

Heat-Pipe Oven: A New, Well-Defined Metal Vapor Device for Spectroscopic Measurements*

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A new, well-defined metal vapor device called the heat-pipe oven has been developed on the basis of the heat pipe, a heat conductive element designed by Grover and his co-workers in Los Alamos. It continuously generates homogeneous vapors of well-defined temperature, pressure, and optical path length. The vapor is confined by inert gas boundaries which remove the window problem and allow a direct pressure measurement without relying on vapor pressure curves. Due to the continuous evaporation and condensation the vapor purifies itself during operation.

INTRODUCTION

In many, mainly spectroscopic applications, it is important to generate well-defined homogeneous metal vapors, for which the density and temperature and, for absolute calibrations, also the total number of particles are well known. This has been done in the past mainly by using furnaces of different types, atomic beam devices, hot cathode diodes, burner systems, or other similar instruments. All these devices have suffered from a number of serious problems and considerable effort has been spent to overcome these difficulties.

First of all, it is essential to establish a homogeneous temperature and density distribution which is constant over a sufficiently long period of time. For this purpose sophisticated arrangements of radiation shields and heat baffles have been used.¹⁻³

The particle density n is in most cases determined from the well-known ideal gas relation,

$$p = nkT, \quad (1)$$

which is valid if the typical range of interaction between the particles is much smaller than the interparticle distance. The partial vapor pressure p inside the system is usually determined by the temperature of the coldest spot of the whole system. Since vapor pressure curves are often poorly known it is always preferable to measure the pressure directly. Even where the vapor pressure curves are sufficiently well known the actual pressure may still be appreciably different due to chemical reactions. For this reason, in atomic beam experiments, the total number of absorbing atoms is measured through the material deposited on an extremely sensitive electrobalance^{4,5} which, however, requires very

careful calibration. In other cases, where temperatures are sufficiently high to generate thermal metal vapor plasmas with high enough electron densities, for example, Cs plasmas at 2500°C, line broadening data are evaluated to determine the densities of interest.

Another very serious and common difficulty is the window problem because, especially at high temperatures, a large number of vaporized species are highly reactive. For this reason baffles with small apertures are often used and the vapor is kept away from the windows by means of traps or by differential pumping. Even when the vapor does not react with the window material, the windows usually have to be kept at a slightly higher temperature than the rest of the system in order to prevent the vapor from condensing on the window and making it opaque. Either technique will introduce uncertainties in determining the total number of particles because of the poorly defined boundary layers and may give rise to radiative transfer problems.

The device described in the following is able to remove the difficulties indicated. It is based on a heat conductive element initially developed by Grover, Cotter, and Erickson,⁶ which they call the "heat pipe" (see also Eastman⁷). For this reason we named our new metal vapor device the "heat-pipe oven." The heat pipe uses the well-known fact that large amounts of heat can be transferred if one evaporates a liquid, transports the vapor through a duct and condenses it again. In order to operate such a device continuously it is necessary to return the condensate back to the evaporator. This has been achieved in case of the heat pipe in a very elegant way by using the effect of surface tension.

The heat pipe consists of a closed tubing, the inner wall of which is covered by a capillary structure acting as a wick. This wick is saturated with a wetting liquid. During operation the liquid evaporates at one end due to an external heat source. By providing a heat sink at the other end the vapor is driven down the tube and condenses again. The condensate is then returned by the capillary forces through the wick to the evaporator. In this manner heat conductivities have been achieved

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⁶ G. M. Grover, T. P. Cotter, and G. F. Erickson, *J. Appl. Phys.* 35, 1990 (1964).

⁷ Y. Eastman, *Sci. Amer.* 218, 38 (1968).

which are orders of magnitude larger than the ordinary thermal conductivity through metal rods of comparable size. Furthermore, the heat pipe has been proven to operate under gravity-free conditions.

DESCRIPTION OF THE HEAT-PIPE OVEN

In order to make the vapor inside the heat pipe amenable to spectroscopic or other measurements the pipe has to be opened. This was done in a way similar to that used by Bohdanský and Schins,^{8,9} who connected one end of the heat pipe through a smaller diameter tubing to an argon pressure vessel. At pressures above 50–100 Torr depending on the test material they were able to separate the gas from the vapor because the vapor stream drives the inert gas to the end of the heat pipe. This experiment provides a simple and accurate method for measuring vapor pressure curves above the indicated pressure limit.

The design of a heat-pipe oven which was developed primarily for spectroscopic measurements is shown in Fig. 1. It consists of a tube the central portion of which is designed like a heat pipe with a capillary structure on the inner surface made, for example, of several layers of woven mesh. Both ends are closed by a demountable unit which connects the tube to a vacuum system and holds the windows. The tube is heated in the middle by an induction heater which is surrounded by a vacuum chamber in order to prevent the outer surface of the pipe from oxidizing. The vacuum chamber is sealed against the tube by means of O-rings. The O-ring seals are cooled by the water in the cooling chamber and allow for any thermal expansion of the pipe. The whole arrangement is completely demountable so that the actual pipe can be easily replaced by another one without replacing any other part of the system.

In order to operate this device, the pipe is first of all filled with an inert gas at a suitable pressure. Heating up the central portion of the tube, the sample of metal will melt down and wet the wick. Depending on the pressure of the inert gas, which initially fills the whole tube, the metal then evaporates at a temperature given by the vapor pressure curve of the metal, for which the vapor pressure equals or just exceeds the inert gas pressure. This causes the vapor to diffuse towards both ends until it condenses again. The condensate returns through the wick back to the heated portion of the tube by capillary action. Finally an equilibrium is reached, in which the center part of the pipe is filled with the metal vapor at a pressure determined by the confining inert gas at both ends of the pipe. Owing to the pumping action of the flowing vapor the inert gas is completely separated from the vapor except for a short transition region whose thickness depends on the pressure and the

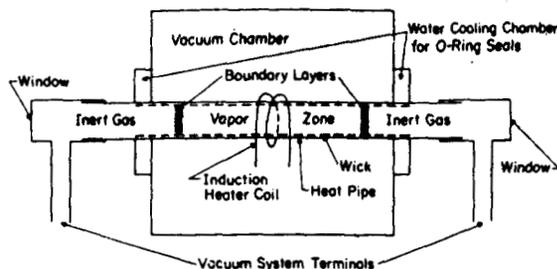


FIG. 1. Schematic arrangement of the heat-pipe oven.

type of vapor and inert gas used. In order to keep this boundary layer sufficiently short, typical operation pressures are about 10 Torr and higher. The length of the metal vapor section depends almost entirely on the power input and the gas pressure, which determines the vapor temperature. At typical temperatures of about 600°C and higher the power is predominantly removed by radiation from the tube wall. For this reason the vacuum chamber is water cooled.

Our first metal vapor device was operated with lithium which seemed to be most promising because of its very high heat of vaporization, its high surface tension and low density. The stainless-steel tube (which is compatible with lithium) has a diameter of 1 in. and a wall thickness of 1 mm. The wick is made of 4 layers of stainless-steel mesh extending out to the water cooling chambers which cool the O-ring seals. The pipe has been run successfully for several days without any detectable loss of lithium and without any deposit on the end windows. With a 34-cm-long vacuum chamber the metal vapor region in the tube has been extended to a length of 25 cm. In order to observe the temperature profile along the tubing the vacuum chamber has a narrow window along its side covering the whole length; this allows temperature measurements by means of a pyrometer. A shutter inside this window substantially reduces sputtering from the outer surface of the tubing onto the window. The temperature of the central vapor zone depends only on the inert gas pressure and was observed to follow the vapor-pressure curve within the error limits. Although the heat-pipe oven is open on both ends its operation was completely stable without any measurable shift of the metal vapor zone as demonstrated in detail below (Fig. 2).

PROPERTIES OF THE HEAT-PIPE OVEN

The heat-pipe oven offers a number of significant advantages over previous devices.

The inert gas between the windows and the vapor zone removes the window problem which, especially at high temperatures, has been a serious problem for all highly reactive species such as, for example, barium, lithium, and a large number of other elements of interest. The inert gas sections also solve radiative transfer

⁸ J. Bohdanský and H. E. J. Schins, *J. Appl. Phys.* **36**, 3683 (1965).

⁹ J. Bohdanský and H. E. J. Schins, *J. Phys. Chem.* **71**, 215 (1967).

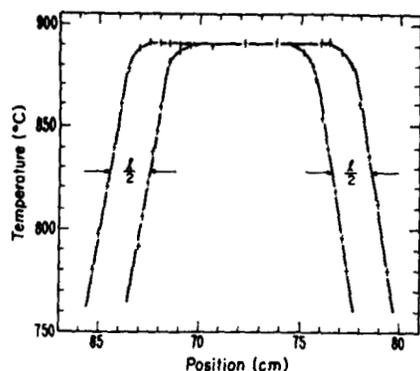


FIG. 2. Temperature profiles measured at two different powers with a heat-pipe oven operating with lithium and using helium as the confining gas at a pressure of 10.9 Torr.

problems which frequently arise due to poorly defined vapor zones as indicated in the introduction.

The metal vapor is very clean because the continuous evaporation and condensation will purify the vapor after some operation time. At a particular temperature the vapor pressure of the impurities will usually be different from the vapor pressure of the dominating species of interest. Those impurities which have a higher vapor pressure have a low enough concentration so that the generated vapors are not saturated. Otherwise these impurities would determine the characteristics of the heat pipe. Therefore, since the vapors are not in equilibrium with the liquid phase they will not condense again but are pumped out to colder parts of the tube where they may condense or stay in the gaseous phase. Those components which stay in the gaseous phase can be pumped out after cooling down the heat pipe. Impurities with a lower vapor pressure than the dominating vapor will either have such a low vapor pressure that they have no effect on, for example, spectroscopic measurements or again the vapors cannot be saturated, in which case they are pumped out as explained before. Therefore, in order to take full advantage of the self-cleaning effect of the heat pipe oven one has to start out with a sufficiently clean sample so that none of the impurity vapors can be saturated and condense again in which case they would get back through the wick to the evaporator.

A further crucial advantage is the fact that the heat pipe oven can be operated continuously under extremely well-known conditions. Since evaporation and condensation take place almost at the same temperature, the temperature will be constant to within at most 1°C over the whole metal vapor zone⁸ without using any radiation shields or heat baffles. Only around the short boundary layers does the temperature drop rapidly as shown in detail below (Fig. 2). A suitable thermocouple gives the temperature of the metal vapor section easily to 1°C accuracy.

The pressure can be determined with high accuracy directly from the inert gas pressure. Pressures of about

10 Torr and higher are in a convenient range where, for example, a simple oil manometer will easily give the pressure to within 1% accuracy. There will be a small pressure gradient within the vapor section in order to establish the vapor stream. However, a simple estimate of the necessary vapor flow for a particular power input shows that the change in pressure and the equivalent change in temperature are always negligible for any practical situation.

Frequently it may be advantageous to insert a heat screen around the actual heat pipe in order to reduce the radiation losses, the necessary vapor flow and, of course, also the input power. This is highly desirable at temperatures of about 1500°C and higher where radiation losses can readily be of the order of 10 kW and larger. At these powers it requires special attention to avoid the formation of hot spots which may damage the heat pipe. These hot spots are generated if the evaporating liquid cannot be replenished fast enough through the wick.

For absolute calibrations one also needs the total number of particles or the effective length of the vapor zone. These quantities are not quite as easily determined as the temperature and the pressure, because now we also have to worry about the two boundary layers between the vapor and the confining inert gas. The shape and the density distribution of these layers are essentially determined by the amount of vapor flow, the condensation process and by the mean-free path for neutral-neutral collisions. Therefore, as in the laminar flow situation, the boundary layer will not be a plane. Hence the temperature distribution of the pipe wall does not directly reflect the partial vapor pressure in the boundary layer.

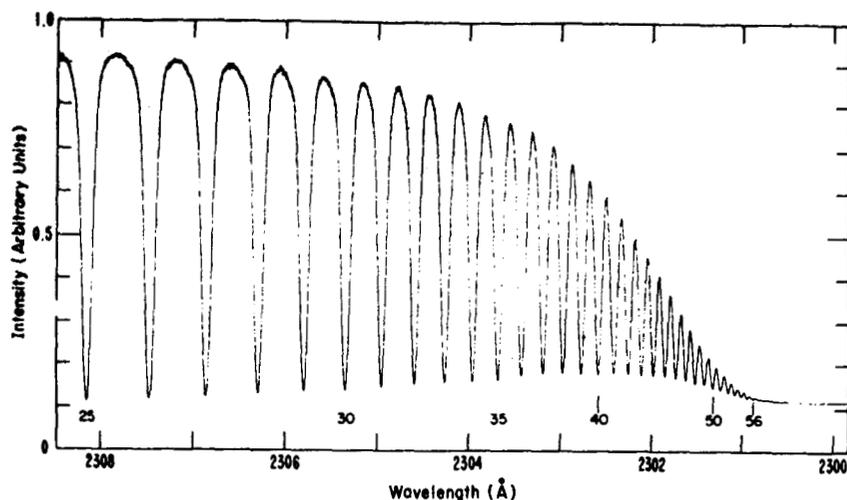
However, without going into details of how the boundary layer is formed there is a simple method to examine quantitatively the effect of the boundary layers. If we keep the pressure of the inert gas constant and change only the power input then we always maintain the same temperature in the central vapor zone and the same type of boundary layer. Hence, it is important to realize that by raising the input power at a constant inert gas pressure one only extends the length of the vapor zone by an accurately measurable amount l without affecting the boundary layers. In other words, one inserts effectively a well defined, homogeneous vapor section of length l . This fact can be exploited in different ways and the most suitable method depends on the accuracy required.

Let us assume we are interested in absorption measurements. Similar considerations hold, of course, also for emission measurements. Then the optical depth τ which is a function of the frequency ν is given by

$$\tau(\nu) = \int \kappa_\nu(x) dx = \int \sigma_\nu(x) \cdot n_p(x) dx, \quad (2)$$

where κ_ν is the absorption coefficient, σ_ν the absorption cross section, and n_p the population density of the par-

FIG. 3. Lithium absorption spectrum of the principal series 2^2S-2^2P measured at a vapor pressure of 8.2 Torr.



ticular absorbing state which in most cases will be the ground state. All three quantities are in general a function of position in the boundary layer and are essentially constant throughout the vapor zone where we denote them by a subscript 0.

If one operates the heat-pipe oven at a constant pressure with two different powers P_1 and P_2 , where we take P_2 to be larger than P_1 , then the difference between the corresponding two optical depths is given without any assumption by

$$\tau_2(\nu) - \tau_1(\nu) = \sigma_{\nu} n_0 \cdot l, \quad (3)$$

because, as discussed before, the two metal vapor zones will only differ by a homogeneous section of length l in which the population density and the absorption cross section are constant. Therefore by measuring the difference of the optical depths one completely eliminates any effect due to the boundary layers.

In case of a single oven this can be done by two successive scans at different powers. A more accurate way would be to operate simultaneously two identical heat pipe ovens at different powers in a customary double beam arrangement which can be adjusted in such a manner that one obtains directly the difference of the two optical depths from the ratio of the two outgoing intensities. By interconnecting both pipes one establishes the same inert gas pressure in each and automatically also the same pressure and temperature of the vapor and the same boundary layers. Such a differential measuring technique is readily performed because of the simple technology and operation of the heat pipe. Usually, however, it will only be advisable if for absolute calibrations one strives for the ultimate accuracy. In many applications, especially if one is interested only in relative measurements, it is sufficient to operate a single heat pipe where one keeps the boundary layers as short as possible with regard to the vapor zone.

From the preceding discussion it appears that for absolute calibrations the crucial quantity is the length l .

Its measurement has been demonstrated in Fig. 2, where temperature profiles are shown for a heat pipe oven which operates with lithium and using helium as the confining gas at a pressure of 10.9 Torr. The temperature along the heat pipe has been measured for two different input powers by means of a pyrometer which has an accuracy of about 5°C and measures down to 780°C . One realizes that within the measured temperature range the temperature gradient is the same on both sides for both powers. Therefore the length l can be determined accurately from the shift of the temperature slopes as indicated in Fig. 2. These slopes are so steep that the accuracy of the length l is usually determined more by the position measurement than by the temperature measurement. Accuracies of about 1 mm can easily be achieved, meaning that for 1% accuracy l should be at least about 10 cm. It should be mentioned that due to the emissivity of the tube wall the apparent temperature measured by the pyrometer is slightly lower than the true temperature measured by a thermocouple. Therefore, Fig. 2, shows a temperature of 891°C around the vapor section instead of 902°C according to the vapor pressure curve of Li as given in the literature. However this does not affect the determination of the length l .

Using higher pressures the temperature gradient became larger and the boundary layers shorter because the mean free path for neutral-neutral collisions is shorter. This is due to the fact that the temperature distribution around the boundary layer is not determined by the ordinary heat conductivity in a gas but by the amount of vapor condensing on the wick. Therefore, the thickness of the boundary layer is determined by the reaching distance of the atoms into the confining inert gas.

The heat pipe oven has already been used successfully in a number of spectroscopic measurements. Figure 3 shows part of an absorption spectrum of lithium using a xenon high-pressure arc lamp as a background source and a 2-m Czerny Turner grating spectrometer

with a resolving power of 120 000 in first order. It is a photoelectric scan of the lithium principal series 2^2S-n^2P which has been observed up to principal quantum numbers of about $n=60$ where the lines are no longer discernible because of the merging. Furthermore, resonance fluorescence spectra of the Li_2 molecule have been observed after excitation by various lines of the argon ion laser.

SUMMARY

A new metal vapor device called the heat-pipe oven has been presented. It has several unprecedented features which should make it very useful in a large number of spectroscopic and other applications. It continuously generates homogeneous vapors of well-defined temperature, pressure, and optical path length. These parameters can be measured easily and accurately in a direct manner. The temperature is constant to within at most 1°C over the whole vapor zone without using radiation shields. This is due to the continuous evaporation and condensation of metal vapor, which also purifies the vapor during operation. The pressure can be measured directly from the pressure of the confining inert gas without relying on vapor pressure

curves. The inert gas boundaries remove the window problem and also make the heat-pipe oven a very suitable instrument for vacuum uv spectroscopy. This is due to the fact that at typical pressures of about 10 Torr the heat-pipe oven can easily be attached to a vacuum spectrometer without a LiF window by differentially pumping the inert gas.

Up to now the heat pipe oven has been designed primarily for spectroscopic applications. However, it is clear that its unique features permit the heat-pipe oven to serve as a starting point for a number of other applications. For example, the application of this device to the generation of metal vapor plasmas is now in progress.

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