

Chemiluminescent reactions in a heat-pipe oven*

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(Received 4 November 1974)

A heat-pipe oven has been used to contain and control the chemiluminescent reaction $\text{Ba} + \text{N}_2\text{O} \rightarrow \text{BaO}^* + \text{N}_2$. The heat-pipe oven permits Ba vapor to be maintained at any desired pressure. Reactions were easily controlled by varying the flow rate of N_2O or pressure of Ba. A large volume (about 20 cm^3) of chemiluminescence was produced and spectra were taken from 0.1 to 5 Torr. In addition to emission from $\text{BaO } A^1\Sigma-X^1\Sigma$, numerous atomic Ba lines also have been observed. This device is well suited to the study and control of chemical reactions between metal vapors and oxidizers.

PACS numbers: 82.40.T, 07.20.H

The traditional technique for producing and spectroscopically observing chemiluminescent reactions between two molecular species has been either by crossed molecular beams¹ or by flame spectroscopy.² For either of these techniques the vapor density of both reacting species is not well determined and the flame has a nonuniform intensity distribution.

We have made an easily controllable chemiluminescent light source by reacting Ba with N_2O in a crossed heat-pipe oven.^{3,4} A heat-pipe oven is a device that generates vapors of well-defined pressure, temperature, and optical path length. The metal vapor is confined by inert-gas boundaries, and therefore, a direct measurement of the inert-gas pressure gives the metal-vapor density without relying on vapor-pressure curves. Although the reaction takes place at the gas-metal-vapor interface and a concentration gradient of reactants exists, this device eliminates the pressure gradients. Figure 1 is a schematic diagram of our experimental arrangement. We used a stainless-steel heat-pipe oven and a stainless-steel mesh wick of the same design and dimensions described by Ref. 4. One arm of the oven was welded closed and filled with about 200 g of barium. From 0.1 to 5 Torr (1 Torr = 133.3 Pa) of He buffer gas was admitted to the oven. Buffer gas pressures were measured to better than 0.05 Torr by means of a capacitance manometer. The rf and resistance heater power were adjusted so that there was barium vapor beyond the cross region of the pipe. N_2O was admitted through one arm of the cross and the flow rate was adjusted by means of an inlet valve. Flow rates of N_2O were about 3×10^{18} molecules/sec corresponding to a Ba consumption of about 0.5 mg/sec at a pressure of 2 Torr. A bright chemiluminescent flame was observed to be approximately uniform throughout the diameter of the heat-pipe oven. The brightness and extent of the reactive zone depended upon the buffer gas pressure and flow rate of N_2O ; the brightest chemiluminescence occurred at the lower pressures. Observed spectral distributions were similar to those described by Jones and Broida,⁵ but in addition numerous atomic lines were observed.

Figure 2 shows the chemiluminescence spectra of the $\text{Ba} + \text{N}_2\text{O}$ reaction at pressures of 0.2 and 2.0 Torr in

the heat-pipe oven. Spectra were not corrected for phototube response. At low pressures, the light was extremely bright, filling the diameter of the cross tube and extending about 5 cm in the direction of the N_2O valve. The spectrum at 0.2 Torr shows a typically headless emission⁵ with many superimposed $\text{BaO } A^1\Sigma-X^1\Sigma$ bands and Ba atomic lines. The atomic lines primarily come from the $6p^3P$ and $3D$ levels. In addition, the Li resonance line at 670.8 nm and the Ca and Sr resonance lines 422.7, 460.7, and 689.3 nm were observed. Li had been put into the pipe during a previous experiment and even after careful cleaning, some Li probably remained diffused into the stainless-steel walls. The Sr and Ca lines arise from their 2000 ppm impurity in Ba. The relative intensity of atomic emission was strongly dependent on the N_2O flow rate which also controlled the position of the flame zone. At 2.0 Torr the atomic Ba emission had virtually disappeared and well-developed BaO band heads were observed. The light intensity was moderately bright. In the heat-pipe oven, conditions could be changed from high to low pressures within a few minutes.

In the reaction of N_2O and Ba, no pumping was necessary to maintain constant pressure even at very large gas flow rates. Apparently, barium was an efficient getter for the N_2 produced and the BaO condensed on the walls of the chamber. We looked for droplet formation by scattering at 90° from both laser and white light sources.⁶ No scatter of this light from condensation

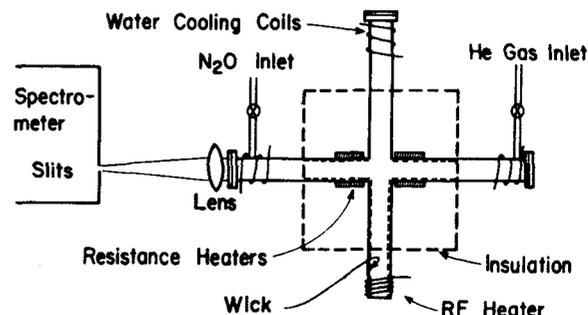


FIG. 1. Schematic diagram of the apparatus.

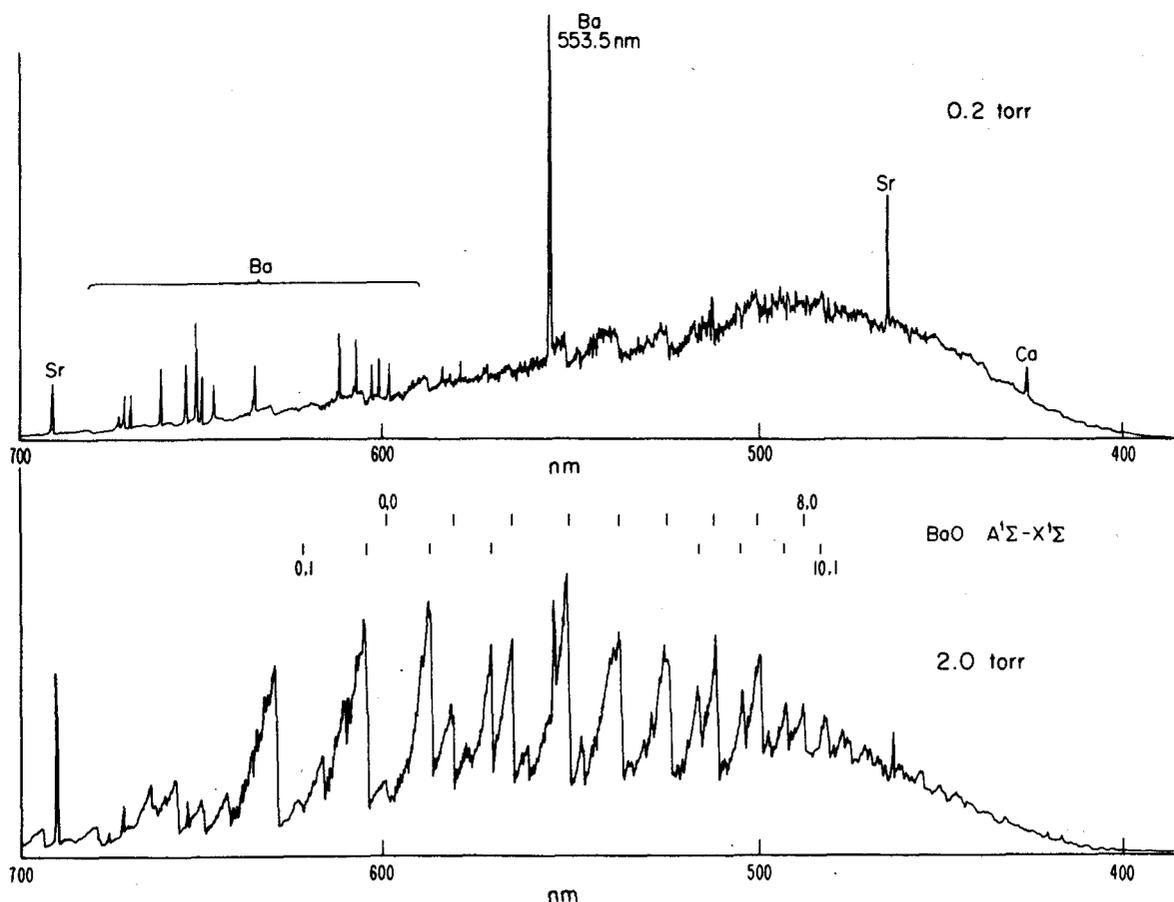


FIG. 2. Spectral distribution of the chemiluminescent reaction of $\text{Ba} + \text{N}_2\text{O} \rightarrow \text{BaO}^* + \text{N}_2$. Upper trace at 0.2 Torr pressure, lower trace at 2.0 Torr.

droplets was observed.

These observations on the chemiluminescence of Ba and N_2O show that heat-pipe ovens provide an excellent means of producing and studying metal vapor and oxidizer chemical reactions with well-controlled pressure and temperature over a large volume.

*Contribution of the National Bureau of Standards.

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