Heat Pipe Oven Applications. I. Isothermal Heater of Well Defined Temperature. II. Production of Metal Vapor–Gas Mixtures

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A concentric heat pipe oven is described, which serves as an oven with a highly homogeneous temperature distribution as required by such applications as crystal growing, thermal treatment of materials, and radiation standards. The design is simpler than conventional ovens with similar temperature stability and homogeneity. The temperature control is replaced by a pressure control. This device is used in a modification of the heat pipe oven that generates homogeneous mixtures of a vapor (such as a metal vapor) and an inert gas at well defined total pressure, partial pressure, temperature, and optical path length. All the features of the previously described heat pipe oven are maintained with the additional option that allows quantitative total and partial pressure measurements without relying on vapor pressure curves.

INTRODUCTION

Recently a new type of oven called the heat pipe oven was developed by Vidal and Cooper[1] as a well defined metal vapor cell for quantitative spectroscopic measurements. Their paper is henceforth referred to as paper I. The design of the heat pipe oven is based on the heat pipe, a heat conductive element first described by Grover, Cotter, and Erickson.[2] The heat pipe oven differs in several unique ways from the well known oven originally described by King,[3] which has been improved many times since then. It continuously generates homogeneous pure metal vapors of well defined pressure, temperature, and optical path length. All parameters can be measured easily and more accurately and directly than for the King furnace without relying on vapor pressure curves. (This is demonstrated in paper I.) The inert gas boundaries, which confine the metal vapor section and allow a direct pressure measurement, remove the window problem.

Over the past three years of working with the heat pipe oven, we have never encountered any problem with condensation of material on the windows, even down to pressures of about 0.01 Torr. The most severe problem we have had is related to the penetration of refractory metals like tantalum, molybdenum, tungsten, and niobium by the alkali metals investigated, in particular lithium. This penetration along grain boundaries was observed by Klueh[4] only when the oxygen dissolved in the refractory metal exceeds some temperature dependent threshold concentration. Another problem is related to the oxygen content of the alkali metal which may attack the refractory material.[5] However, both corrosion problems may be overcome by using refractory metals alloyed with a small percentage of yttrium, zirconium, or hafnium.[6,7] We have successfully used a small sample of yttrium as a getter inserted into the center portion of the heat pipe oven.

Owing to its confining low pressure inert bas boundaries, the heat pipe oven is also a suitable instrument for vacuum uv spectroscopy. This has been shown in the wavelength range around 200 Å by Ederer, Lucatorto, and Madden[8]

using the National Bureau of Standards 180 MeV synchrotron. Other successful applications have been resonance fluorescence measurements of alkali molecules such as Li_2[9] and NaLi.[10] The latter molecule was observed for the first time with a modified heat pipe oven that allows the simultaneous generation of two different metal vapor zones in the oven as described by Hessel and Jankowski.[11] Among other applications, ir alkali metal vapor lasers[12] and the Zeeman effect of long spectral series in the absorption spectrum of barium[13] have been investigated by means of the heat pipe oven.

In all these experiments, with the partial exception of Ref. 12, the heat pipe oven has been operated with pure metal vapors as originally described. If the concentration of impurities is small the heat pipe purifies itself during operation as explained in paper I. In a number of applications, such as foreign gas broadening or light scattering experiments, it is desirable to have a system in which one has not only pure vapors but also mixtures of vapors with nonreactive gases. For quantitative measurements one then needs the total and partial pressures of the constituents.

In the first part of this paper a concentric heat pipe oven is described whose temperature distribution is highly homogeneous and whose temperature can be maintained at a well defined value. Owing to the simplicity of its design, the oven will be useful by itself in many technical applications. In the following sections we describe how this oven is used as a heater for a device that is capable of handling mixtures of vapors and gases in a well defined manner without losing the unique features of the heat pipe oven.

ISOTHERMAL HEATER OF WELL DEFINED TEMPERATURE

In designing an oven with a homogeneous temperature distribution one could use in principle either the closed heat pipe as described by Grover, Cotter, and Erickson[2] or the open ended heat pipe as described in paper I. Both
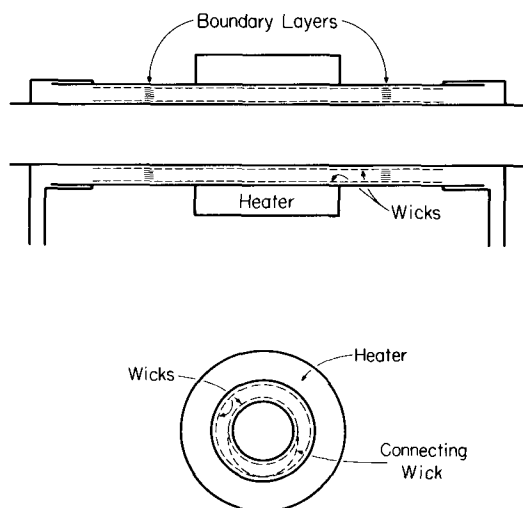
FIG. 1. Schematic diagram of the concentric heat pipe oven which serves as the isothermal heater of well defined temperature.

devices provide the high heat conductivity to insure a homogeneous temperature distribution. They differ, however, in a few very significant features, which make them suitable for different problems. Let us assume that both types of heat pipes operate with the same vapor. In case of the closed heat pipe the temperature and the pressure inside the pipe are determined by the power balance along the pipe and the temperature is the same over the whole length (see also Eastman[14]). In case of the open heat pipe the temperature is only a function of the confining inert gas pressure as long as the vapor flow velocities are not too large.[1] At large flow velocities the temperature is lowered due to the Bernoulli term in the pressure balance.[15] The power put into the open heat pipe does not affect the temperature, but only the length of the heated zone.[1]

It is therefore clear that for an oven of well defined temperature the open ended heat pipe oven appears to be more advantageous because it requires only a well controlled pressure. For the closed heat pipe a sophisticated regulation of the power is necessary based on a temperature measurement which has to be much more accurate than the pressure measurement of the open ended heat pipe oven. This is most clearly illustrated by an example. If we use lithium as a heat pipe material we find from the vapor pressure curve that at a temperature of 1000 K a 1% change in temperature corresponds to a 20% change in pressure. Consequently, if one wants to control the temperature to within ± 1 K, the buffer gas pressure has to be maintained within $\pm 2\%$. This may readily be done, if the open ended heat pipe has a large enough external pressure reservoir. Since the pressure in the external pressure reservoir follows the ideal gas law, it means that one needs to control the buffer gas temperature of the pressure reservoir to 2% at room temperature to maintain the oven temperature constant to 0.1%.

It should be noted that this argument applies also to other materials since the slope of the vapor pressure curves (dP/dT) is about the same for similar vapor pressures of the other materials.

Figure 1 shows schematically the concentric open ended heat pipe oven which serves as the isothermal heater of well defined temperature. It may be looked at as an ordinary heat pipe oven with a tube inserted along the axis of the pipe. Our first device was built out of stainless steel with lithium as the working material. The outer wick of the inner tube and the wick of the outer heat pipe are both made out of four layers of stainless steel mesh. The bottom part of Fig. 1 shows a cross section of the center part of the concentric heat pipe oven. The diagram shows the heater, the inner and outer pipe with the wicks, and a loop shaped wick, which connects the outer and inner wick. It is the purpose of the connecting wick to provide a path back to the outer wick, and to avoid accumulation of the vapor material on the inner wick. Without the connecting wick the outer wick is eventually depleted and the outer pipe will form hot spots.

The concentric heat pipe oven should be useful in a number of applications where an oven with a well defined homogeneous temperature distribution is required such as thermal treatment of materials and crystal growth. For thermal processing a series of concentric ovens may be arranged where the temperature of every individual oven may be adjusted separately by its inert gas pressure. Furthermore, the concentric oven should also be useful as a radiation standard from the visible range of the spectrum down to the microwave range. By inserting diaphragms on one end of the heated zone and closing off the other end of the heated zone one obtains a blackbody radiation source in the visible and infrared. In the microwave region the inner tube may easily be shaped into a waveguide to provide a well matched radiation standard.

PRINCIPLE OF THE MODIFIED HEAT PIPE OVEN FOR VAPOR–GAS MIXTURES

The modified heat pipe oven is shown schematically in Fig. 2. On the upper left is a diagram of the open ended heat pipe with the mesh structure on the inner side, which serves as a wick. Let us assume in the following discussion that the pipe is surrounded by an ideal heater that heats the pipe homogeneously (uniform temperature) over the length L. Later we replace the ideal heater by the concentric heat pipe oven described above.

Initially the pressure of the inert gas throughout the pipe is P_a and the heat pipe is filled with a material whose vapor pressure curve is given in Fig. 2. If one starts to heat the pipe, the metal vapor pressure follows the vapor pressure curve and reaches pressure P_b and temperature T_b at some subcritical power of the heater. The corre-
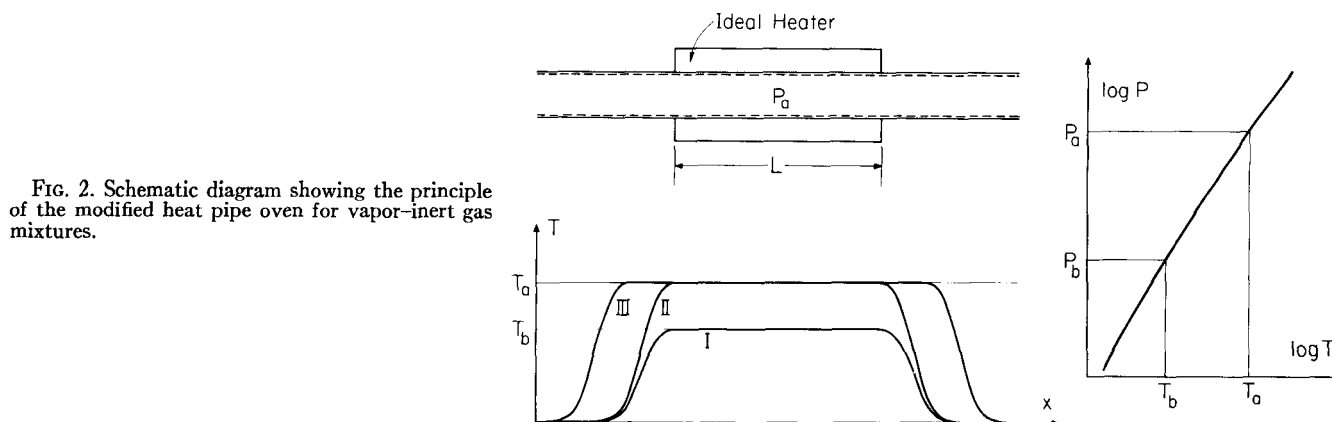
FIG. 2. Schematic diagram showing the principle of the modified heat pipe oven for vapor–inert gas mixtures.

sponding temperature profile along the heat pipe oven is shown schematically in Fig. 2 (curve I). At this point one has a total pressure throughout the pipe, P_a, in the heated region a partial pressure of the evaporating material, P_b, and an inert gas pressure $P_a - P_b$. By further increasing the heater power the temperature distribution along the pipe moves up until it reaches the critical temperature T_a that is determined by the confining inert gas pressure which outside the metal vapor region is P_a (curve II). The system now works as a heat pipe oven.[1] A further increase in power does not change the maximum temperature T_a as long as P_a is kept constant. It will only extend the length of the metal vapor section (curve III) so that a balance between the heater power and the power losses due to radiation and/or conduction is established.

In order to achieve this operation in which the vapor is mixed with the inert gas one needs an ideal heater that is able to operate at any subcritical power. Furthermore, the vapor pressure curve has to be known well enough to derive the partial pressure P_b from the measured temperature T_b. In practice, both problems are linked and are by no means trivial. At subcritical temperatures like T_b we have lost the high heat conductivity of the heat pipe and hence do not have the homogeneous temperature distribution and the homogeneous partial pressure over the length of the heater. The only remaining advantage of the inner heat pipe with respect to the well known King furnace is provided by the wick, which returns the liquid metal to the heater section and avoids the common problem of material accumulation at the ends of the furnace.

In principle, one could resort at this stage to the old art of building a heater that provides a homogeneous temperature distribution by using a sophisticated system of heat shields, baffles, heaters, and temperature controls. This method has been described in many papers, such as those by King and Stockbarger[16] or Tomkins and Ercoli.[17] However, it is certainly simpler to use the concentric heat pipe oven descrived above. This has been done in an arrangement shown schematically in Fig. 3. The inner open-

ended heat pipe, which corresponds to the heat pipe in Fig. 2, is concentrically surrounded by a second open-ended heat pipe. If the outer heat pipe is operated as an ordinary heat pipe oven, it provides the homogeneous temperature distribution of an ideal heater for the inner pipe. By using the same metal vapor in the outer as well as in the inner pipe we have achieved another important feature, namely, the total pressure P_b of the outer pipe is identical to the partial pressure in the inner pipe as long as P_a is larger than P_b. This occurs because the temperature is the same in both pipes.

In some cases it may still be advantageous to operate with two different materials in the outer and inner pipe, although one loses the simple and direct method of determining the partial pressure. This may happen if the material of interest in the inner pipe is not very suitable for a heat pipe operation due to small heat of vaporization, small surface tension, high viscosity, or large density. One then should try to find combinations of materials whose vapor pressure curves are not too far apart. We also note that in working with less suitable heat pipe materials, it is advisable to make the heated section (for example, by an induction heater) only slightly shorter than the desired length of the vapor section inside the heat pipe oven. This minimizes the vapor flow and the flow of the liquid in the wick and reduces the possibility of hot spot formation.

Our first modified heat pipe oven was operated with lithium and helium. Both pipes were made of stainless steel, the inner one of 25 mm diameter and the outer one of 50 mm diameter. The three wick structures as shown in Fig. 3 were all made of four layers of stainless steel mesh. As indicated in Fig. 1 the inner wick of the outer pipe and the outer wick of the inner pipe are again connected by a loop-shaped wick structure (four layers of stainless steel mesh) in the center part close to the heated section. The outer pipe is heated by an induction heater. For simplicity we have built the first versions of the modified heat pipe oven of stainless steel because we have been mainly inter-
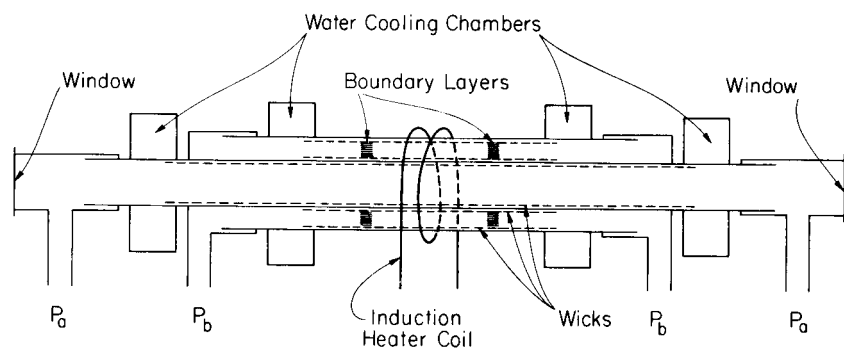
FIG. 3. Schematic arrangement of the modified heat pipe oven for vapor–inert gas mixtures.

ested in small partial pressures of the metal vapor, i.e., about 1 Torr and smaller. For the corresponding temperatures of about 1020 K and lower, stainless steel has a reasonable life time even with lithium.[7]

PROPERTIES OF THE MODIFIED HEAT PIPE OVEN

As in the ordinary heat pipe oven, the confining non-reactive gas boundaries prevent any chemical reactions of the metal vapor with the end windows of the inner heat pipe. Usually the inner heat pipe does not work in the heat pipe oven mode as long as the inner pressure P_a is larger than the outer pressure P_b. However, the wick structure of the inner pipe still avoids accumulation of the material at the end of the pipe by returning it to the heater section by surface tension.

The high heat conductivity of the outer heat pipe, which works like an ordinary heat pipe oven, insures a homogeneous temperature T_b and consequently a homogeneous vapor pressure P_b. Both parameters, T_b and P_b, can be measured easily and directly without relying on vapor pressure curves. For example, the temperature T_b is measured by means of a thermocouple. The pressure P_b is immediately measurable from the confining inert gas pressure in the outer pipe.[18]

The length of the heated section can be easily varied and is determined by the input power so that a power balance is achieved between the input power and the power losses due to radiation and/or conduction through the pipe walls. In this manner the optical path length can be varied without changing the gas and vapor densities.

For identical materials in the inner and outer pipe, one of the essential new features of the modified heat pipe oven is that the total pressure P_b of the outer pipe is identical to the partial pressure of the inner pipe.

For quantitative spectroscopic measurements such as oscillator strengths one has to know, in addition to the densities or partial pressures, the total number of absorbing particles or the effective length. It may be suspected that in keeping the pressure P_b and the length of the heated zone constant, the total number of absorbing particles may still be a function of the total pressure P_a in the inner

pipe due to changes in the density distribution of the transition region between the vapor and the buffer gas. We looked at the absorption spectra of the lithium continuum around 2300 Å and we found that at least for $P_b \gtrsim 0.3$ Torr the total number of absorbing particles was constant within the accuracy of the measurement.

To obtain the absorption spectrum we used a xenon high pressure arc lamp to provide the continuous background radiation. The spectrum was taken in second order with a 2 m Czerny–Turner grating spectrometer (resolving power in first order is 120 000) using a solar blind multiplier 541F−05M−14 whose long wavelength cutoff point is around 3500 Å. The first measurement was taken after connecting the outer and inner pipe so that $P_a = P_b$. In this situation both the outer and the inner pipe work as a heat pipe oven. The outer and inner pipes were then disconnected and the pressure P_a was increased, keeping P_b and the heater power constant. By measuring the optical depth τ in the continuum, which is directly proportional to the total number of absorbing particles, it was found that for lithium pressures $P_b \gtrsim 0.3$ Torr, the optical depth was independent of the total helium pressure P_a within 3% up to 500 Torr. The pressure P_a was increased by factors of about 2 for each subsequent measurement until we reached 500 Torr. The optical depth was measured most carefully for pressures P_a slightly larger than P_b where the inner heat pipe ceases to work as a heat pipe oven. The accuracy was essentially limited by the stability of the xenon high pressure arc lamp, whose intensity was monitored separately, and the photon emission noise of the photomultiplier, whose dark current was negligible.

For pressures below about 0.3 Torr and a maximum length of the heated zone of about 20 cm, the optical depth in the continuum was too small to permit accurate enough measurements of changes in the total number of absorbing particles. We have tried to use the total absorption of some of the higher lithium resonance lines. Owing to the apparatus profile of the instrument whose width is comparable or even larger than the widths of the resonance lines, the errors in deconvolution are too large to obtain accurate optical depths integrated over a line. This is due to the well known fact that unlike an emission spectrum, the

FIG. 4. Double beam arrangement with two modified heat pipe ovens in which contributions to the absorption from the boundary layers may be suppressed.
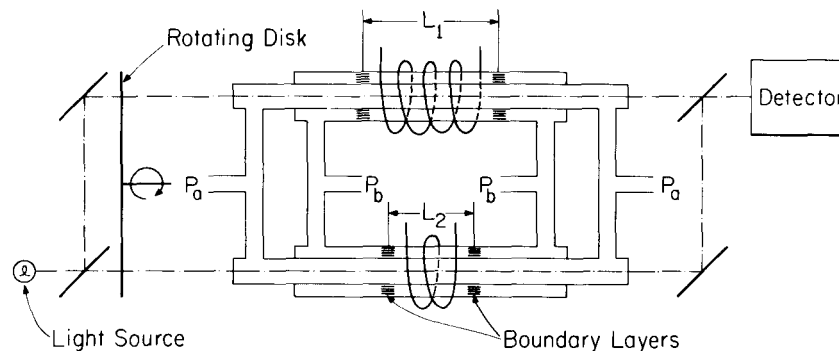
apparent total absorption integrated over a spectral line may be seriously affected by the apparatus profile because it has to be folded with the exponential of the optical depth, $\exp[-\tau(\nu)]$, and not directly with the profile $\tau(\nu)$. Knowing the broadening parameter and the temperature, and assuming for $\tau(\nu)$ a Voigt profile, one could in principle apply a curve of growth analysis that is independent of the apparatus profile because it exploits only the equivalent width. Since at the moment broadening constants are not available with sufficient precision, this technique has not yet been applied. These data may rather be used to obtain the broadening constants.

Finally, one has to look carefully at the transition region of the vapor zone to the pure gas zone in the inner pipe. According to Fig. 2 one expects a region of homogeneous partial pressure and a rapid decrease where the temperature profile drops off. In order to account for the absorption in the transition region and to determine the length of a zone with homogeneous partial pressure, one can apply the two methods described in paper I with an obvious modification.

In the first method, the pressures P_a and P_b are kept constant and the heater power is varied. This changes the over-all length of the vapor zone without affecting the density distribution in the boundary as long as one stays far enough away from the cooling chambers shown in Fig. 3. In this manner one measures the optical depth $\tau_1(\nu)$ and $\tau_2(\nu)$ at a high and a low power input and, by taking the difference

$$\tau_1(\nu) - \tau_2(\nu) = \sigma(\nu)nl,$$

the contribution of the boundaries is canceled. Some caution is necessary if the instrument profile affects the absorption spectrum. The length l corresponds again to the difference in the measured width of the two temperature profiles for the two different input powers as shown in Fig. 2 of paper I. $\sigma(\nu)$ is the absorption cross section and n is the population density of the absorbing state which in most cases will be the ground state.

The second method is shown schematically in Fig. 4. Here one avoids the two successive scans of the absorption

spectrum by scanning the difference in the optical depths $\tau_1(\nu) - \tau_2(\nu)$ with a conventional double beam arrangement. If one uses two identical modified heat pipe ovens with the two outer heat pipes as well as the two inner heat pipes connected together, the pressures P_a and P_b will be the same in both pipes, hence the transition regions will have the same density distributions in both ovens. The rotating disk opens alternately the two optical paths of the double beam arrangement. The measurement is made in two steps. First the heat pipe ovens are turned off and the system is adjusted so that the absorption is identical in both optical paths. Then both ovens are turned on with one of them having a longer heated zone than the other; the difference in the two optical depths is obtained immediately from the ratio of the two outcoming intensities. The length l is again given by the difference $L_1 - L_2$.

It should also be noted that in a number of applications in which, for example, only relative intensity measurements are required, the contribution from the transition regions may not matter, especially if P_a is very much larger than P_b.

The coaxial heat pipe arrangement should also be a good device for generating plasmas in mixtures of metal vapors and inert gases such as those found in metal vapor lasers. Except for possible demixing effects, this device should provide a constant partial pressure of metal vapor, even in the presence of high discharge currents, because of the temperature stability of the outer heat pipe. In other devices,[19] high powers produce additional heating which then increases the partial pressure, whereas in the coaxial heat pipe one expects only a change in the length of the heated zone with no change in pressure or temperature. Even this change could be minimized by readjusting the power balance of the system, e.g., even cooling the outer pipe. Preliminary tests have already been performed in which a barium discharge was maintained in a heat pipe using only the power supplied to the discharge. The heating power was used initially to start the discharge.

ACKNOWLEDGMENTS

those leading to the basic idea of the modified heat pipe oven.

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