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MEASUREMENTS OF THE EXTINCTION PARAMETERS OF HOT SEEDED HYDROGEN AT HIGH PRESSURES

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16. Abstract

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FOREWORD

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TABLE OF CONTENTS

SUMMARY

Aerosols of submicron-sized refractory particles are currently being considered as propellants for gas core rockets and for thermal radiation shielding in nuclear rocket nozzles. In order to be useful for these applications, the particles should be strongly absorbing in the ultraviolet, visible, and near infrared regions of the spectrum and should withstand high temperatures at high pressures without reacting or vaporizing. Submicron-sized particles of refractory metals generally satisfy both of these requirements.

Measurements of the extinction parameter of tungsten-hydrogen aerosols were made as a function of wavelength from 2500 Å to 5800 Å at pressures from 1 to 115 atmospheres and temperatures from 300°K to 2500°K. The measured extinction parameter at room temperature corresponded closely, over this pressure range, to the value of the extinction parameter calculated using the Mie theory. For temperatures above about 1000°K, the extinction parameter was found to increase with further increases in temperature. This increase at 2000°K was about a factor of two for a pressure of one atmosphere, about a factor of 4 at about 10 atmospheres pressure, and much greater at 100 atmospheres pressure. At 1 atmosphere the extinction parameter of a well dispersed tungsten aerosol increased from 18,000 cm^2/gm at room temperature to 30,000 cm^2/gm at 2000°K. At 100 atmospheres, the extinction parameter of a less well dispersed tungsten aerosol increased from 8,000 cm^2/gm at room temperature to 120,000 cm^2/gm at 2300°K. The opacity of tungsten-seeded hydrogen at temperatures of about 2000°K or more is shown to be much greater at high pressures than at one atmosphere pressure.

INTRODUCTION

The absorption of thermal radiation by a gas is usually greatly enhanced by the addition of very small particles of a suitable material. If the particles are small enough, they may remain essentially in thermal and dynamic equilibrium with the gas as the gas is heated by radiation to the particles with subsequent conduction from the particles to the gas.[1] Particle-seeded hydrogen is currently being considered as the propellant for the gaseous core nuclear rocket.[2]

Two gaseous core nuclear reactor concepts that are currently being investigated for nuclear rocket propulsion are the coaxial flow reactor,[3,4] in which a slow moving central stream of gaseous fissioning fuel heats a fast moving annular stream of particle-seeded gas primarily by thermal radiation, and the nuclear light bulb reactor,[5,6] in which fissioning fuel is contained in a transparent partition and a particle-seeded gas is heated by thermal radiation through the partition. The particles in the gas are necessary to insure sufficient radiant heat transfer to the working fluid and to minimize heating of the containment vessel. The performance of a gaseous core nuclear rocket would be far superior to any present-day rocket, chemical or nuclear.

Since a gas-core nuclear rocket would rely on thermal radiation from the hot fissioning core to heat the hydrogen propellant, particle seeding is essential to its operation. Whereas gases alone tend to absorb thermal radiation in lines and bands, the absorption characteristics of particle clouds vary gradually with wavelength. Particle clouds of some refractory materials have been shown to have very high absorption coefficients such that a small mass fraction of such a particle seed material dispersed in a gas will make the gas highly absorbing.

The objective of the investigation described in this report was the measurement of the radiant heat attenuation of an aerosol which may serve as the gas core nuclear rocket propellant, and is an extension of the research described in NASA CR-1504. This experiment utilized a tungsten-hydrogen aerosol heated to temperatures as high as 2500°K and under pressures up to 115 atmospheres. The hydrogen aerosol was produced by dispersing submicron-sized particles of tungsten in hydrogen gas. The principal property

broad spectrum, visible and ultraviolet, light passing through it with the attenuation being measured as a function of wavelength. Other characteristics of the aerosol that were examined included the nature and extent of chemical reactions between the seed material and the hydrogen and the degree of dispersion of the seed material obtained before and after heating. Chemical equilibrium calculations and vapor pressure data[7] for the refractory metals indicate that tungsten is a prime candidate for the seed material in the gas core nuclear rocket.

BACKGROUND

Both the open cycle and light bulb gas core reactor concepts have in common the requirement of transferring thermal radiant energy from the uranium fuel region to the hydrogen.[1-6] As indicated by experimental data[8,9] on uranium plasmas, most of the emission spectrum appears to be concentrated below 7000 Å. Hydrogen at temperatures below about 8000°K is transparent to radiation of wavelengths longer than 1216 Å where the first transition in the Lyman series occurs. As the hydrogen is heated, more of the hydrogen atoms are in excited states and can absorb radiation of longer wavelengths. Above about 8000°K the absorption by the hydrogen becomes significant and is sufficient to strongly attenuate the radiant energy from a uranium plasma.[10,11] For propellant temperatures below this, some other mechanism of heat transfer must be employed. The hydrogen could be seeded with a gas that would absorb in the 900-7000 Å region but the process would be line absorption and the integrated absorption would not be sufficient. Seeding by small particles is favored because they give a very broad absorption that varies uniformly with wavelength. There are several criteria that the seed particles have to meet. Their optical absorption must be large so that a particle to propellant mass ratio smaller than about one percent will be sufficient to absorb the radiant energy. Unless the particles are fissionable, their neutron absorption cross section must be low or the critical mass of the fissile fuel may be significantly increased. A material with a high vaporization temperature and a low vapor pressure is desirable. The chemical reaction rate with hydrogen should be small. These considerations indicated that submicron-sized refractory metal particles may be the best candidates. Carbon was favored early in the development of the gas core nuclear reactor concept; however, Shenoy, et al.[12,13] have shown it to have a reaction rate with hydrogen that makes it unsuitable as a seed material. Carbon is attractive, except for the chemical reaction, because it has a low density and low molecular weight and is highly absorbing for thermal radiation.

Other materials such as alumina and silica which would seem to offer these advantages, being known as good attenuators of light, are unsatisfactory because they are almost purely scattering and absorb very little energy. Also, high temperature high pressure hydrogen tends to reduce the oxide re-

fractory materials.

Heat transfer mechanisms in the gas core reactor are complex. In the central region of the cavity is the gaseous uranium core which is essentially opaque to radiant energy.[14] As a result of the high opacity, radiant energy is emitted from very near the surface of the uranium plasma, which may have approximately a spherical geometry in the case of the co-axial flow concept. The temperature in the interior of the uranium plasma may extend up to 100,000°K, but the temperature profile is very severe and a skin temperature in the neighborhood of 20,000°K is expected. The seeded propellant closest to the core and that near the nozzle would have been heated high enough so that no solid particles can exist and the radiant energy absorption is due to the heated hydrogen and the vaporized or chemically reacted seed material. Krascella[10] and Patch[11] have calculated the absorption of pure hydrogen as a function of pressure, temperature, and wavelength. Krascella[15] also calculated the Rosseland mean opacity of tungsten vapor using a semiempirical model. The Rosseland mean opacity is an average of the spectral absorption coefficient over the black body spectrum.

Near the walls of the reactor cavity and near the hydrogen propellant entrance, the seed material would be in the form of submicron-sized particles in dynamic equilibrium with the hydrogen. The opacity of this aerosol is due to the seed particles since the hydrogen is transparent in these cooler zones. The seed material serves to transfer the radiant energy to the hydrogen and to shield the cavity walls from a high heat flux. The particles attenuate the radiant energy by two processes: scattering and absorption. Only the absorption process serves to heat the hydrogen propellant, but the scattering is equally important since it increases the average path length traveled by the photons.

The energy absorption and subsequent heating of the particles depend on the ohmic heating of the particles by conduction electrons that have been excited by the incident radiation.[16] In the two extremes, a perfect insulator and a perfect conductor, no energy would be deposited. In the case of the insulator, the electromagnetic wave would be reflected from the tightly bound electrons and in the case of a perfect conductor, re-emission in the original form would take place because no ohmic heating could occur. Metallic particles are better able to absorb radiant energy because they are conducting

and have significant resistivity.

In addition to absorption, radiant energy is scattered without change in wavelength by the particle. The scattering is the sum of three processes, reflection from the surface of the particle, refraction of light transmitted through the particle, and diffraction due to the interference of the electromagnetic wave as it passes by the edge of the particle. The cross section for diffraction is approximately equal to the geometric cross section of the particle. Since reflection, refraction, and absorption are competing processes on the surface of a particle, the total cross section of these processes is approximately equal to the geometrical cross section. The total interaction of the radiant energy is the sum of the absorption and scattering processes, with a cross section of approximately two geometrical cross sections, and is termed the attenuation or extinction process.

The extinction process as it has just been described is greatest for the case where the diameter of the particle is on the order of the wavelength λ divided by π. Thus, the absorption of radiant energy in the near infrared, visible, and ultraviolet regions of the spectrum is greatest for submicron-sized particles of diameters in the range of 0.05 to 0.5 micron. The scattering from a particle in this size range has a complicated angular dependence. Particles much larger produce highly forward scattering and particles much smaller produce nearly isotropic scattering which is known as Rayleigh scattering.

For particles as large as the wavelength or larger, the cross sections are proportional to the square of the radius, whereas the mass is proportional to the cube of the radius, so the cross sections per unit mass of particle material are inversely proportional to the particle radius. For particles much smaller than the wavelength of the incident thermal radiation, the total attenuation per unit mass falls off rapidly and approaches the value of the absorption-reflection process, which also decreases. In this instance the previous discussion concerning diffraction scattering is no longer applicable, since the diffraction scattering is negligible for such small particles. The very small particles can be totally absorbing (if there is no reflection, e.g. carbon) but the absorption parameter is smaller than that of particles of the optimum size. The steady state radiant energy transport equation for small particles suspended in a transparent medium is

given by,

$$\frac{1}{\rho}\frac{dI(\lambda,\Omega)}{ds} = -\mu_e(\lambda)I(\lambda,\Omega) + j(\lambda) + \mu_s(\lambda)\int_0^{4\pi} I(\lambda,\Omega')p(\lambda,\cos\theta)\frac{d\Omega'}{4\pi}$$

where

I = the intensity of radiant energy in a unit wavelength interval at λ traveling in direction Ω

s = distance

Ω = solid angle

j = emission coefficient

p = scattering amplitude function

θ = angle of scattering

ρ = density of the aerosol.

It is seen from this equation that the four important parameters one must know in order to evaluate radiant heat transfer through particle-seeded gases are:

$\mu_e(\lambda)$ the extinction parameter

$\mu_s(\lambda)$ the scattering parameter

$p(\lambda,\cos\theta)$ the scattering amplitude function

and $j(\lambda)$ the emission coefficient.

These parameters must be known over the applicable temperature, wavelength, and pressure range. In addition, $p(\lambda,\cos\theta)$ should be known for scattering angles from 0 to 180 degrees.

The Mie theory,[17] published in 1908, is the best known and most useful theory which describes the absorption and scattering of electromagnetic radiant energy by particles. It applies to homogeneous spherical particles of any diameter situated in a homogeneous transparent nonmagnetic medium. Mie solved Maxwell's equations with the appropriate boundary conditions and evaluated the total scattered energy as well as the total energy removed from an incident beam, thus arriving at the scattering and extinction cross sections, σ_s and σ_e.

Krascella[18] applied a transformation procedure developed by Aden[19] to the Mie equations to calculate the effect of particle size, wavelength, and particle temperature on particle opacity in those regions of the ultraviolet, visible, and infrared spectra for which complex index of refraction data were

available. Shenoy[12] used Krascella's program to extend these calculations to other types of particles and to a broader wavelength range.

Svatos[20] published a solution to Maxwell's equations for extinction by flattened ellipsoids; however, at present there is no theory that accurately predicts absorption and scattering characteristics of irregularly shaped particles. Submicron-sized particles of refractory materials are generally highly irregular in shape, so the Mie theory can only be used as an approximation to the absorption and scattering characteristics of these particles.

Since the Mie theory calculates the extinction cross section, σ_e, and the scattering cross section, σ_s, the absorption cross section is given by $\sigma_a = \sigma_e - \sigma_s$, and the extinction, absorption, and scattering parameters are given by

$$\mu_e = \frac{\sigma_e}{\rho_p V}, \qquad \mu_a = \frac{\sigma_a}{\rho_p V}, \qquad \text{and} \qquad \mu_s = \frac{\sigma_s}{\rho_p V}$$

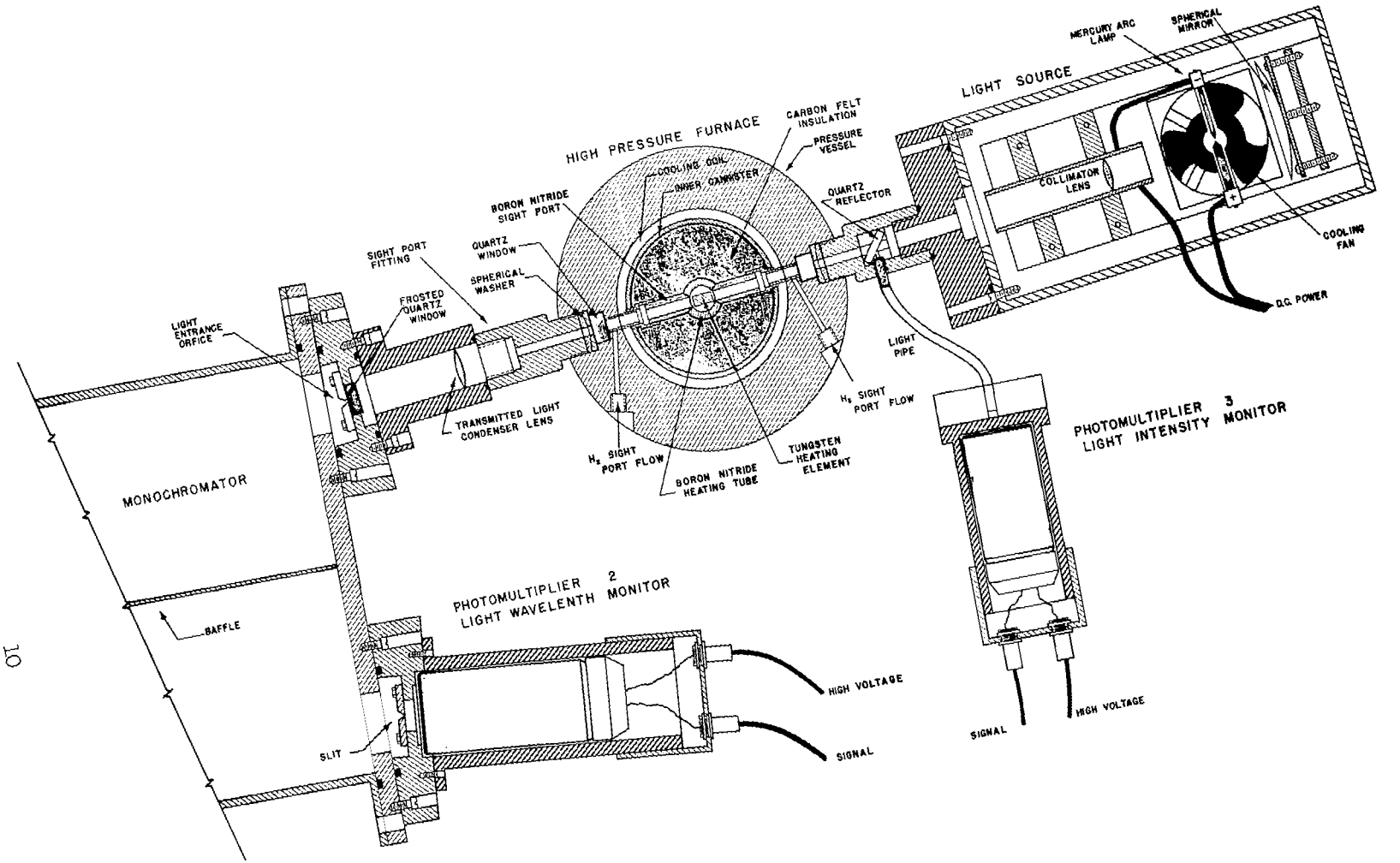
where ρ_p is the mass density of the particle material and V is the volume of the spherical particle.

Mie's solution, though derived for a single sphere, also applies to absorption and scattering by any number of spheres, provided that they are all of the same diameter and composition and provided also that they are randomly distributed and separated from each other by distances that are large compared to the particle radius. Under these circumstances there are no coherent phase relationships between the light that is scattered by the different spheres, and the total scattered energy is equal to the energy scattered by one sphere multiplied by their total number. Similarly, for a distribution of sizes, the energy scattered by the spheres of each particular size may be summed to obtain the total scattered energy.

INSTRUMENTATION

The experimental apparatus is designed to measure the transmission of light through a heated and pressurized aerosol and to measure simultaneously the particle density of the aerosol. The objective is to measure the extinction parameter of the aerosol as a function of wavelength of the incident radiant energy. This is accomplished by passing a beam of radiant energy through the furnace containing the hydrogen aerosol and using a monochromator to record the transmitted beam intensity as a function of wavelength. The transmitted intensity is first measured with only the hydrogen gas present and then measured again after a seed material has been introduced into the hydrogen at the same temperature and pressure. The ratio of the two measured intensities gives the attenuation. By measuring the particle density of the aerosol simultaneously with the intensity measurements the extinction parameter (in cm^2/gm) can be calculated.

Figure 1 is a schematic of the apparatus through which the radiant energy beam traverses. The light source is a mercury arc lamp which produces a focused beam of ultraviolet and visible light which passes through the sight port of the furnace and through the aerosol column flowing up through the center boron nitride tube in the furnace. Experience has shown that operating the arc at 75 watts instead of its 100 watt rating increases the stability of the arc. The intensity of the beam of light transmitted through the furnace is two orders of magnitude greater than the highest thermal emission from the hot furnace. Photomultiplier 3 monitors a reflection of the incident beam of light from a quartz window placed at 45 degrees in the path of the beam. This signal is used to correct the signals from the other photomultiplier tubes for fluctuations in the light source intensity. A quartz lens in the arc lamp focuses the beam of light down to approximately one-quarter inch diameter as it passes through the center of the furnace. Another quartz lens in the sight path on the other side of the furnace focuses the transmitted beam onto a diffusing screen and through a pinhole into the monochromator. The effect of this lens and diffusing screen is to optically integrate spatial intensity variations resulting from refraction by the dense, turbulent hydrogen. Light passing through the pinhole strikes the diffraction grating assembly at the far end of the monochromator. The



...romator Assembly - Furnace End

diffracted light of a selected wavelength is focused by the grating onto the slit, and the intensity of light passing through the slit is monitored by photomultiplier 2. A system of four baffles in the monochromator prevents reflections from the incoming beam from interfering with the diffracted light. Part of the beam incident upon the diffraction grating strikes a set of mirrors which direct this portion of the beam to a photomultiplier tube at the far end of the monochromator. This signal is used to correct for the effects of variations in the aerosol density on the signal being recorded by photomultiplier 2.

The pressure chamber of the furnace (Figure 2) is constructed from mild steel tubing with a 7 1/2 inch outside diameter and a 4 1/2 inch inside diameter. The interior length is 18 1/4 inches. Both ends are capped with two inch thick steel plates secured by eight cap screws. The 1 1/2 inch wall thickness far exceeded the strength needed for a 1500 psi working pressure but was used for ease in designing and machining the through-the-wall fittings.

The aerosol generator that has been used to obtain the high pressure tungsten data is shown in Figure 3. The design is a modified version of the Wright Dust Feed Mechanism developed by Wright.[21] The scraper head is supported on a one-quarter inch OD pipe that is secured to the base and through which the aerosol is carried out. The gas enters through a hollow pinion shaft surrounding the outlet pipe. The hollow pinion shaft is labeled "inner pinion" on the figure. The "outer pinion shaft," which is threaded, rides on the inner pinion shaft and a nut on the top of the inner pinion shaft prevents the pressure from lifting the outer pinion shaft up the inner pinion shaft. Both the base of the seed chamber and the threaded outer pinion are gear driven, but gear ratios of the two are slightly different so that, for approximately every five revolutions of the outer pinion and the seed material chamber, the seed material chamber has advanced one thread down on the threaded outer pinion. This slow advance of the seed material in the chamber onto the fixed scraper head while the two are rotating with respect to each other provides a gradual scraping of the seed material which is carried away by the gas passing up through the inner pinion, over the scraper head, and down through the scraper head support pipe. The seed material must be packed so that it will remain in an inverted position

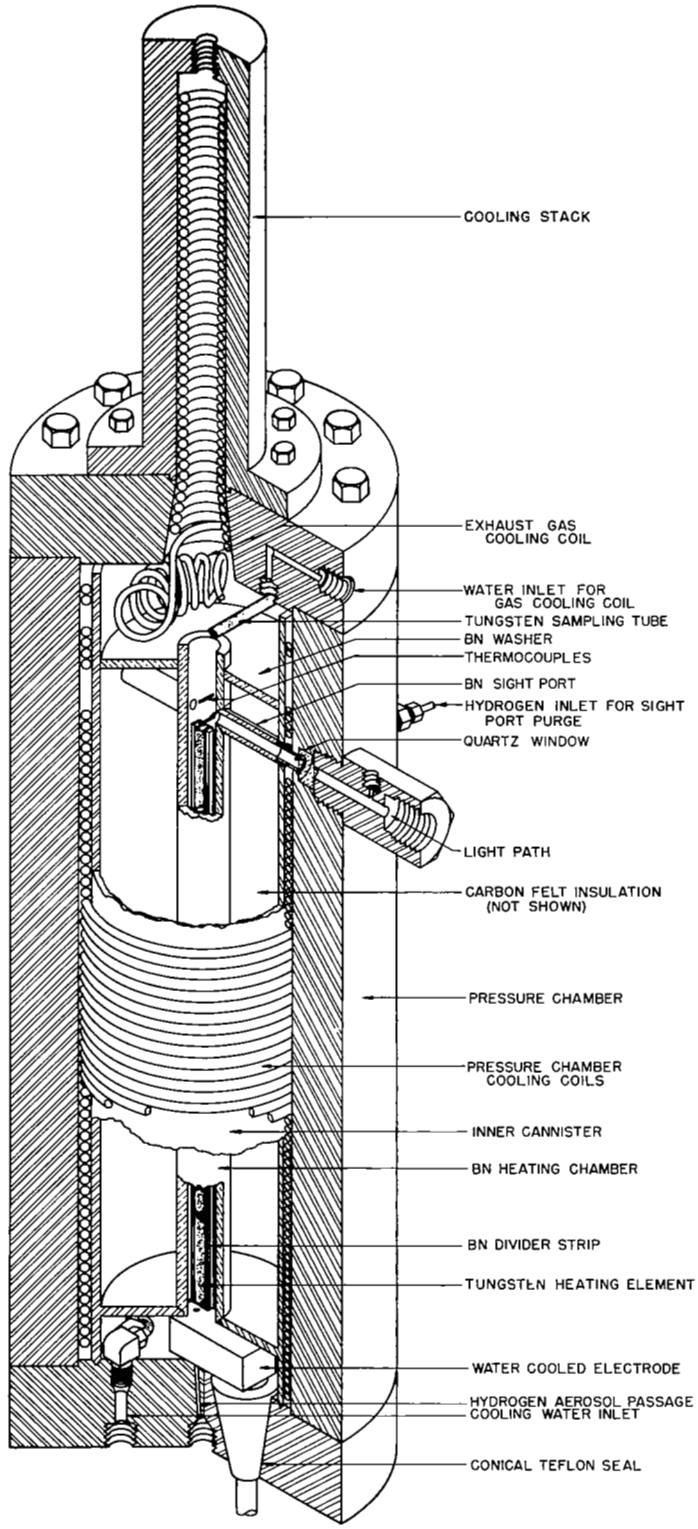


Figure 2. High Pressure Furnace

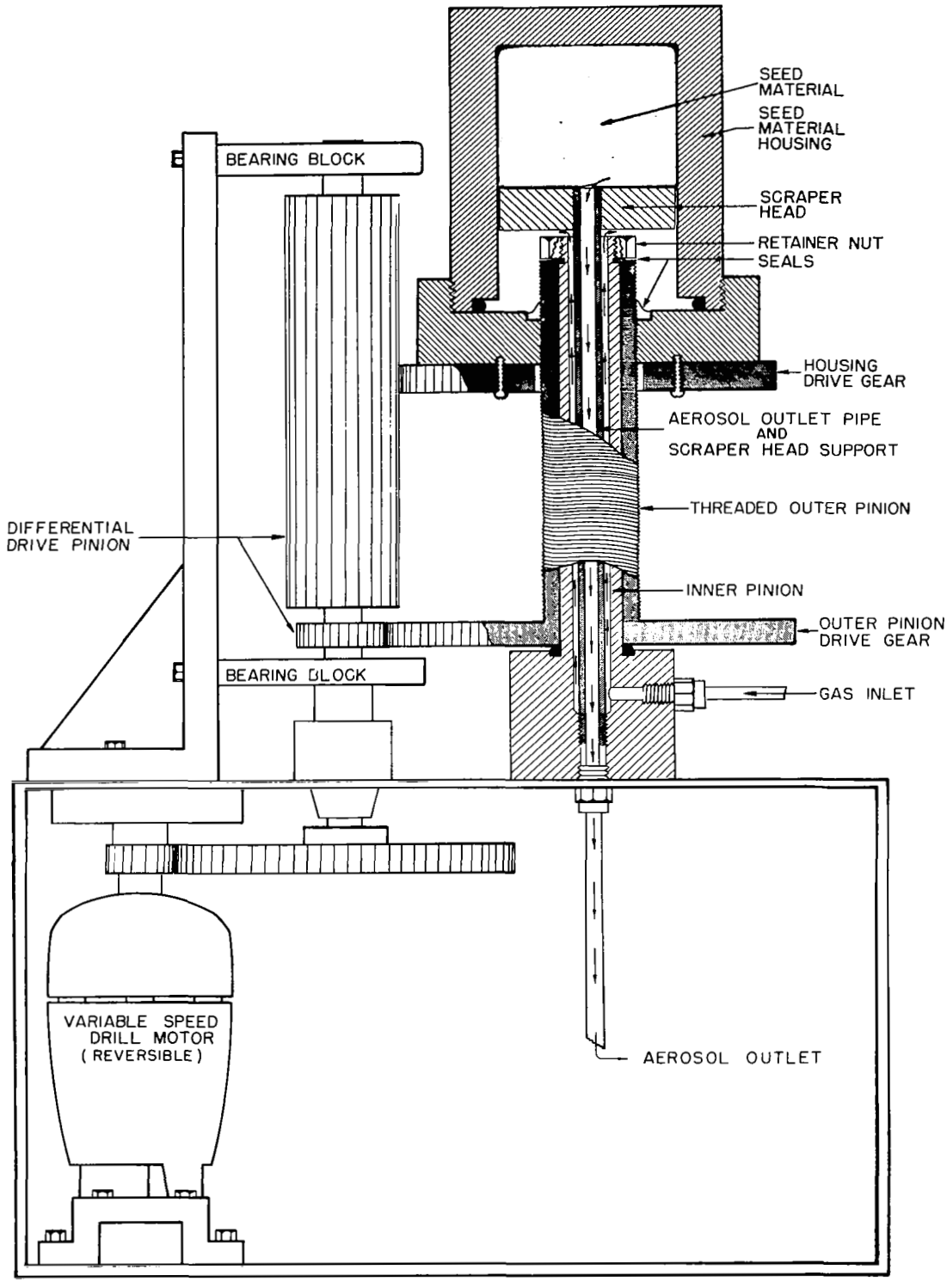


Figure 3. High Pressure Wright Dust Feed Aerosol Generator

until scraped off. Tungsten is ideal in this respect. The real advantage of this aerosol generator is the positive entrainment of seed material in the aerosol lines even at low volume flow rates. Plus, the device can be operated at variable speeds to provide the desired seed density.

The aerosol generator will produce a more highly dispersed aerosol if the aerosol is passed through a nozzle. A pressure upstream of twice that downstream is needed to insure sufficient shear forces on the aerosol. This is difficult to achieve at the higher working pressures because of the limited supply pressure available and therefore prevented the use of a nozzle at high pressures.

At the bottom of Figure 2 can be seen one of the water-cooled copper power leads and its conical Teflon seal. The pressure inside the chamber produces a leak-tight seal by acting on the conical base of the power lead and its seal. The hydrogen aerosol enters a fitting at the base of the furnace and splits into two streams as it passes through the vee-shaped passages machined into the bottom plate of the furnace. One of the two passages is shown in the figure. A hole in each of the power leads is aligned with one of the inlet passages and the aerosol passes into the boron nitride heating chamber. The heating chamber is a 15 inch long tube with a one inch outside diameter and a five-eighths inch inside diameter. The heating element is a strip of tungsten which is separated by a one-eighth inch strip of boron nitride at the bottom. The tungsten strip is nominally 28 inches long, one-half inch wide, and 50 mils thick. The boron nitride divider strip shown in Figure 2 has been modified so that it extends only half way up the tungsten strip, thereby improving flow conditions in the furnace. The power supply for the furnace is rated for 20 Kw direct current. Just above and just below the sight path are tungsten-three percent rhenium and tungsten-26 percent rhenium thermocouples in boron nitride shields. At the top of the boron nitride heating chamber is located a boron nitride sampling tube through which a sample of the aerosol is withdrawn for density measurements.

The size distribution of aerosols is important when measuring the extinction parameter. The approach used in this research to measure the size distribution was to sample the aerosols and make electron micrographs which show the degree of agglomeration, the sizes of the agglomerates, and the size dis-

tribution. Tests with the electrostatic device (Figure 4) used for depositing the particles on electron microscope grids showed that a random collection of particles was obtained.

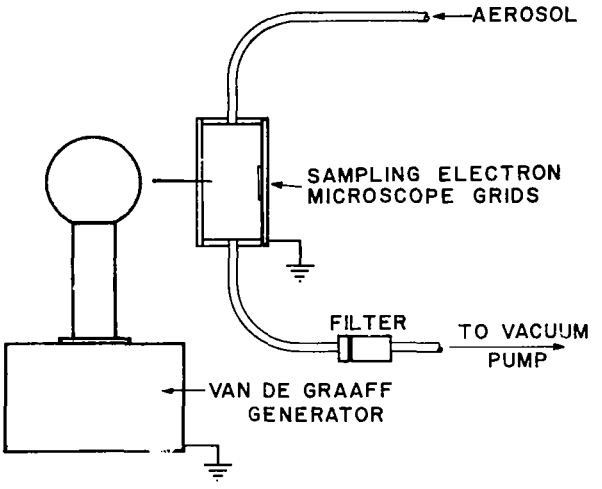


Figure 4. Electrostatic Sampling Device

Above the sampling tube the aerosol passes through 12 inches of copper cooling coils as it travels up the cooling stack. The gas is cooled as it travels up the cooling stack and then it goes to the seed filter where the seed material is recovered by passing the aerosol, still under pressure, through a 4.7 cm diameter fiberglass filter. The hydrogen passes through a throttling valve which holds the pressure in the furnace system to the desired operating pressure. The hydrogen leaving the throttling valve is passed through a flowmeter and then exhausted to the atmosphere.

The signals from the three photomultiplier tubes are recorded simultaneously by an oscillograph and a tape recorder. The output from the thermocouples in the furnace is displayed by a digital voltmeter and recorded manually. The pressure in the furnace, and the pressure changes in the density volume measurement tanks (as indicated by manometers), are also recorded manually.

There are two sampling systems used to simultaneously measure the concentration of seed material in the hydrogen. One sample is taken at the entrance of the furnace and the other sample is taken at the top of the boron nitride heating chamber. These two samples are used to study the effect of temperature on the reaction or vaporization of the seed.

Temperature measurements are made with shielded tungsten three percent rhenium versus tungsten 26 percent rhenium thermocouples. These thermocouples are capable of operation in hydrogen at 2500°K. The temperature is measured immediately below and immediately above the light beam with two thermocouples.

DATA REDUCTION AND ANALYSIS

Three signals are recorded by the oscillograph. The first is the intensity of the focused beam of light from the mercury arc (I_L) monitored by photomultiplier 3 (Figure 1). The second signal recorded (I_H) is the intensity of light transmitted through the aerosol in a narrow portion of the ultraviolet spectrum which has passed through the aerosol. This is necessary in order to correct for variations in the aerosol density. The third signal is the wavelength dependent signal (I_w) from photomultiplier 2. A superscript $\underline{B}$ is added to denote that the intensity was measured before particles were introduced into the furnace and the superscript $\underline{P}$ is added when the signal was measured with particles present.

The density of the aerosol changes with time, while the intensity of the signal depends on the wavelength. Since the grating is driven by a motor at a constant speed, it is convenient to use wavelength, instead of time, as the independent variable for the changing aerosol density.

The extinction parameter $\mu_e(\lambda)$ is given by

$$\mu_e(\lambda) = \frac{K(\lambda)}{\rho(\lambda)}$$

where $K(\lambda)$ is the linear attenuation coefficient and $\rho(\lambda)$ is the density of the aerosol at that time when photomultiplier 2 is observing light of wavelength λ. The calculation of $\mu(\lambda)$ involves two separate calculations. One is the calculation of $K(\lambda)$ and the other is the calculation of $\rho(\lambda)$. $K(\lambda)$ is given by the expression

$$K(\lambda) = \frac{1}{X} \ell n \frac{I_w^{B\ corr}}{I_w^{P\ corr}}$$

where X is the aerosol path length. The superscript "corr" means that I_L has been used to correct the I_w signal for variations due to fluctuations in the light source intensity.

The average aerosol density (ρ_{av}) is obtained by drawing a sample of the aerosol at a constant rate during the spectrum scan. The instantaneous aerosol density, $\rho(\lambda)$, is then calculated from the signal I_H using the equa-

tion

$$\rho(\lambda) = \frac{\rho_{av} L}{\sum_{i=1}^{N} \ell n \frac{I_H^{B\ corr}}{I_H^{P\ corr}(\lambda_i)} \Delta\lambda_i} \cdot \ell n \frac{I_H^{B\ corr}}{I_H^{P\ corr}(\lambda)}$$

where $L = \sum_{i=1}^{N} \Delta\lambda_i$

and $\Delta\lambda_i$ is chosen small enough so that ρ does not change significantly while the wavelength λ observed by photomultiplier 3 changes by $\Delta\lambda_i$.

In order to evaluate the error in $\mu_e(\lambda)$ the percentage of uncertainty in the measurements of I_w, I_H, I_L, and ρ is estimated and the resulting error is calculated as a function of λ. The error in the light intensity measurements is due, principally, to the accuracy to which the intensity levels can be measured from the oscillograph recording. The estimated error in ρ reflects the accuracy with which a sample of aerosol can be collected and weighed.

The analysis equations are derived and discussed in detail in reference 22.

RESULTS

The tungsten seed material was obtained as a powder of submicron-sized particles. The electron micrographs in Figure 5 illustrate the sizes and shapes of these particles as they were collected from the aerosol. The upper electron micrograph was taken of an aerosol that had been formed by passing it through a nozzle. The lower electron micrograph is of an aerosol formed without using a nozzle and exhibits less dispersion. Most of the particles in the well-dispersed aerosol are in the size range between 0.1 and 0.2 micron while the particles in the less-dispersed aerosol are between 0.2 and 0.5 micron. The well-dispersed aerosol was used in measuring the extinction parameter at pressures from 1 to 12.5 atmospheres. The data at higher pressures are measurements of an aerosol formed without a nozzle, so the particle agglomerates were similar to those in the lower electron micrograph. The difference in average particle sizes produced a corresponding difference in the measured extinction parameters as predicted by Mie theory.

The principal sources of error in the data resulted from dilute aerosols and the difficulty in obtaining an accurate density measurement. The scatter in the values of the extinction parameter at different wavelengths, during the same operating conditions, was greater for less dense aerosols. The fact that the extinction parameter varied as much as 20 percent for several measurements at the same operating conditions can be attributed to these errors. The inherent difficulties in obtaining an accurate density measurement were due to the disturbance of the flow pattern in the heating chamber caused by sampling and the deposition of seed material on the sampling tube walls as the result of coagulation, thermal precipitation, and gravity induced precipitation. An increased sampling rate tended to reduce the precipitation rate but increased the disturbance of the aerosol flow in the heating chamber. The optimum sampling rate was a compromise to minimize the sum of the two effects.

No chemical reaction between the hydrogen and the tungsten was expected or observed. One of the beneficial effects of using hydrogen as the carrier gas was more efficient heat transfer from the heating element to the hydrogen, and in turn, to the particles. When the heating element was above approximately 1500°K, the temperature would increase about five percent when particles

With Nozzle Dispersion

1.0 Micron

Without Nozzle Dispersion

Figure 5. Electron Micrographs of Tungsten Particles with and without Nozzle Dispersion

were added to the gas, indicating that the particles were aiding the heat transfer process by absorbing the radiant energy and further heating the hydrogen.

The extinction parameters of tungsten-hydrogen aerosols were measured over a range of temperatures in five pressure regimes: 12, 45, 70, 100, and 115 atmospheres. In addition, the extinction parameter data for tungsten-hydrogen aerosols at one atmosphere from reference 12 are used for showing trends due to pressure.

Figures 6 through 10 are plots of the extinction parameter data as a function of wavelength for various operating points. As a comparison, the theoretical values of the extinction parameter calculated from Mie theory are plotted for four particle diameters (0.02, 0.1, 0.2, and 0.5 micron). The data exhibiting a great deal of scatter in the values of the extinction parameter for different wavelengths are characteristic of measurements made with a dilute aerosol. The spectrum averaged value of the extinction parameter for room temperature data at 1 and 12.5 atmospheres is about 20,000 cm^2/gm. Since these data are for a nozzle-dispersed tungsten aerosol, they represent the extinction parameter for tungsten particles in the size range 0.1 to 0.2 micron discussed in the previous section. The room temperature data for higher pressure tungsten-hydrogen aerosols were not generated with a nozzle so the particles were not as well dispersed. The spectrum averaged value of the room temperature extinction parameter at high pressures is about 8,000 cm^2/gm. This is approximately two-fifths the value obtained for the nozzle-dispersed aerosols measured at one and 12 atmospheres. An examination of electron micrographs of these particles (Figure 5) indicates that the average particle diameter for well-dispersed aerosols is approximately 2.5 times smaller than for aerosols formed without using a dispersion nozzle. This is significant in that the room temperature values of the extinction parameter of the well-dispersed aerosols are about 2.5 times greater than for the less-dispersed aerosols, as is predicted by the Mie theory. Since the difference between the room temperature extinction parameter data at low and high pressures can be accounted for by the change in the average particle diameter, there is no observed pressure dependence of the extinction parameter at room temperature.

A closer examination of Figures 6-10 indicates that there is a slight

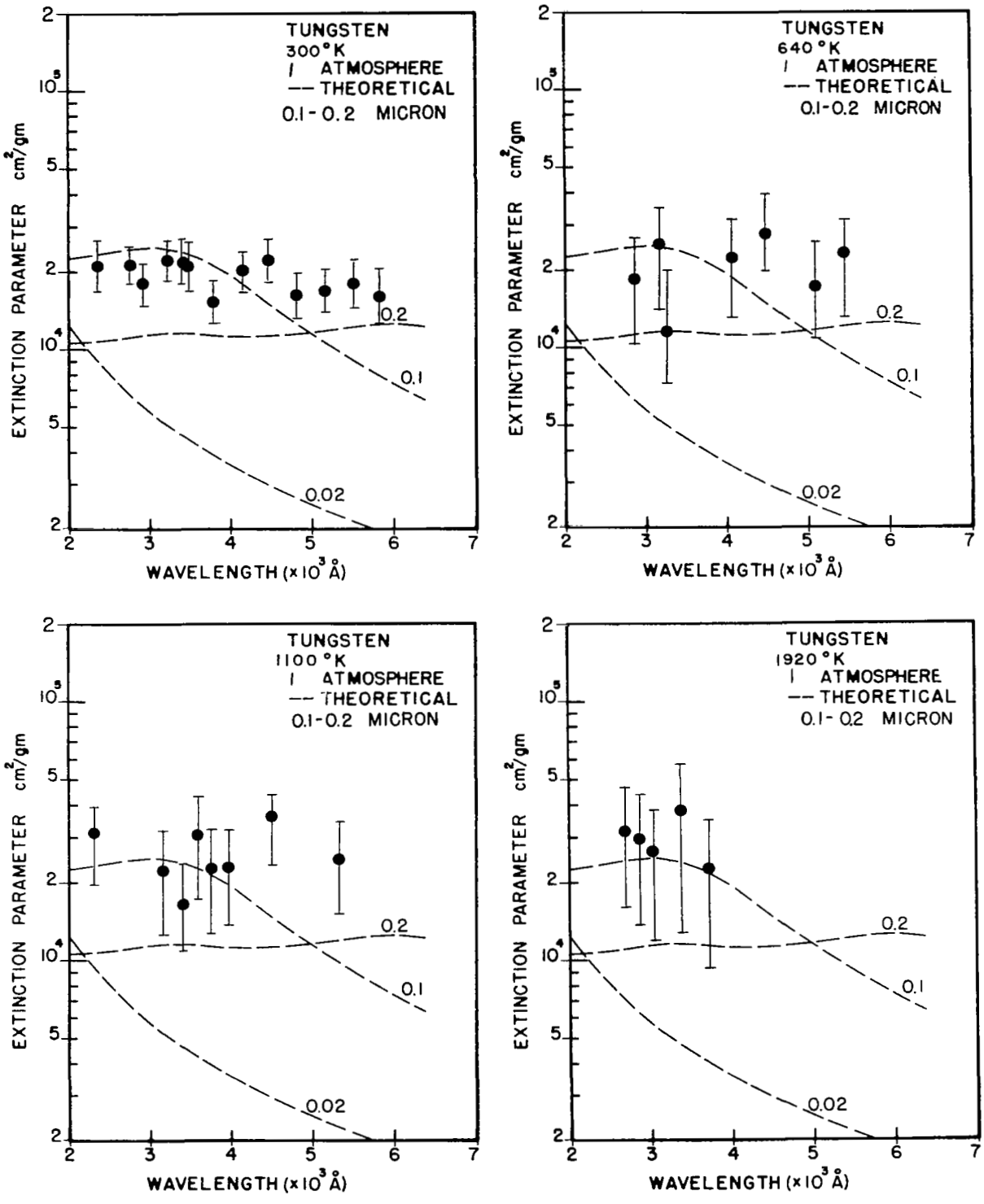


Figure 6. Extinction Parameter of Tungsten-Hydrogen Aerosols at One Atmosphere Pressure (data from reference 12)

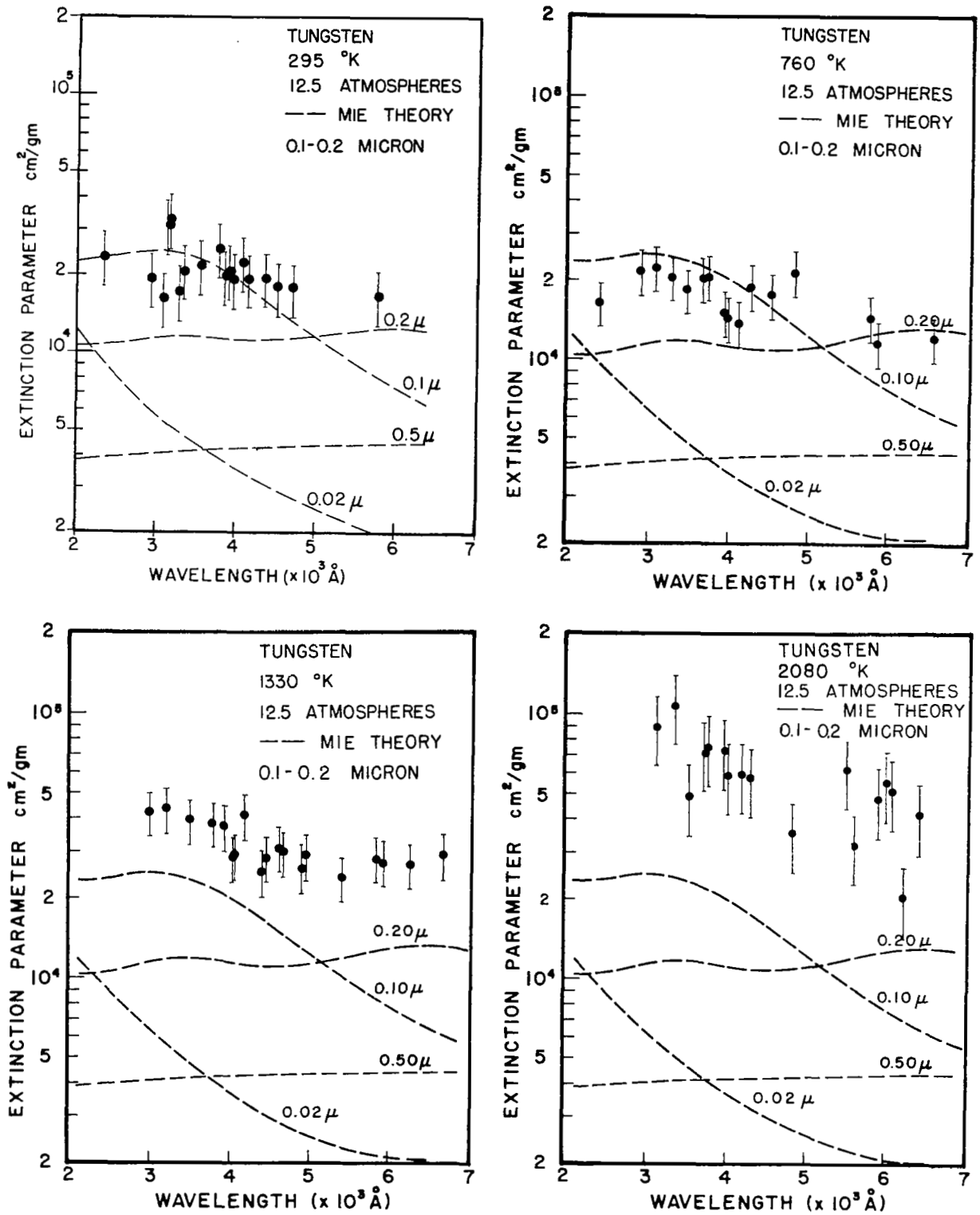


Figure 7. Extinction Parameter of Tungsten-Hydrogen Aerosols at 12.5 Atmospheres Pressure

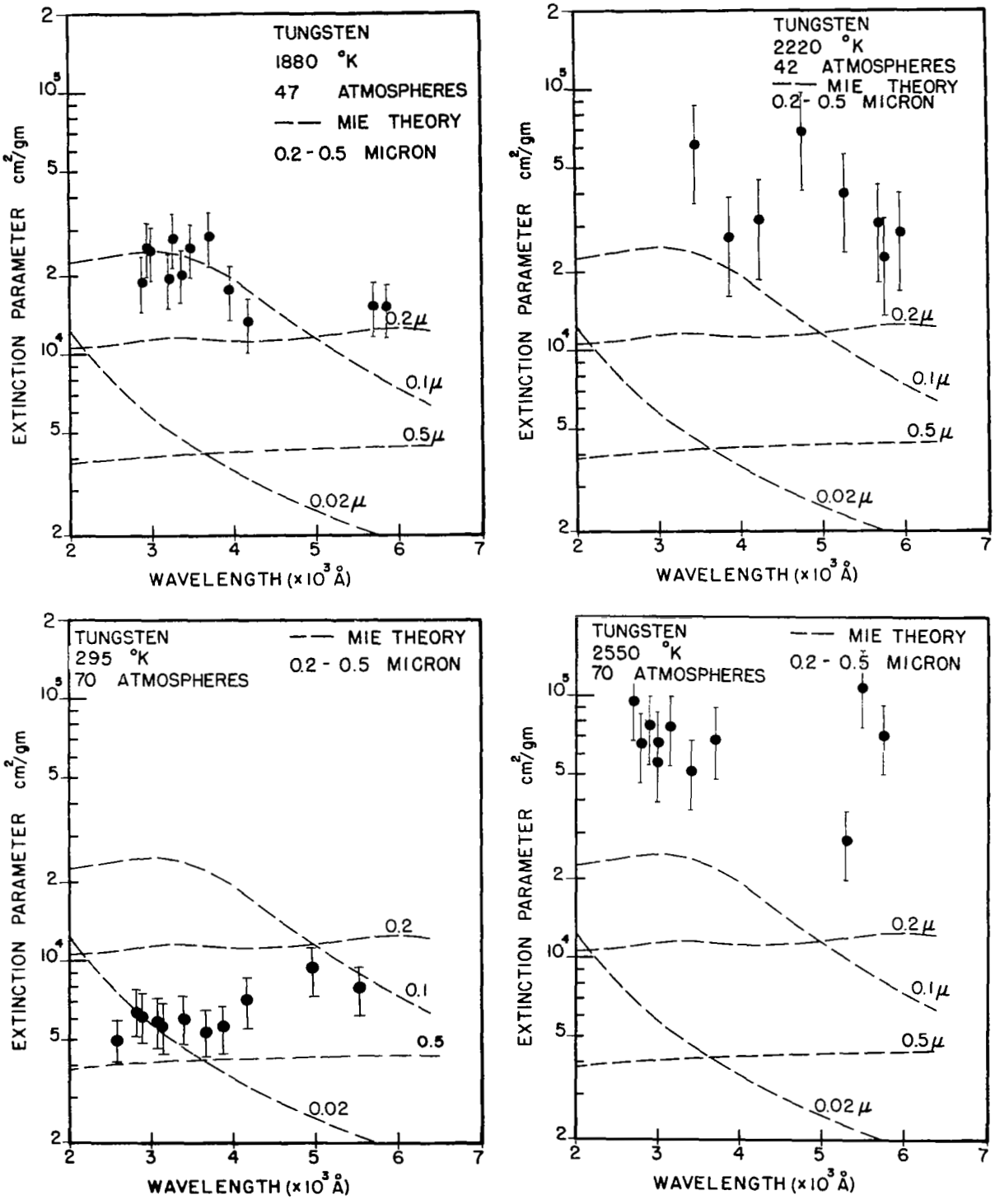


Figure 8. Extinction Parameters of Tungsten-Hydrogen Aerosols at 42, 47, and 70 Atmospheres Pressure

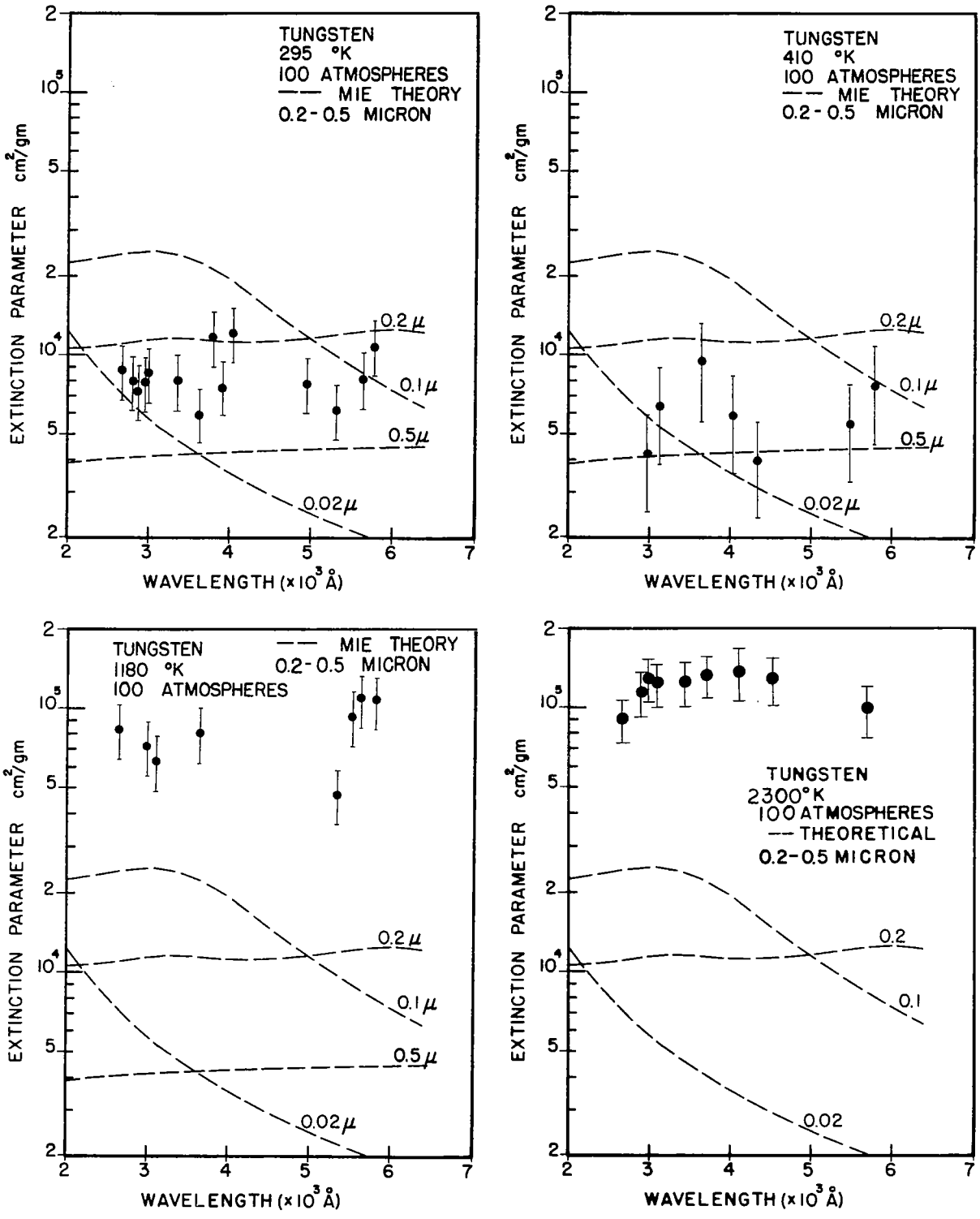
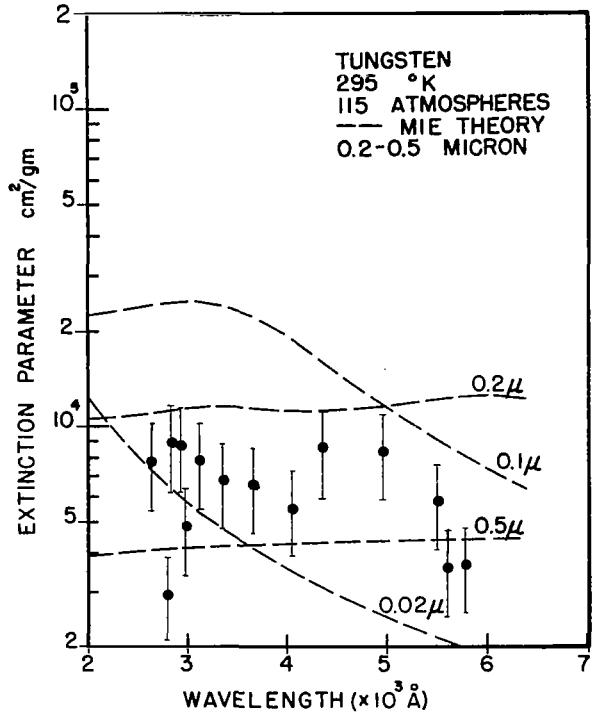
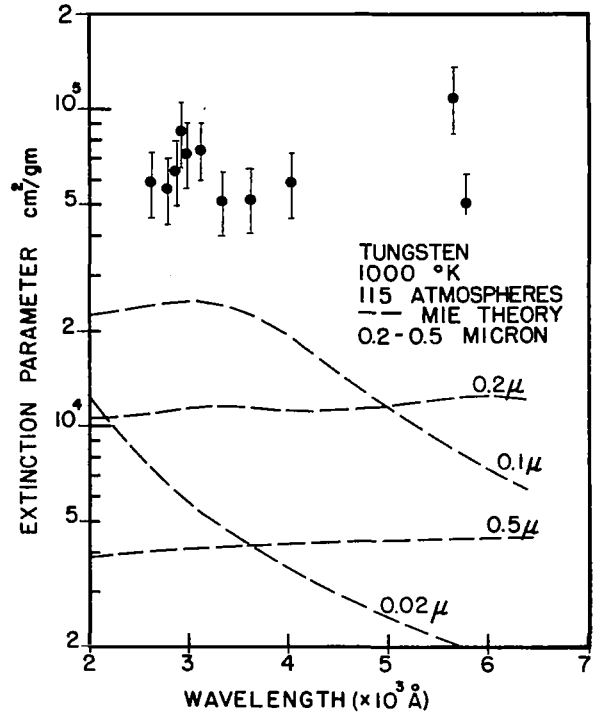


Figure 9. Extinction Parameter of Tungsten-Hydrogen Aerosols at 100 Atmospheres Pressure





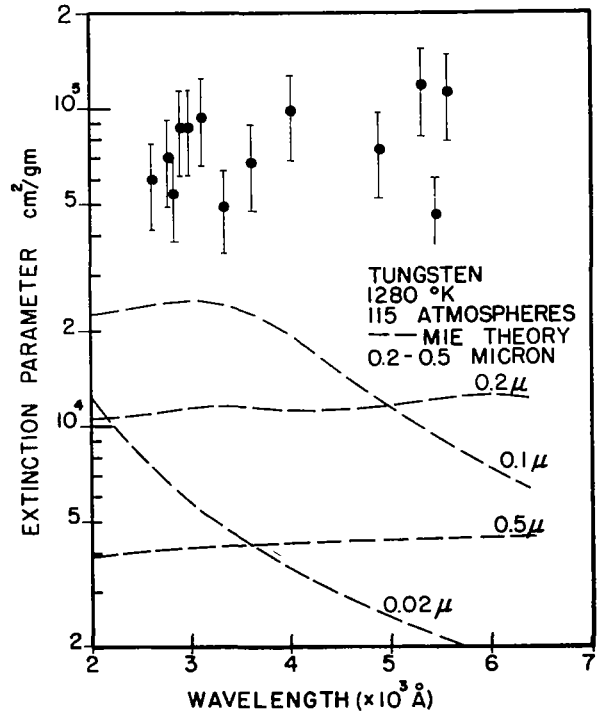


Figure 10. (Above and Lower Left) Extinction Parameter of Tungsten-Hydrogen Aerosols at 115 Atmospheres Pressure

wavelength dependence in that the values of the extinction parameter are generally higher in the ultraviolet region than in the green to yellow portion of the spectrum. In order to examine the dependence, the spectrum over which the measurements were made, 2500 Å to 5800 Å, was divided into three sections and the extinction parameter values for the individual wavelengths in each section were averaged. The three sections were chosen as follows: 2500 Å to 4000 Å (ultraviolet), 4000 Å to 4900 Å (blue and violet), and 4900 Å to 5800 Å (green and yellow). The average values obtained were plotted

as a function of temperature for different pressure regimes in Figures 11 and 12. The wavelength dependence is apparent since the ultraviolet and blue-violet averaged values are usually higher than the green-yellow averaged values.

Other trends become apparent in the data plots of Figures 11 and 12. The values of the extinction parameter increase with temperature and the rate of increase appears to be pressure dependent. Shenoy[12] concluded that the tungsten-hydrogen aerosol opacity at one atmosphere pressure was not temperature dependent since the variation in magnitude of the extinction parameter as a function of temperature was about the same as the estimated error associated with the measurements. A review of the one atmosphere data for tungsten-hydrogen aerosols for a correlation with the high pressure, high temperature data, showed that a slight increase with temperature can be observed.

The rate at which the extinction parameter increases with temperature becomes larger as the pressure increases as is illustrated by Figures 11 and 12. The extinction parameter at room temperature is about 2.5 times smaller at 40 to 115 atmospheres than it is at one and 12.5 atmospheres. This difference is explained in the previous section as being the result of the decreased tungsten particle size when the dispersion nozzle is used at the lower pressures. As the temperature of the high pressure aerosol approaches 1000°K, a significant increase in the extinction parameter is observed and, at 100 atmospheres and 2300°K, the extinction parameter has increased 15 times over its value at room temperature. A partial explanation for this phenomenon is a reduction in the size of the agglomerated particles. It is possible that agglomerated particles could receive enough energy from particle-to-particle and gas-to-particle collisions to cause partial fragmentation. Inspection of the tungsten particles in the electron micrographs of Figure 5 indicates that the particles are really agglomerates which consist of individual particles as small as 0.01 micron in diameter. However, the optimum particle size for attenuating the radiant energy used in this experiment is between 0.1 and 0.2 micron diameter. The Mie theory predicts that a further reduction in the particle size would result in a decrease in the extinction parameter. The theoretical extinction parameter for a 0.1 micron diameter tungsten particle at 2400 Å wavelength is 20,000 cm^2/gm.

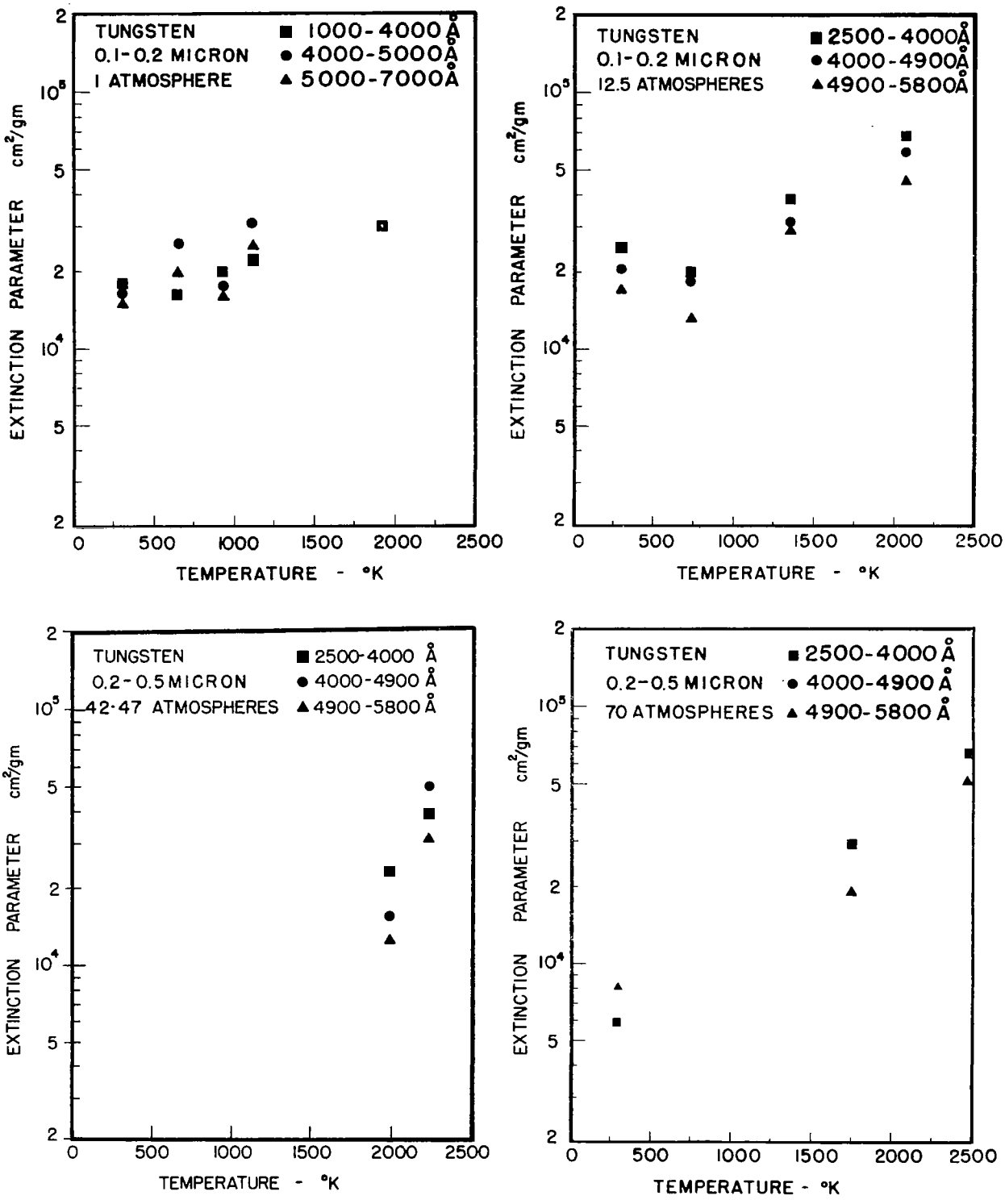


Figure 11. Spectrum Averaged Extinction Parameters of Tungsten-Hydrogen Aerosols as a Function of Temperature at Pressures from 1 to 70 Atmospheres

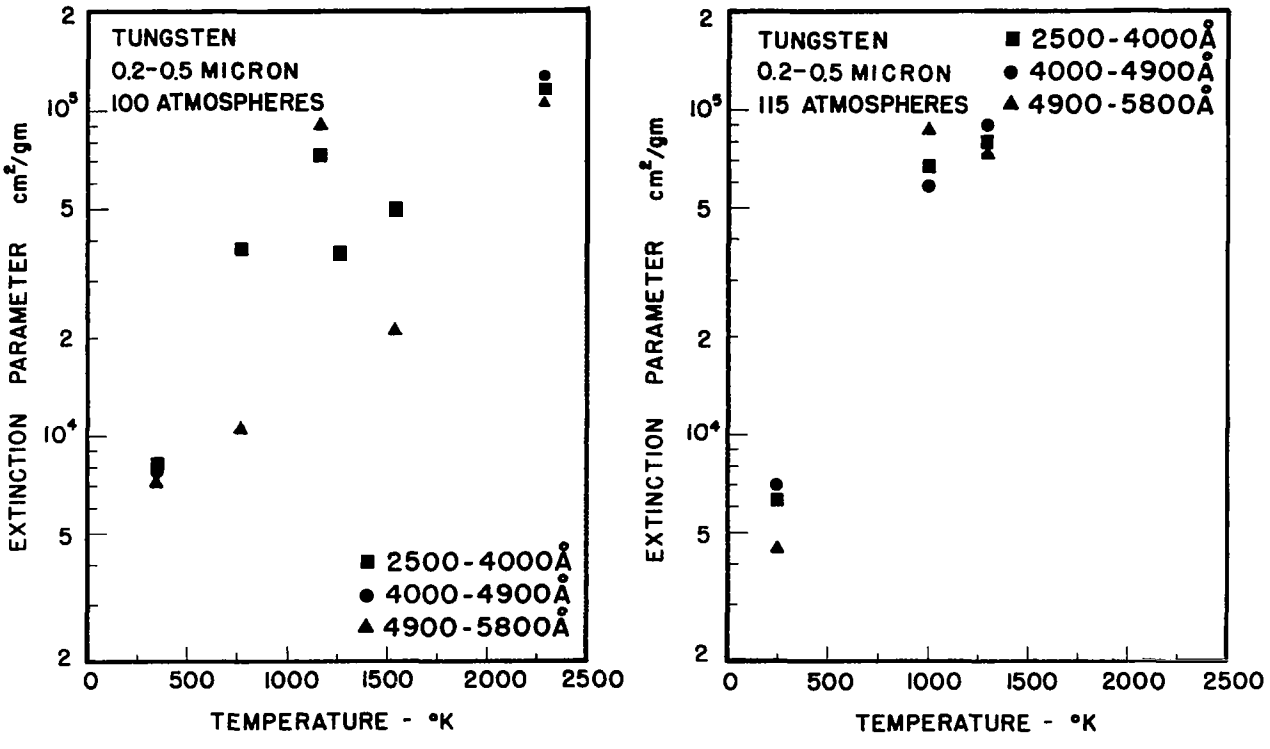


Figure 12. Spectrum Averaged Extinction Parameters of Tungsten-Hydrogen Aerosols as a Function of Temperature at Pressures of 100 and 115 Atmospheres

Even if the optimum particle size were being formed, there still exists a factor of five increase in the extinction parameter that is not explained by the Mie theory.

Cory and Bennett[23] measured a decrease in light transmitted through a refractory particle cloud at high temperatures and moderate pressures. The decrease in transmitted light was more significant as the pressure was raised. Their experiment consisted of heating a tantalum carbide-hydrogen aerosol in a quartz tube by exposing the tube to the focused light from a xenon flash-lamp. As a result, the temperatures and pressures obtained were very transient conditions, lasting a few milliseconds. They suspected that thermionic emission from the particles produced a positively charged particle surrounded by a cloud of electrons that were trapped by the potential well of the positively charged particle. The increased absorption by this system was thought to be an inverse bremsstrahlung effect whereby the electrons could capture and emit photons by momentum exchange with the electric field. In order to

verify the existence of the electrons they measured the conductivity of the aerosol and found that it increased to approximately 20 mho/cm at the maximum flash power point.

The literature[24-26] cited by Cory and Bennett does not provide a conclusive description of this process. The concept of inverse bremsstrahlung was used by Visvanathan[24] in describing free carrier absorption in semiconductors. The description of the electron cloud generated by thermionic emission and held in place by the resultant positive charge on a particle is given by Soo.[25] He describes some assumptions that are required before the electron cloud model can be established. First is the consideration of a single particle in a finite volume in order to equate the thermionic current density to the current density of electrons in the cavity. Second, the gas in the cavity is considered only in regard to its first ionizational potential and not with respect to the effects of gas molecule-particle collisions. Soo also states that as much as 80 percent of the potential bound electrons can be set free by turbulence in the gas, indicating that a hot aerosol of metallic particles may have a significant conductivity as shown by Cory and Bennett.[23]

Calculations were made to predict the increase in the extinction parameter due to free electrons. It was found that the effective work function for submicron-sized tungsten particles would probably have to be smaller than the 4.5 eV work function of large tungsten pieces before a significant number of electrons would be produced by thermionic emission at 1000°K to 2000°K. Even after enough electrons are present in the aerosol to increase the conductivity, the calculation of the increase in extinction parameter showed that the cross section for a free electron[26] (6.65×10^{-25} cm^2) is too small to be significant in the overall extinction process.

A closer consideration of the particle-gas system at high temperature and pressure indicates that the concept of a free electron in the system may not be valid. The following parameters apply to an aerosol of 5×10^{-6} gm/cm^3 density formed from 0.1 micron diameter tungsten particles dispersed in 100 atmosphere hydrogen at 1000°K:

Particle diameter	0.1 micron
Particle surface area	3.14×10^{-10} cm^2
Density of particles in aerosol	4.9×10^{8} particles/cm^3

Mean distance between particles	12.6 microns
Density of H_2 molecules	0.8×10^{21} molecules/cm^3
Mean distance between molecules	1.1×10^{-3} microns
Mean free path of H_2 molecules	2.36×10^{-3} microns
Collision frequency between H_2 and particle	1.85×10^{16} collisions/sec-particle
Average translational energy of H_2 molecule	0.129 eV

The hydrogen gas at 100 atmospheres and 1000°K is approximately 1/25 the density of liquid hydrogen. It may be possible for the free electrons in the gas to interact with one or more hydrogen molecules under these conditions so that radiant energy is absorbed by the system. There is an even greater tendency for H_2^- ions to form if there is a high density of electrons surrounding a charged particle. Any absorption or scattering interactions of photons with ions in the aerosol would tend to be pressure dependent, since the number of molecule-particle collisions is pressure dependent.

Another process by which the extinction parameter could be expected to increase with temperature and pressure is the pressure induced broadening of tungsten spectral lines. Krascella[27] has calculated the Rosseland mean opacity for tungsten vapor as a function of temperature and pressure. This process would probably not be applicable unless it could be shown that a significant amount of tungsten atoms exists in the aerosol at temperatures below the vaporization temperature.

More information describing the tungsten-hydrogen aerosol at high pressure and temperature may be necessary before a particular photon absorption-scattering process can be cited with confidence. Knowledge of the electron density and electron distribution would permit a first order analytical study. The extension of the measurement of the extinction parameter to higher temperatures may also provide information that will indicate the predominance of one extinction process over the other possibilities.

CONCLUSIONS

A well-dispersed tungsten-hydrogen aerosol at room temperature was shown to have a spectrum averaged extinction parameter of approximately 20,000 cm^2/gm. Electron micrographs of this aerosol showed the average diameter of the tungsten particle agglomerates to be between 0.1 to 0.2 micron. In order to achieve this degree of dispersion the aerosol was passed through a nozzle after mechanical agitation had mixed the tungsten agglomerates with the hydrogen carrier gas.

Since it was difficult to use a nozzle in the aerosol inlet tube at high pressures, the aerosol was formed by mechanical agitation only. The diameters of the tungsten particle agglomerates formed by this method were mostly in the range of 0.2 to 0.5 micron. A spectrum averaged extinction parameter of approximately 8,000 cm^2/gm was measured for the room temperature, high pressure aerosol. The reduction in the extinction parameter at high pressures and room temperature was the result of the larger particle diameter, which is predicted by Mie theory.

A plot of spectrum averaged extinction parameters as a function of temperature for aerosols at a given pressure showed an increase of the extinction parameter with temperature at temperatures above about 1000°K. The rate at which the extinction parameter increased became larger as the pressure of the aerosol increased. A spectrum averaged extinction value of 120,000 cm^2/gm was measured for tungsten-hydrogen aerosol at 100 atmospheres pressure and 2300°K, whereas the value for the same aerosol was 8,000 cm^2/gm at room temperature.

Partial deagglomeration of the tungsten particles can only account for about 20 percent of the increase in the extinction parameter. Physical processes which have been considered as possible explanations for the increase are the formation of an electron cloud around a thermionically emitting particle[23,25] (photon absorption by inverse bremsstrahlung[24]), the interaction of photons with an electron associated with one or more H_2 molecules, and pressure induced broadening of tungsten atom spectral lines.[27] The complexity of the high temperature aerosol system prevents any particular process from being cited at this time as a dominant factor in the pressure and temperature dependent extinction parameter. The photon scattering by free electrons alone

has been discounted as a possible explanation for the increase in extinction parameter with temperature and pressure. More information is needed on the electron density and electron distribution and their interactions in order to proceed with an analytical study of the relative importance of each photon interaction process. The measurement of extinction parameters at higher temperatures than those reported here may provide useful insight into the nature of the process responsible for a pressure and temperature dependent extinction parameter.

The heat transfer in the gas core nuclear rocket will probably be enhanced by the pressure and temperature trends in these data. Since the extinction process is the sum of the absorption and scattering processes, it is not apparent whether the increase in the extinction parameter is due to an increase in only one or both processes. An increase in either process will tend to reduce the percentage of seed material required in the propellant, but to different degrees.

The seeded aerosol in the actual gas core reactor will exhibit an additional photon absorption process, photoemission, when exposed to the extremely intense photon field from the uranium core. The extremely high levels of ionizing radiation present in the reactor will also contribute to ionization in the propellant.

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