceramic coatings. However, in most recuperator designs a range of material surface temperatures will be encountered in indentifiable zones and the lower temperature metallic materials will be satisfactory for some parts of the design (Merrigan, 1980, 1981; Strumpf and Miller, 1980). Therefore investigation has been conducted on the use of nonrefractory metal tubing for high temperature heat pipes with a surface barrier coating established for oxidation and sulfidation protection. This configuration is limited in temperature range to somewhat lower levels than the ceramic envelope configurations, but is of interest for transition sections of counterflow recuperators between high temperature ceramic surfaces and conventional metallic heat exchanger materials.

Normal operating fluids for the temperature range of interest are the alkali metals. The entire range from 1100 K to 2000 K may be covered by sodium and lithium. Potassium may be used to extend the operating range to lower temperatures. These alkali metal working fluids may attack ceramic materials at high temperatures so compatibility tests were conducted with silicon carbide and lithium, and with high purity alumina and sodium to determine the magnitude of the problem (Ranken and Lundberg, 1978). These tests indicated that for silicon carbide, a metallic liner was necessary to protect the ceramic from attack by the operating fluid. This requirement has led to the investigation of various means of providing a layered pipe wall structure having an outer layer of an oxidation resistant ceramic and an inner layer of alkaline metal resistant refractory metal. The methods investigated have included the use of chemical vapor deposition of a layer of tungsten on the inner surface of ceramic tubing, the development of free-standing shells of tungsten by the CVD process that were then overcoated with a chemically vapor deposited silicon carbide, and the use of drawn tubing of molybdenum as a substrate for a chemically vapor deposited layer of silicon carbide. In addition the use of high purity alumina without a metallic layer and with sodium as a working fluid has been investigated.

The need for a metallic liner raises questions with regard to chemical reactions at the metal-ceramic interface and compatibility of the physical characteristics of both the metal and ceramic materials and of their reaction products. Coefficients of thermal expansion must be well matched between the refractory metal liner and the ceramic envelope to prevent deterioration of the bond and consequent increase in the thermal resistance of the tube wall. Investigations conducted on the program have consequently included reaction rate modeling for tungsten-silicon carbide reactions and verification of the apparent reaction constants in experiments (Lundberg, 1979, 1980). These experiments have indicated that tungsten layers of about 0.25 mm thickness will provide a life of about 20 years at normal operating temperatures.

HEAT PIPE EXPERIMENTS

CVD Silicon Carbide Heat Pipes

The first successful operation of a high temperature ceramic heat pipe was achieved using chemical vapor deposit (CVD) silicon carbide as a container material and CVD tungsten as a protective liner (Keddy and Rankin, 1979). Controlled deposition of the tungsten was used to give a textured surface for wicking. The working fluid used was sodium with the heat pipe closure accomplished by a tungsten-to-tungsten braze using a palladium-cobalt brazing alloy. These heat pipes were operated in air and in combustion gases at temperatures to about 1200 K. Peak measured heat transfer through a 2 cm diameter heat pipe was about 2 kilowatts with operational limits estimated to be higher. After about 100 hours of operation and 30 cold starts one of these pipes was sectioned and the tungsten-silicon carbide interface examined for evidence of deterioration or reaction. The reaction zone thickness was found to be in agreement with predictive models developed on the program. However, some evidence of liner-shell bond deterioration was observed.

In order to reduce stress at the interface between the tungsten and silicon carbide a configuration was tried in which an intermediate layer of graphite was used. The low modulus of elasticity of the graphite provided accommodation between the tungsten and silicon carbide. This design was based on similar construction that has been used for flame heated thermionic diode shells with good success (Balestra, Miskolczy, and Wang, 1976). Four graphite cores were prepared from silicon substrate grade graphite bar stock by gun drilling. These were coated on the exterior with CVD silicon carbide and then with CVD tungsten on the inner surface. Radiographic inspection of the finished tube prior to assembly disclosed radial cracks in the tungsten liners throughout the tubes. This cracking was attributed to property variation in the graphite along the length of the tube. In order to achieve more uniform structure in the core tube an investigation of the use of extruded silicon carbide-carbon composite tubing was undertaken and is continuing.

Alumina Heat Pipes

Use of alumina as a container material for high temperature heat pipes is attractive from the standpoint of availability, cost, and sodium compatibility. Therefore, the initial investigations of ceramic materials for the program were concentrated on alumina. Both CVD niobium lined alumina tube configurations and metallized alumina tubes using a low density slurry coated inner structure for wicking were tried. None of these early attempts were operated as heat pipes successfully. The low thermal shock resistance of the alumina caused failure of sodium heat pipes during startup. The use of alumina was therefore abandoned despite its advantages and despite development of a good high temperature sealing technique using a Y_2O_3-Al_2O_3 eutectic braze. However, later inquiries as to potential application of ceramic heat pipes in the wet acid environments characteristic of sulfur dioxide scrubbers prompted the assembly of an alumina heat pipe using toluene as a working fluid and intended for operation at temperatures in the 350 to 420 K range.

Two prototype pipes were assembled using a 25 mm o.d. by 3 mm wall by 610 mm long, closed end, aluminum oxide tube and a fitted alumina plug. The tube was lined with 2 layers of 100 mesh 304 stainless steel screen held against the inside wall with a 302 stainless steel coil spring. The aluminum oxide plug was tapered to match a 5^{o} taper in the tube for a glass seal joint. The plug was fitted with a 5 mm Kovar fill tube which was vacuum brazed to the aluminum oxide with a 71.15 silver-28.1 copper-0.75 nickel alloy braze.

The resulting heat pipe configuration is shown in Fig. 1. This heat pipe was operated initially in a hot oil bath for performance verification. Following these performance checks the pipe was operated using an electric resistance furnace for heat input and a gas gap calorimeter for loading. Performance taken in these tests is given in Fig. 2. This pipe has been operated intermittently over a period of about a year with no signs of deterioration. It appears that the alumina envelope is satisfactory under the lesser thermal shock conditions encountered with an organic working fluid and reduced temperatures.

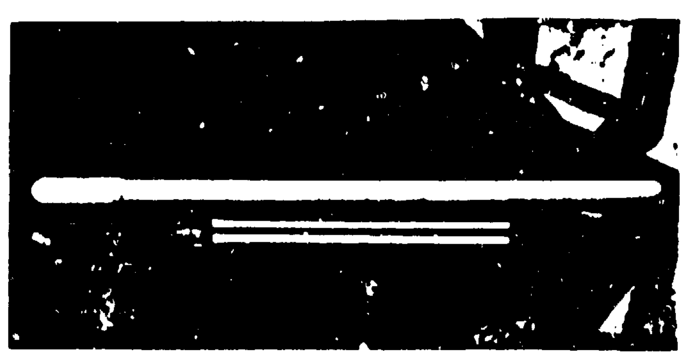


Fig. 1. Alumina-toluene heat pipe.

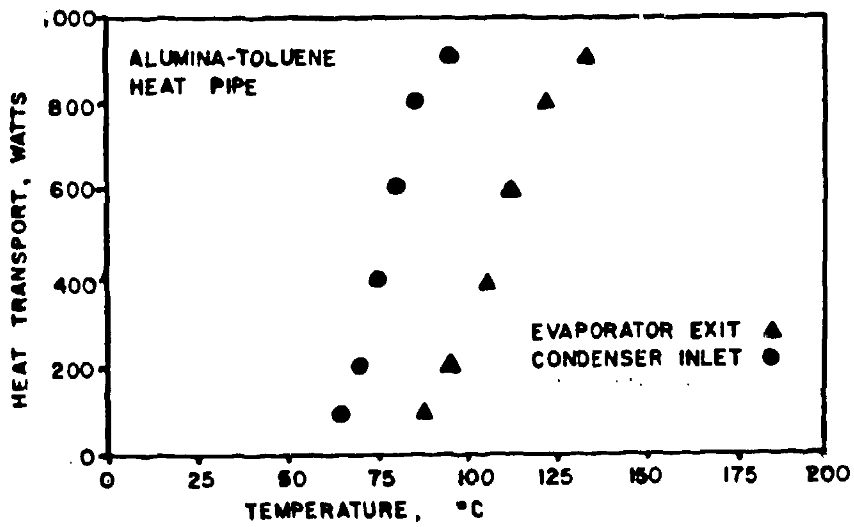


Fig. 2. Performance data for alumina heat pipe using toluene as a working fluid.

Molybdenum Core Heat Pipes

An alternative to the deposition of a protective metallic layer on the inner surface of ceramic tubing for alkaline metal heat pipes is to start with a refractory metal tube and deposit a ceramic coating on the outer surface (Merrigan and Keddy, 1980). This method of assembly permits complete filling and capping of the heat pipe before the ceramic coating process and therefore can be used to produce a heat pipe that is completely ceramic incapsulated. By contrast if a ceramic tube is closed by brazing or welding the materials used in the closure process must be compatible with the operating environment or must be protected by a separate corrosion resistant cover. Molybdenum was used as a core material for this configuration because of its availability as seamless tube and because of familiarity with molybdenum fabrication techniques developed in other Los Alamos programs (Lundberg and Martinez, 1980). Molybdenum tube samples 1.6 cm diameter by 15 cm length with 0.05 cm wall thickness were fabricated from low carbon arc cast tubing material and coated with fine grain, equiaxed CVD silicon carbide in thicknesses ranging from 0.1 mm to 0.5 mm. An intermediate layer of 0.1 mm tungsten was used on some of the specimens to reduce formation of molybdenum silicide at the interface. Five tube samples were coated. The thickest coating layer failed in cooldown from the approximately 1400 K coating temperature. In failure the coating fractured into pieces about 0.5 mm by 0.05 mm in size with the longer dimension along the axis of the tube. The failure removed most of the coating from the tube. The remainder of the tubes were placed in a vacuum furnace and cycled from 300 K to 1600 K at 40 minutes per cycle. The two of the five tubes having intermediate tungsten layers survived six cycles in the vacuum furnace and were then moved to a gas furnace where they were directly exposed to the gas flame which was cycled off and on at 30 minute intervals. After 30 cycles the tubes were removed and sectioned for metallographic analysis. No evidence of interlaminar separation was found and reaction zones at the interfaces were minimal. A molybdenum heat pipe was fabricated using the same tubing with 150 mesh molybdenum screen wick, molybdenum end caps and electron beam welding for assembly. Hafnium washers were used for oxygen gettering in the pipe. This heat pipe was filled with 11 grams of sodium, the fill tube pinched off, and a cover cap welded in place. After operation to verify performance, the heat pipe was coated with a layer of 0.1 mm CVD tungsten, followed by approximately 0.25 mm of equiaxed CVD silicon carbide (Fig. 3). Inspection after coating showed a single coating flaw in the area of the hemispherical end cap on the end of the tube opposite the fill tube. The heat pipe was placed in test despite the flaw and operated with a calorimeter to obtain performance data and then with radiation loading for about 150 hours. No general deterioration of the coating was observed during this time but the end cap flaw grew to a pit approximately 1.5 mm diam and 2.0 mm deep through oxidation of the molybdenum substrate. At 150 hours the test was terminated rather than risk venting of the heat pipe. Temperature during the extended period of the test was about 1300 K.

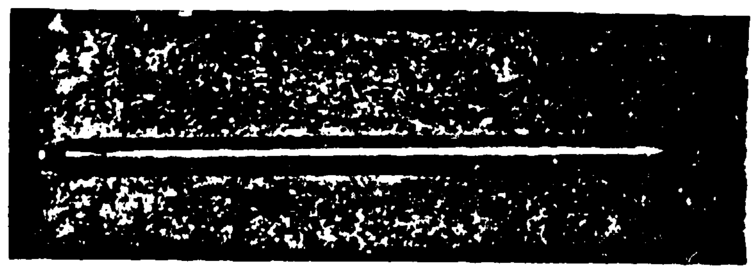


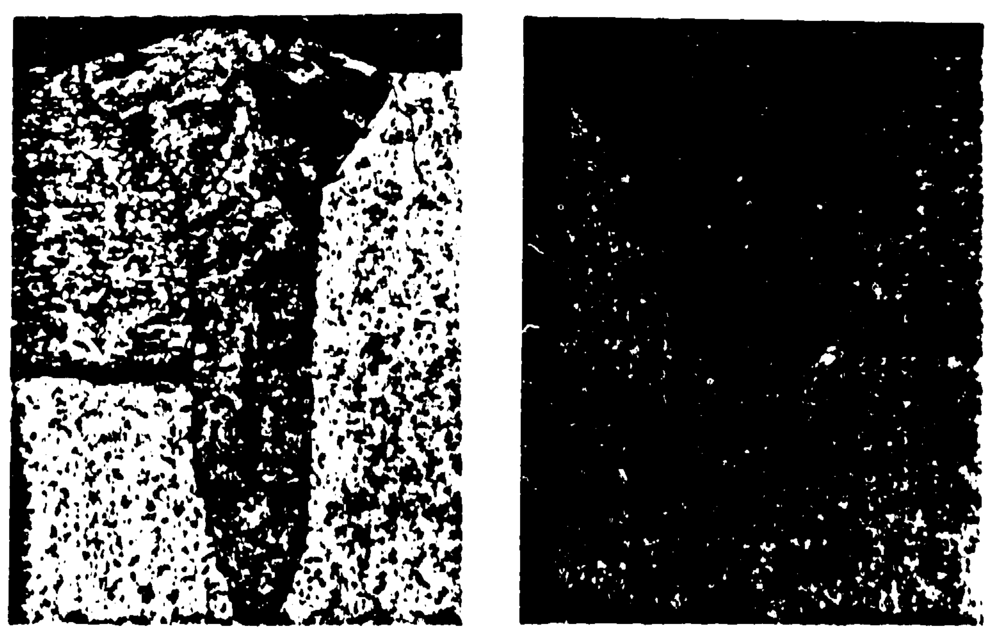
Fig. 3. Molybdenum core heat pipe after coating with silicon carbide.

This method of fabrication of ceramic heat pipes is more expensive than the alternative use of coated ceramic tubing, however, the experience suggests that with some improvement in coating technique this method could be used to produce heat pipes capable of extended operation at about 1500 K.

High Temperature Metallic Heat Pipes

Oxidation resistant metallic surfaces have been investigated for use in intermediate temperature ranges where solid ceramic heat exchange elements may not be necessary. For example, in counterflow heat pipe heat exchangers surface temperatures of the tube rows near the low temperature side may be cool enough under normal operating conditions to use metallic tubes even though the remainder of the recuperator requires ceramic elements (Merrigan, 1980). In connection with the ceramic heat pipe program a review and investigation of metallic tube materials for use in high temperature heat exchangers was initiated. Initial review suggested the use of iron-chromium-aluminum alloys as container materials for operation in oxidizing and sulfiding gas streams. These alloys develop heavy, protective oxide surface layers when exposed to high-temperature oxidizing atmospheres, and are commonly used in high-temperature electrical heating elements because of their exceptional oxidation resistance. Varieties having more than 22% chromium and about 5% aluminum have been found to have excellent resistance to atmospheres containing sulfur compounds.

As a test of the concept a heat pipe was assembled from Kanthal A-1 seam welded tubing. Fecralloy plate material was used for the pipe end caps. Kanthal A-1 is an iron-base alloy containing 22% chromium, 5.7% aluminum and 0.5% cobalt, and is produced by AB Kanthal in Sweden. The manufacturer claims this alloy can be used successfully in an oxidizing atmosphere up to 1625 K. Fecralloy is also an iron-base alloy containing 16% chromium, 5% aluminum and 0.3% yttrium. This alloy is produced by Allegheny Ludlum Steel Corp. in Pennsylvania. Because small diameter tubing was not available in these materials, 4130 steel tubing was used for the fill tube. Joining of the fecralloy end caps to the Kanthal tube was attempted by electron beam welding without a filler metal using a variety of weld parameters. The penetration and metal flow characteristics of the test joints were good and no problems were encountered in achieving consistently acceptable weld beads. All of the weld samples were leak tight when measured on a 10^{-6} scale. However, metallographic examination indicated a problem with fusion zone hot cracking. Figure 4, a conventional electron beam weld, shows the extensive solidification cracking encountered in high restraint zones. A defocussed beam was used to eliminate the centerline root cracking, but as indicated in Fig. 5, significant interdendritic solidification cracking remained. Gas shielded tungsten electrode welding (TIG) was therefore used in initial assembly for end cap and fill tube attachment. The resulting assembly was leak checked satisfactorily, filled with sodium and operated at low power for wet-in. The fill tube was then pinched off and prepared for a cover tube. A leak was discovered in the heat pipe after machining for cover tube attachment. This was caused by machining in the weld area. A thicker fecralloy end plate was fabricated and the pipe reassembled, using TIG welding for the cover tube attachment. TIG welding is used in fabrication of the Kanthal seam welded tubing with resulting microstructure shown in Fig. 6. No problems have been encountered with this weld method as yet in tests.



Figs. 4 and 5. Metallographic sections showing fusion zone cracking in electron beam welds of fecralloy/Kanthal joint combination.

The heat pipe used 150 by 150 mesh 316 SS screen wick and hafnium foils for oxygen gettering. Sodium was used as a working fluid. The heat pipe was wet in at 1173 K and operated briefly at 1273 K in air using an radio frequency induction heat source as shown in Fig. 7. These tests were run before installing the cover tube and repeated on the reassembled pipe.

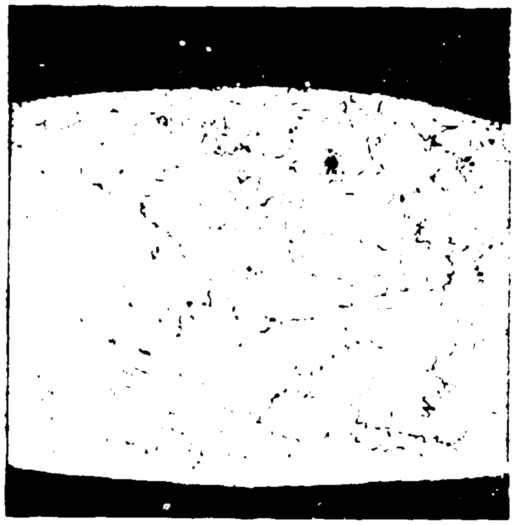


Fig. 6. TIG seam weld in Kanthal tube.

Fig. 7. Kanthal heat pipe performance verification test.

After initial operational verification the heat pipe was conditioned in air at 1321 K in order to develop a uniform oxide layer. The heat pipe was then performance tested in a natural gas furnace. The gas flame impinged directly on the evaporator of the heat pipe, and the condenser was fitted with a gas-gap calorimeter for output measurement as shown in Fig. 8. Performance of the pipe is given in Fig. 9. The heat throughput in these tests was limited by the capacity of the calorimeter and absolute performance limits for the pipe were not determined. After performance testing the Kanthal heat pipe was installed in a natural gas fired life test set-up as shown in Fig. 10. The heat pipe is mounted in a firebrick furnace with half of its length heated by the hot combustion products and the other half air cooled by convection. The combustion air is preheated by passing over the heat pipe's condenser. Chromel-Alumel thermocouples are spot-welded to the heat pipe along its length with the heat pipe temperatures monitored and recorded periodically during operation.

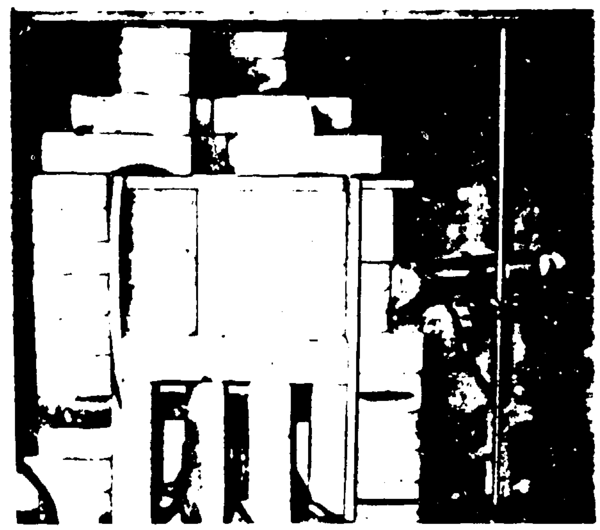


Fig. 8. Kanthal heat pipe calorimeter test.

The pipe is operated each working day in this furnace at about 1200 K with a total of about 100 hours operating time accumulated as of this writing. The surface condition of the pipe shows no sign of deterioration as of 100 hours. A typical temperature profile for the pipe in life test is given in Fig. 11 for thermocouple spacing of 5.7 cm.

CURRENT CERAMIC HEAT PIPE DEVELOPMENT

Current baseline configuration for ceramic heat pipe development employs a multi-layer structure with a core of extruded silicon-carbide-carbon composite material (Merrigan and Sandstrom, 1981). Figure 12 illustrates the extrusion process while Fig. 13 shows the microstructure of a silicon carbide-carbon composite with CVD silicon carbide surface coating. Optimization of the properties of the extruded material is continuing with an investigation of the use of sintering aids to improve density. The inner surface of this composite tube is coated with CVD tungsten and the outer surface with silicon carbide as shown in Fig. 14. Surface structure of the silicon carbide is shown in Fig. 15. Further development of the tungsten deposition process is intended to provide a graded structure with fine grain material at the core interface and more coarse grain structure at the free surface for wicking.

Closure and sealing of this heat pipe configuration is by means of an electron beam weld between the tungsten liner and a formed tungsten end cap. Two means of providing oxidation protection for the tungsten are available. The tungsten weld may be made prior to the final CVD silicon carbide deposition and the entire surface coated in this final step or a protective end cap of the composite material may be bonded over the tungsten cap using a ceramic or glass sealant and the final CVD silicon carbide coating applied over the composite envelope.

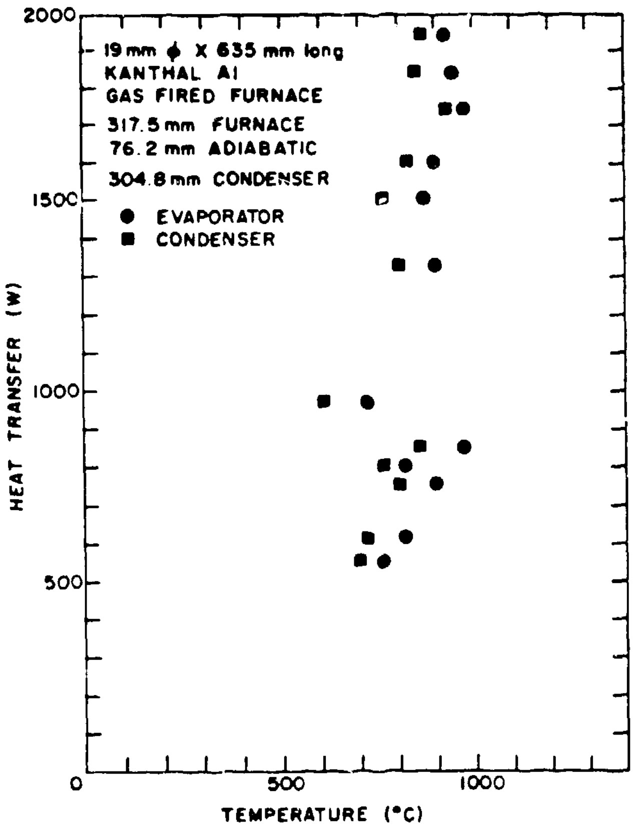


Fig. 9. Performance data for Kanthal heat pipe.

Fig. 10. Kanthal heat pipe life test.

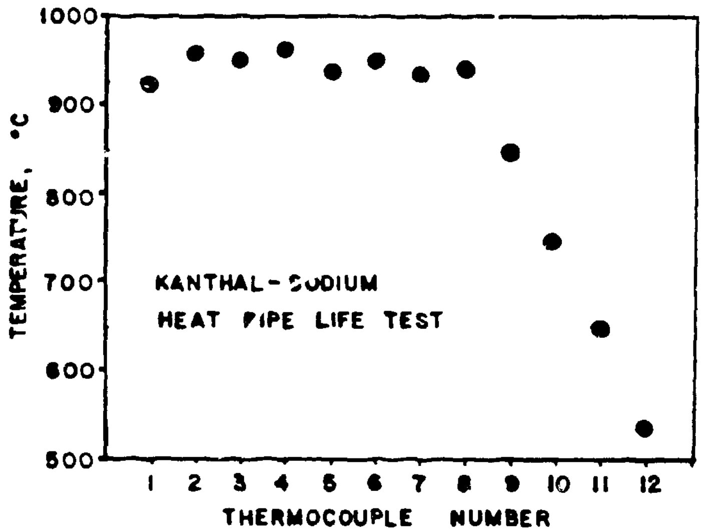


Fig. 11. Kanthal heat pipe life temperature profile. Thermocouple spacing is 5.7 mm.

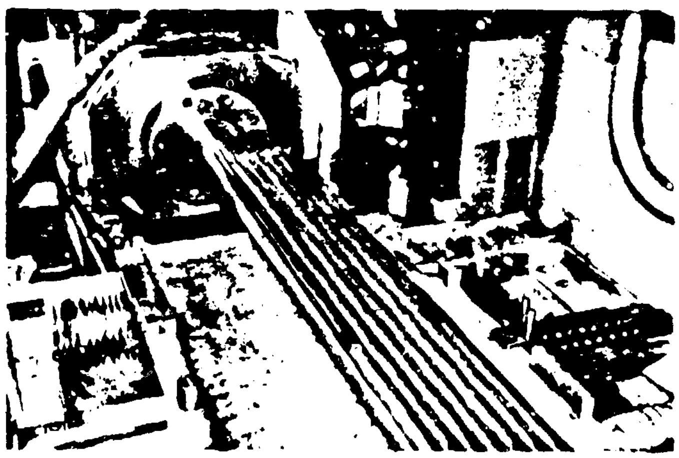


Fig. 12. Silicon carbide-carbon extrusion process.

CONCLUSIONS

Heat pipes capable of operating in oxidizing atmospheres at temperatures in the range of 1100 K to 1500 K have been assembled from solid ceramic materials, ceramic coated refractory metals, and oxide protected non-refractory metals. Ceramic heat pipe fabrication technology has been applied to lower temperature operation in wet acid environments using alumina and is potentially useful in a

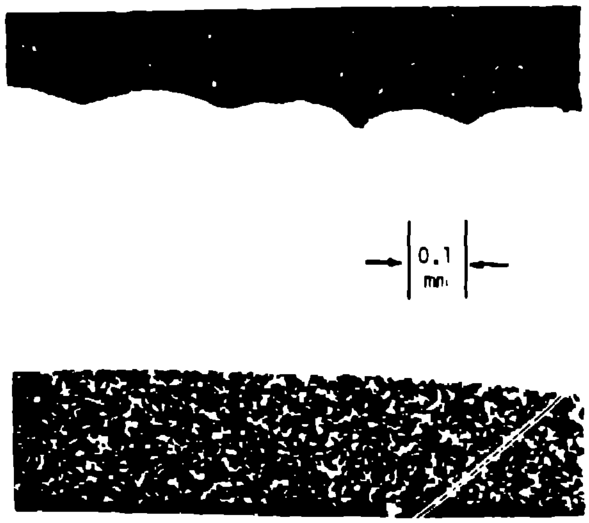


Fig. 13. Silicon carbide-carbon composite with CVD silicon carbide coating.

Fig. 14. Cross-section of SiC/C tube liner and SiC coating 13.0 mm o.d.

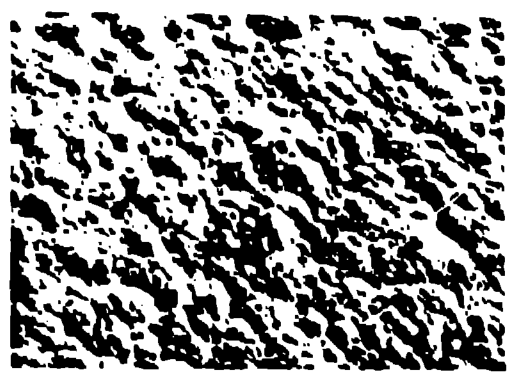


Fig. 15. SEM of CVD SiC Surface.

variety of other applications involving erosive or corrosive environments. Alumina has been demonstrated to be an adequate ceramic heat pipe material for use with organic working fluids although lacking the necessary thermal shock resistance for use with liquid metals. Current high temperature ceramic heat pipe development is based on the use of CVD tungsten and silicon carbide coatings on an extruded silicon carbide-carbon composite core.

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