NASA Contractor Report 3244

Spectral Properties of Gaseous Uranium Hexafluoride at High Temperature

N. L. Krascella

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N. L. Krascella
United Technologies Research Center
East Hartford, Connecticut

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SUMMARY

The present experimental investigation was undertaken to provide spectral data for uranium hexafluoride (UF_6) - argon mixtures at temperatures and wavelengths of interest to the plasma core reactor (PCR) concept.

An experimental study was conducted to determine relative spectral emission and spectral absorption data for UF_6 - argon mixtures at elevated temperatures. These spectral data are required to assist in the theoretical analysis of radiation transport in the nuclear fuel-buffer gas region of a PCR. Current spectral data for UF_6 do not cover the wavelength range between approximately 600 to 5000 nanometers. In addition, no data exist at elevated temperatures of interest to the PCR concept.

Relative emission measurements were made for UF_6 - argon mixtures over a range of temperatures from about 650 to 1900 deg K and in the wavelength range from about 600 to 5000 nanometers. All emission results were determined for a total pressure of 1.0 atm. Uranium hexafluoride partial pressures varied from about 3.5 to 12.7 mm Hg.

Absorption measurements were attempted at 600, 625, 650 and 675 nanometers for a temperature of 1000 deg K. The uranium partial pressure for these determinations was 25 mm Hg.

Generally, the results exhibit appreciable emission for hot UF_6 - argon mixtures at wavelengths between 600 and 1800 nanometers. The relative emission increases strongly with temperatures. At wavelengths greater than about 1800 nanometers, essentially no significant emission was observed at all temperatures.

No measurable absorption was observed at wavelengths between 600 and 675 nanometers for UF_6 at a temperature of 1000 deg K. The absorption determinations indicate cross sections of less than 10^{-22} cm^2.

INTRODUCTION

Extensive experimental and theoretical research has been conducted with respect to various aspects of fissioning gaseous uranium plasmas over the past two decades. These studies have been generally directed toward high performance nuclear space propulsion systems (Ref. 1). In addition to space applications, the PCR concept has been recognized as a possible candidate energy source for various terrestrial applications (Ref. 2). The very high operating temperatures generated by fissioning plasmas tend to enhance efficiencies of various thermodynamic cycles compared to conventional power generating systems or nuclear reactors with solid fuel elements. Furthermore, the high operating temperatures associated with gaseous fissioning reactors provide a source of very intense electromagnetic radiation with a number of possible applications involving direct coupling of energy by radiative processes.

Although most of the applications of the PCR require extensive basic research and technological development, the potential benefits from the use of these devices warrant continued investigation of the concept. Possible PCR space and/or terrestrial applications are:

(1) High-thrust, high-specific-impulse space propulsion systems.
(2) Advanced high-temperature, closed-cycle gas turbine electrical power generation.
(3) MHD conversion systems for electrical power generation.
(4) Photochemical and/or thermochemical processing.
(5) Direct pumping of lasers by deposition of fission fragment energy in UF_6 or other lasing gas mixtures.
(6) Optical pumping of lasers by thermal and/or nonequilibrium radiation emitted by a gaseous fissioning UF_6 or uranium plasma.

A typical unit cavity of a PCR device is illustrated in Fig. 1. In the PCR concept a high-temperature, high-pressure plasma is sustained via the fission process in a uranium gas injected as UF_6 or other uranium compound. Containment of the plasma is accomplished fluid-mechanically by means of an argon-driven vortex which also serves to thermally isolate the hot fissioning gases from the surrounding wall.

For applications which employ thermal radiation emitted from the plasma, an internally-cooled transparent wall can be employed to isolate the nuclear fuel, fission fragments, and argon in a closed-circuit flow loop and permit transfer of the radiant energy from the plasma to an external working fluid. For applications which employ fission-fragment-induced short wavelength nonequilibrium radiation emitted from the plasma, the working fluid such as lasing gases can be either mixed with fissioning gas or injected into the peripheral

buffer gas region such that there is no blockage of radiation due to the intrinsic absorption characteristics of transparent materials at short wavelengths.

Three fundamental areas of research are required to demonstrate the feasibility of the PCR concept: (1) nuclear criticality; (2) fluid mechanical confinement; and, (3) transfer of energy by radiation processes. Various aspects of these areas of technology are currently being investigated at UTRC. In addition, cavity reactor experiments which employ gaseous UF_6 are currently being performed at Los Alamos Scientific Laboratory (LASL) as part of the planned NASA program to determine PCR feasibility.

The present report summarizes recent results of emission and absorption measurements in hot UF_6 - argon mixtures at various wavelengths, temperatures and pressures. These data are required to provide basic absorption data for radiation transport calculations and relative emission data for subsequent comparison of theoretical calculations with experimental results.

Identification of commercial products in this report is to adequately describe the materials and does not constitute official endorsement, expressed of implied, of such products or manufacturers by the National Aeronautics and Space Administration.

LIST OF SYMBOLS

I_o	Source lamp or incident intensity, arbitrary units
I_t	Transmitted intensity, arbitrary units
I_{UF_6}	UF_6 intensity per unit pressure, arbitrary units
I_A	Argon intensity, arbitrary units
r	Relative position along optical path, dimensionless
$\dot{m}$	Mass flow rate, G/Sec
T	Absolute temperature, deg K
N	Mol fraction, dimensionless
P	Pressure, atm or mm Hg
M	Molecular or atomic weight, G/Mol
L	Path length in optical plenum = 10.2 cm
Ln	Natural logarithm
λ	Wavelength, nanometers
σ	Absorption cross section, cm^2
R	Detector spectral response, arbitrary units

TEST EQUIPMENT

The equipment used in the experimental evaluation of the spectral properties of UF_6 - argon mixtures consisted of three major components. These were the plasma torch-optical plenum assembly, the monochromator, and the UF_6 transfer system. A schematic of the overall system is shown in Fig. 2; details of these components as well as other auxiliary equipment are discussed in the following sections.

Plasma Torch-Plenum Assembly

The UTRC plasma torch facility was used to provide the UF_6 - argon mixtures for all experimental determinations. A schematic of the plasma torch illustrating the major torch components is depicted in Fig. 3. The principal components of the torch are a pin-type cathode, a hollow conical anode, an argon injector, a UF_6 injector, and two external magnets. The cathode is a hemispherical-tipped 2 percent thoriated tungsten rod 0.1 cm in diameter and was provided with means for independent water cooling. The cathode extended partially into the water-cooled copper anode as shown in Fig. 3. Argon was injected tangentially through four equally spaced 0.016-cm diameter holes located at the base of the cathode assembly. The aerodynamic swirl imparted by the argon injection system was augmented magnetically by means of two external magnets located concentrically about the anode as shown in Fig. 3.

A Rapid Electric, Model SRV/MAN, 200 kW dc power supply was used to provide power to the discharge. The external magnets were supplied by Hypertherm Model H-444 dc power supplies rated at 16.2 kW (90 v, 180 A) with a 60 percent duty cycle. The large magnet was connected to two parallel Model H-444 supplies to provide higher current (360 A) and, thus, a greater magnetic field. The smaller magnet was supplied by one Model H-444.

A UF_6 injector was located immediately adjacent to and downstream of the anode. Preheated UF_6 was injected into the argon stream heated by the torch via two 0.16-cm diameter holes. The aerodynamically mixed high temperature gas stream was then introduced into the cylindrical stainless-steel plenum (see Figs. 2 and 3). The plenum was equipped with six optical ports (three oppositely positioned pairs) which enable viewing the high temperature gas stream spectroscopically in absorption or emission. The optical path through the mixed gas stream was 10.2 cm long. The exhaust gases were subsequently neutralized in a sodium bicarbonate scrubber exterior to the laboratory. In addition, a scanning thermocouple system (tungsten - 5 percent rhenium; tungsten - 26 percent rhenium) was installed such that temperature scans could be made in the plasma parallel to the optical path. A typical temperature profile is illustrated in Fig. 4.

Monochromator

A Jarrell-Ash quarter meter monochromator was used for the spectral determinations. The low emission intensities from the UF_6 - argon mixtures necessitated use of the 100 μ entrance and exit slits for all measurements. A series of four diffraction (reflection) gratings were available for use and were blazed at nominal wavelengths of 600, 1000, 2000 and 4000 nanometers. A two speed motor (15 and 30 rpm) was installed to provide wavelength drive and was normally used at the lower speed. Spectral scans were limited to a duration of approximately 1.5 minutes due to torch operating constraints and to limit exposure of equipment to UF_6.

Two detectors were utilized with the previously described gratings. A photodiode detector (EG & G, model HUV-1000B) was used with the 600 nanometer grating while a liquid nitrogen cooled Indium Antimomide (InSb) infrared detector (Judson model J-10) was used in conjunction with the infrared gratings. The InSb detector has a nominal low wavelength cut-off at about 1000 nanometers. Typical spectral response curves for these detectors are illustrated in Fig. 5.

Sapphire windows were provided to confine the hot UF_6 - argon mixtures to the optical plenum. The window holders were equipped with a small argon purge flow to assist in keeping the windows free of UF_6 deposits (see Fig. 2). A separate argon flow system was used to purge the monochronomator housing of atmospheric gas which strongly absorbs in the infrared region (principally water vapor).

A tungsten-halogen lamp of known relative emission characteristics in the visible and infrared regions (maximum calibrated wavelength 2600 nanometers) was used to calibrate the optical system and to cross calibrate the 600 and 1000 nanometer gratings. The "standard" lamp was also used as a source for the absorption measurements.

UF_6 Transfer System

The transfer system consisted of a two liter Monel supply canister rated at 200 atm with appropriate shut-off and metering valves as indicated in Fig. 2. Two chromel-alumel thermocouples were installed in the canister to monitor the temperature of the UF_6 liquid and gas phases. A Matheson linear mass flow meter was used to determine UF_6 mass flow rates during various experiments. The mass flow meter output was continuously monitored on a Hewlett-Packard dual channel recorder during all tests involving UF_6 flow. The canister, valves, mass flow meter, and all lines in the UF_6 transfer system were electrically heated by means of Variac controlled heater tapes. Chromel-alumel thermocouples were placed in various components to monitor temperatures at strategic locations in the UF_6 flow loop. An argon-flow meter system was incorporated in the UF_6 transfer loop to permit on-line, periodic calibration of the Matheson flow meter.

EXPERIMENTAL PROCEDURES

Preliminary experiments were conducted to establish operating parameters in terms of argon flow rates, power supply settings and corresponding attainable gas temperatures in the optical plenum. These tests were conducted with argon only. As noted in a previous report (Ref. 3), gas temperature is determined primarily by the argon flow rate and to a lesser degree by the applied voltage or current.

Standard procedure involved initiation of the discharge on argon and adjustment of argon flows and arc current until the desired gas temperature was attained. A short wait was usually required prior to UF_6 injection to permit adequate warming of the UF_6 injector ring. Upon injection of preheated UF_6 and subsequent stabilization of the system, temperature scans were conducted to ascertain the temperature profile in the plenum, adjacent and parallel to the optical path. Subsequently, spectral emission scans were commenced. Termination of a run was accomplished by arresting UF_6 flow and initiating a purge of hot argon from an auxiliary system until the lines were clear of UF_6. The torch was usually operated during the purge cycle.

Spectral scans with UF_6 were usually followed by a scan of the tungsten-halogen lamp to determine the condition of the sapphire windows and to serve as a check on the calibration of the optical system. No evidence of window degradation due to UF_6 deposition was found as exemplified by this procedure.

Absorption determinations were performed at fixed wavelengths (no spectral scan) using the tungsten-halogen lamp as a source. After initiation of the discharge, the intensity of the transmitted (essentially I_o) beam was determined without UF_6 flow. (The argon intensity of emission was always negligible.) Subsequently, UF_6 flow was initiated and the combined intensity ($I_t + I_{UF_6}$) was ascertained. Finally, the lamp was extinguished and the emission from UF_6 (I_{UF_6}) was measured at the same conditions (fixed wavelength and temperatures). Thus, the transmitted intensity could be determined as follows:

$$(I_{UF_6} + I_t) - I_{UF_6} = I_t \tag{1}$$

The partial pressure UF_6 was calculated from a knowledge of the mass flow rates of argon and UF_6 and the total system pressure as follows:

$$P_{UF_6} = \bar{N}_{UF_6} P_t \tag{2}$$

$$\bar{N}_{UF_6} = \frac{(\dot{m}/M)_{UF_6}}{(\dot{m}/M)_{UF_6} + (\dot{m}/M)_{Argon}} \tag{3}$$

Previous experience with Matheson linear mass flow meters indicated deterioration of the system after extensive use with UF_6 (Ref. 3). Therefore, periodic calibration of the Matheson flow meter was effected by means of on-line argon flow calibration system.

RESULTS AND DISCUSSION

The following paragraphs summarize the spectral results obtained with respect to the high temperature emission and absorption characteristics of UF_6.

Spectral Emission

A series of twelve rapid emission scans were conducted at four temperatures from approximately 1000 to 1800 K using to 600, 1000, 2000 and 4000 nanometer gratings. The purpose of these survey runs was to ascertain regions of significant UF_6 emission in the wavelength region between 600 and 5000 nanometers over the temperature range of interest for subsequent more detailed spectral examination. Therefore, no data reduction was effected for this group of experimental runs. A summary of these spectral runs is compiled in Table I in terms of the grating used, temperature and UF_6 partial pressure. Total pressure for all runs was 1.0 atmospheres. The scanning thermocouple was not used during these scans but remained fixed at the center of the optical plenum.

Examination of the experimental traces of emission as a function of wavelength showed significant emission from UF_6 only in the wavelength region between 600 and about 1800 nanometers. (Previous studies of emission from UF_6 at wavelengths less than 600 nanometers were reported in Ref. 3.) At wavelengths greater than 1800 nanometers the emission from UF_6 was negligible and barely discernable. Thus, subsequent detailed spectral runs were conducted in the wavelength region between 600 and 1800 nanometers.

Twenty-four detailed spectral emission scans were made in the wavelength region between 600 and approximately 1800 nanometers after completion of the survey runs. The 600 and 1000 nanometer gratings were used in these studies. Experimental parameters included temperature, UF_6 and argon mass flow rates, UF_6 partial pressure and total pressure (1.0 atmospheres for all cases). A summary of these parameters is presented in Table II.

Standard procedure involved periodic calibration of the spectral response of the optical system using the tungsten-halogen lamp as a relative calibration source for each of the two grating-detector combinations utilized in the emission measurements. Thus, direct comparison of measured emission from UF_6 could be made for each spectral region investigated. Similarly, spectral scans were taken at each temperature with argon flow only to assure that the contribution to emission from argon was negligible compared to UF_6 emission. Emission from argon was always found to be negligible compared to emission from UF_6.

A typical experimental trace in the wavelength region between 600 and 900 nanometers is reproduced as a function of wavelength in Fig. 6. The results depicted in Fig. 6 also show the relative emission from argon (dotted curve

and the relative emission from an UF_6-argon mixture at the same temperature. Except for the strong contribution from argon lines, the continuum due to argon is negligible with respect to the UF_6 emission. The argon line structure precluded determination of UF_6 emission throughout much of the region between 700 and 900 nanometers except for regions between the strong lines at wavelengths of 725, 788, 875 and 900 nanometers. Similar spectral results were obtained for other temperatures and at longer wavelengths. The emission from UF_6 at longer wavelengths was considerably reduced in intensity however.

A summary of the emission data for UF_6 in the wavelength range between 600 and 900 nanometers is illustrated in Fig. 7 for five temperatures between about 1100 and 1900 deg K. These data have been corrected for instrument response and have been normalized with respect to the partial pressure of UF_6 in the system. Generally, the profiles for UF_6 emission are essentially structureless and decrease rapidly with increasing wavelength at any given temperature. The emission increases by about a factor of 300 in going from a temperature of 1100 to 1900 deg K.

Similar experimental spectral emission results for UF_6 are shown as a function of wavelength in the region between 900 and 1800 nanometers in Fig. 8. These data have been corrected for instrument response and normalized with respect to the UF_6 partial pressure as in the previous figure. The emission data are illustrated for a temperature of 1270 and 1460 deg K. The data for the two wavelength regions of Figs. 7 and 8 are compared in Fig. 9 at approximately equivalent temperatures. Exact temperature setting from run to run was not possible due to minor variations in the argon flow and UF_6 flow into the torch and to minor variations in the arc current or voltage.

The results in Fig. 9 show reasonable agreement in the two wavelength ranges examined for approximately the same temperatures. The apparent discrepancy at wavelengths around 900 nanometers for the two wavelength ranges is due to decrease in instrument response at the extremes of the respective grating-detector combinations which results in low recorded signal levels. The experimental data in Figs. 8 and 9 show the continued decrease in emission from UF_6 at longer wavelengths. Essentially no emission was observed at wavelengths greater than about 1800 nanometers at any temperature investigated. Similarly, negligible or no emission was observed at any wavelength for temperatures below 1100 deg K.

Qualitatively these results are in agreement with the spectral emission data reported in Ref. 4 at shorter wavelengths and at similar temperatures.

Two emission scans were made at different flow rates of UF_6 to ascertain the effect of UF_6 partial pressure on spectral emission intensity. These runs were conducted at a temperature of 1420 deg K and for UF_6 partial pressures of 7.3 and 12.7 mm of Hg, respectively. A wavelength range from 600 to 900 nanometers was scanned. Results of these determinations are shown as a function of wavelength in Fig. 10. The data exhibit no effect of UF_6 on the relative emission per unit UF_6 pressure. Therefore, an optically thin plasma is implied for the conditions of these experiments.

Absorption Experiments

A series of four runs were conducted to attempt determination of the absorption cross section of UF_6 at selected wavelengths. All absorption runs were made at a temperature of 1000 deg K and at four fixed wavelengths of 600, 625, 650 and 675 nanometers. The tungsten-halogen lamp was used as a source for these measurements as described in a previous section of this report. The partial pressure of UF_6 was increased to a nominal value of 25 mm of Hg in order to increase the number density of absorbers in the optical path since the path length in the apparatus is fixed at 10.2 cm.

No apparent absorption was indicated for these operating parameters. Since the capabilities of the equipment allows measurement of cross section of the order of about 5×10^{-21} cm^2, it is concluded that the absorption cross section at the indicated wavelengths was at least 10^{-22} cm^2. Additional measurements were not attempted since the maximum flow rate had been attained in the system and the path length could not be increased.

An absorption cross section of less than 10^{-22} cm^2 in these wavelengths is in quantitative agreement with the data reported in Ref. 4 in the visible region of the spectrum.

CONCLUSIONS

In summary, the emission and absorption characteristics of hot UF_6 - argon mixtures have been experimentally examined over a range of wavelengths and temperatures. The results indicate only significant or measurable emission from UF_6 in the approximate wavelength range from 600 to 1800 nanometers and at temperatures in the range from 1100 to about 1900 deg K. Absorption measurements were attempted at wavelengths between 600 and 765 nanometers.

The following general conclusions are inferred from these experimental spectroscopic investigations:

1. Significant emission from hot UF_6 - argon mixtures occurs only in the wavelength range between 600 and 1800 nanometers and in the temperature range between 1100 and 1900 deg K.

2. Negligible emission is observed at wavelengths greater than approximately 1800 nanometers and for temperatures below about 1100 deg K.

3. Emission from argon is negligible at all temperatures and wavelengths examined except for the argon line region between 700 and 900 nanometers.

4. The emission spectrum for UF_6 exhibits effectively no spectral structure between 600 and 1800 nanometers.

5. No absorption was measured at wavelengths between 600 and 675 nanometers at a temperature of 1000 deg K indicating a cross section of less than about 10^{-22} cm^2 for UF_6.

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TABLE I

SURVEY SCANS

Run Number	Grating (NM)	Temp. °K	P_{UF_6} mm Hg
1	600	1075	7.5
2	1000	1080	7.5
3	4000	1060	7.5
4	600	1310	6.4
5	1000	1330	6.9
6	4000	1320	6.7
7	600	1700	5.9
8	1000	1710	5.8
9	4000	1720	5.7
10	600	1820	3.8
11	1000	1820	3.6
12	4000	1830	3.8

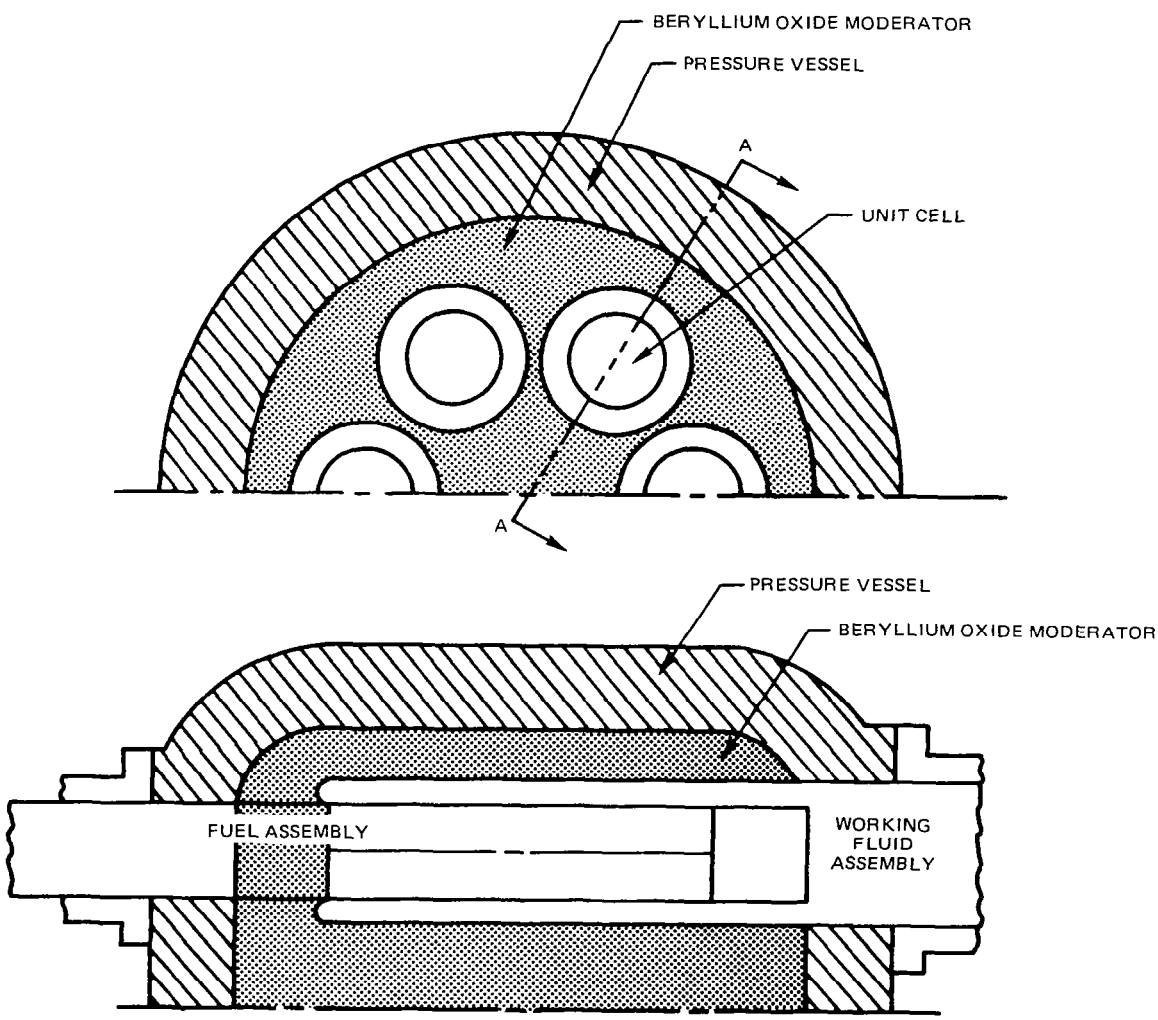
TABLE II

SUMMARY OF EXPERIMENTAL RUNS

Run Number	Grating (NM)	Temp. °K	Press - UF_6 mm Hg	Comments
1	600	660	7.8	Emission - UF_6
2	600	680	7.8	Emission - UF_6
3	600	1075	7.5	Emission - UF_6
4	600	1100	6.9	Emission - UF_6
5	600	1320	6.4	Emission - UF_6
6	600	1340	6.9	Emission - UF_6
7	600	1480	6.9	Emission - UF_6
8	600	1490	12.7	Emission - UF_6
9	600	1490	6.9	Emission - UF_6
10	600	1700	5.9	Emission - UF_6
11	600	1720	5.9	Emission - UF_6
12	600	1875	3.8	Emission - UF_6
13	600	1880	3.5	Emission - UF_6
14	600	1890	3.8	Emission - UF_6
15	600	1080	-	Emission - argon
16	600	1450	-	Emission - argon
17	600	1710	-	Emission - argon
18	1000	690	7.8	Emission - UF_6
19	1000	1470	6.8	Emission - UF_6
20	1000	1450	7.3	Emission - UF_6
21	1000	1280	7.2	Emission - UF_6
22	1000	1260	6.9	Emission - UF_6
23	1000	1390	-	Emission - argon
24	1000	1060	-	Emission - argon
25	600	1000	25.1	Abs - UF_6 at 600 mm
26	600	1000	25.1	Abs - UF_6 at 625 mm
27	600	1000	25.1	Abs - UF_6 at 650 mm
28	600	1000	25.1	Abs - UF_6 at 675 mm

FIG. 1

SCHEMATIC OF A PLASMA CORE REACTOR



SCHEMATIC OF UF_6 PLASMA TORCH SYSTEM

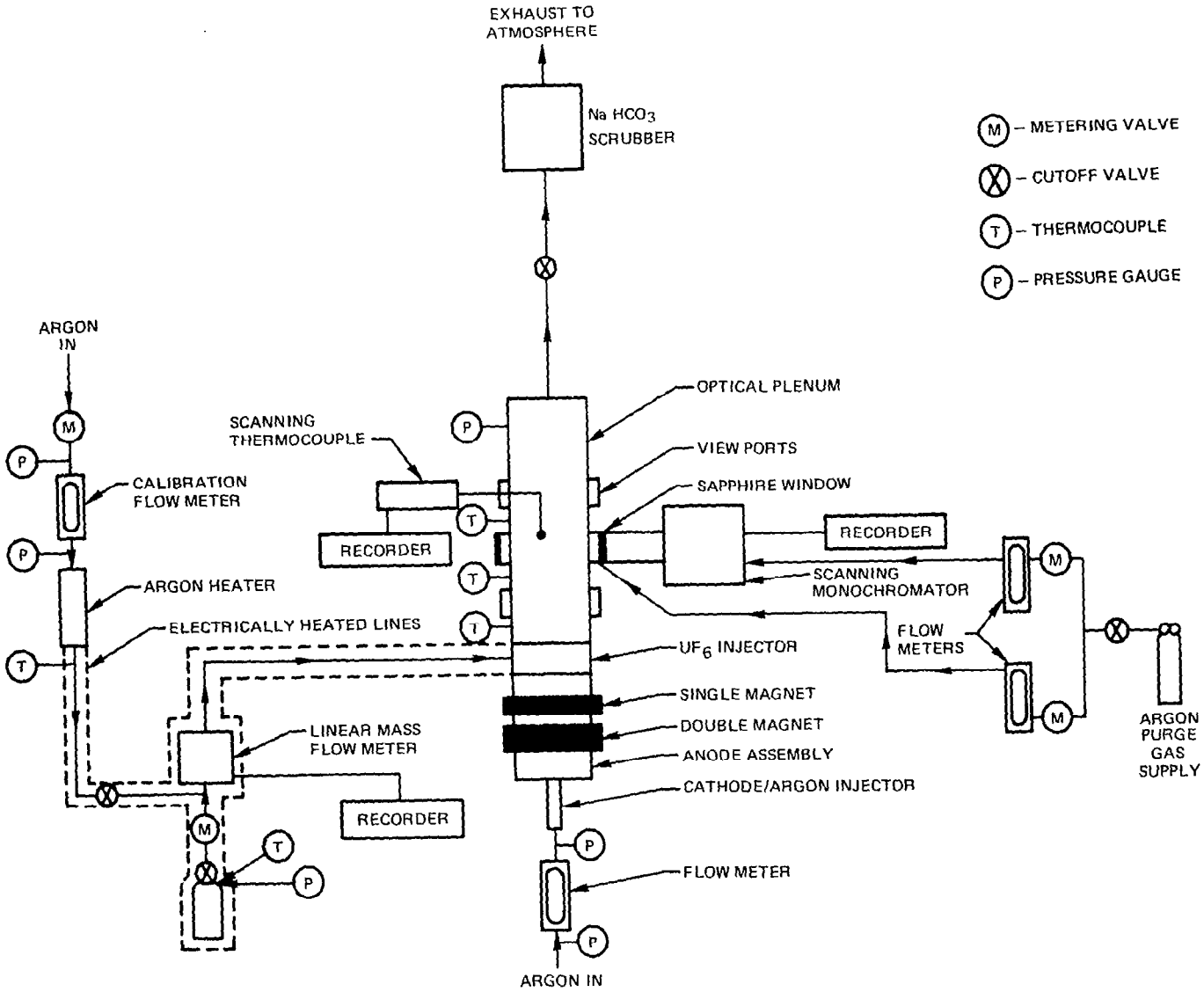


FIG. 2

SCHEMATIC OF PLASMA TORCH ASSEMBLY

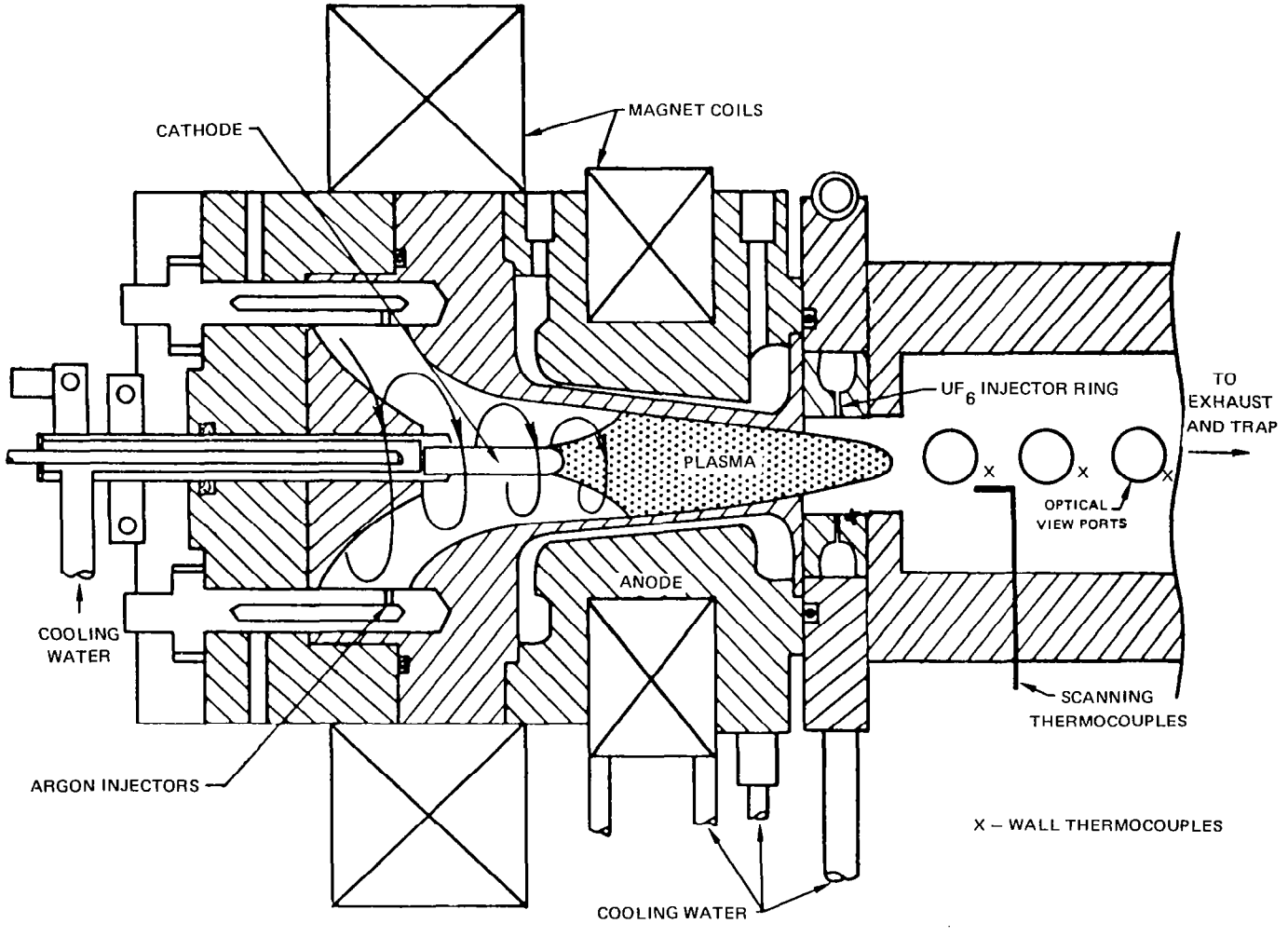
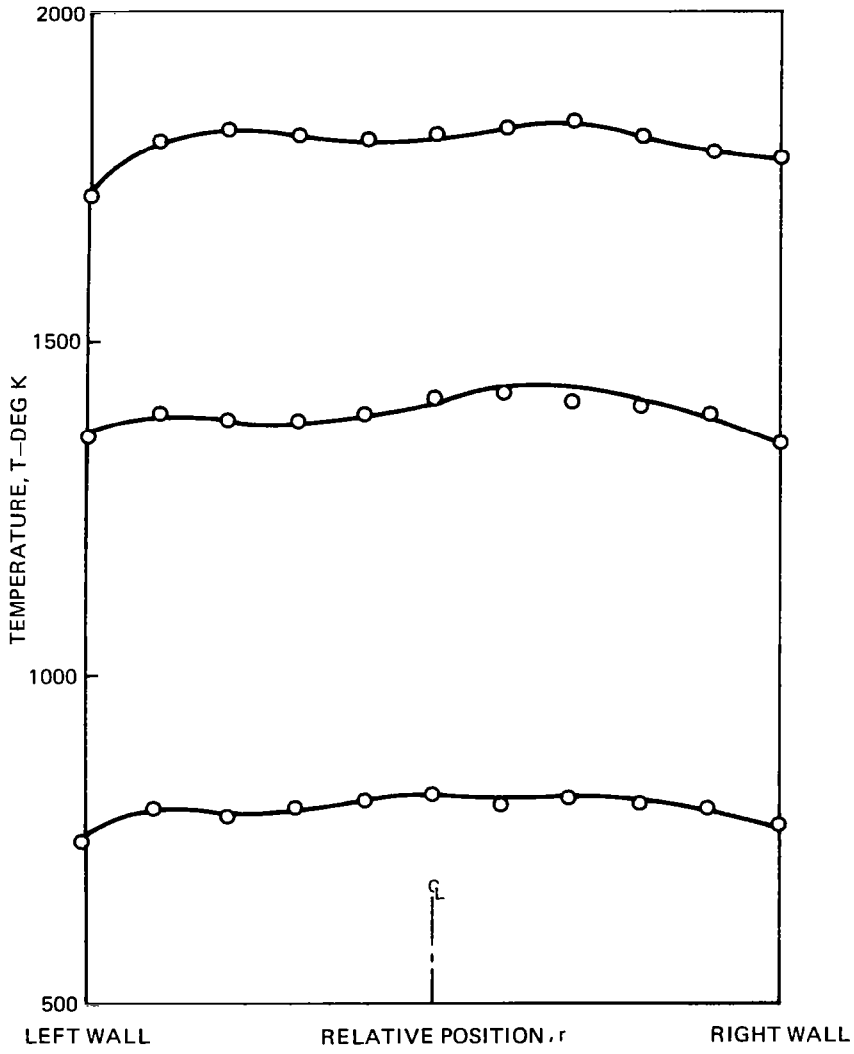


FIG. 3

FIG. 4

TYPICAL TEMPERATURE PROFILES ALONG THE OPTICAL PATH IN THE PLENUM

(DATA TAKEN AT OPTICAL PORT ADJACENT TO UF_6 INJECTOR)



DETECTOR RESPONSE AS A FUNCTION OF WAVELENGTH

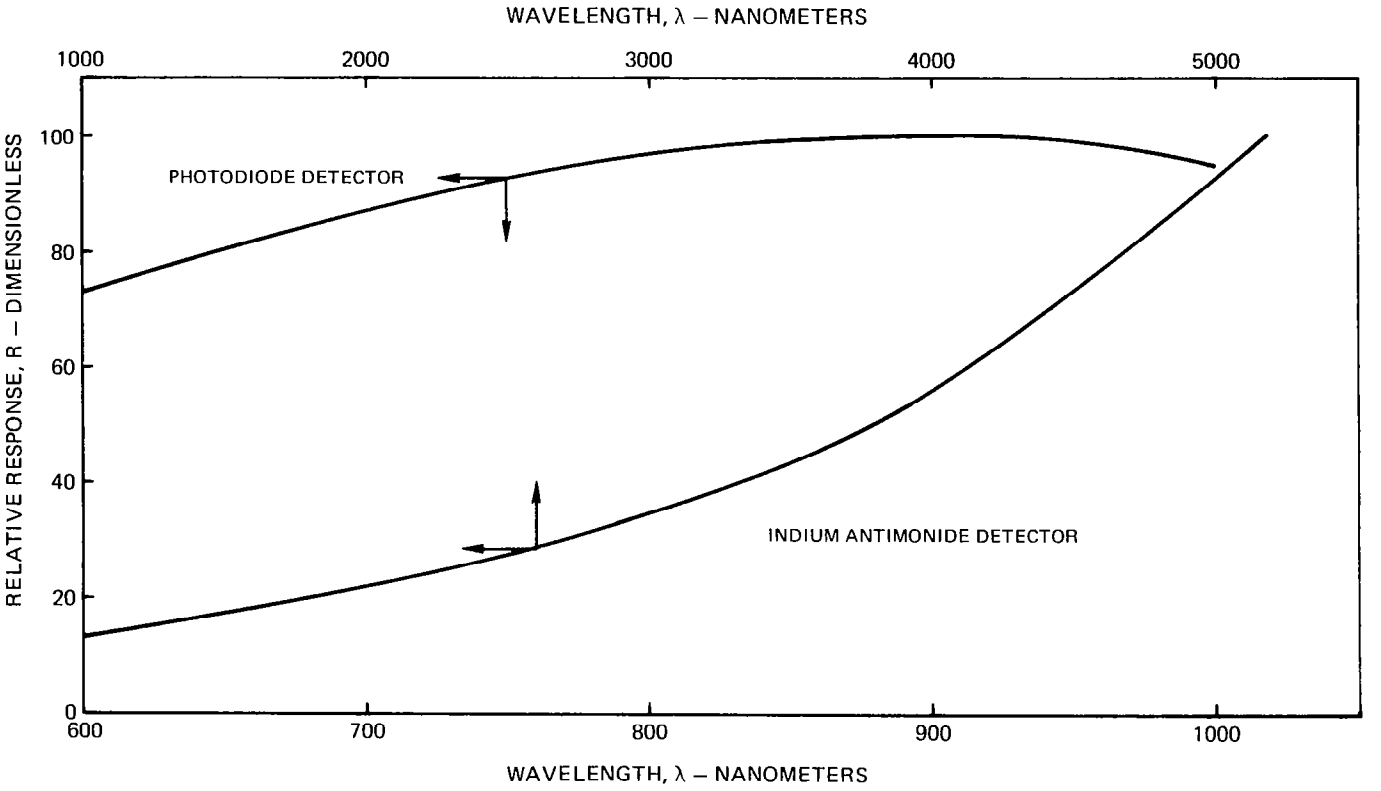


FIG. 5

TYPICAL EXPERIMENTAL SPECTRAL EMISSION RESULTS

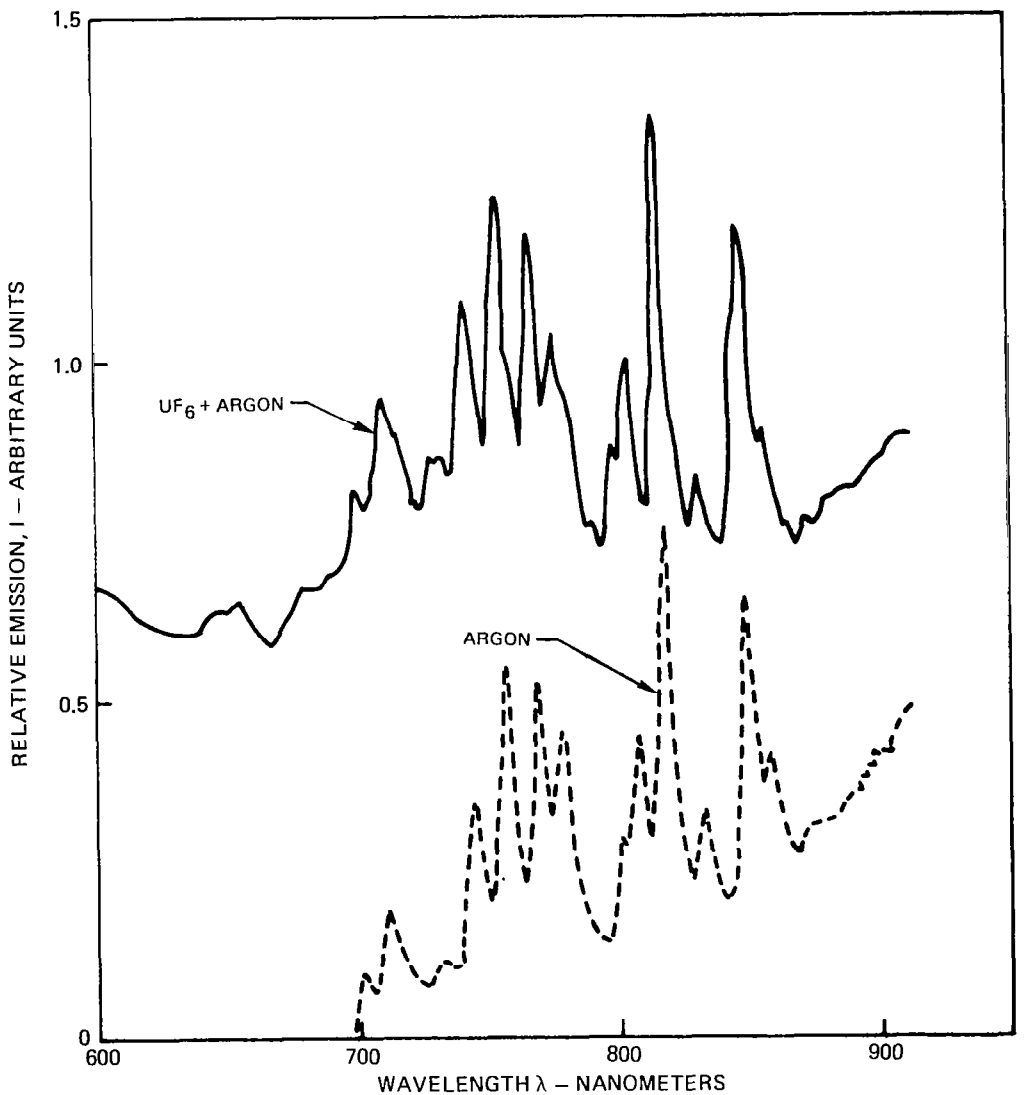


FIG. 6

FIG. 7

RELATIVE EMISSION PER UNIT UF_6 PRESSURE BETWEEN 600 AND 900 NANOMETERS

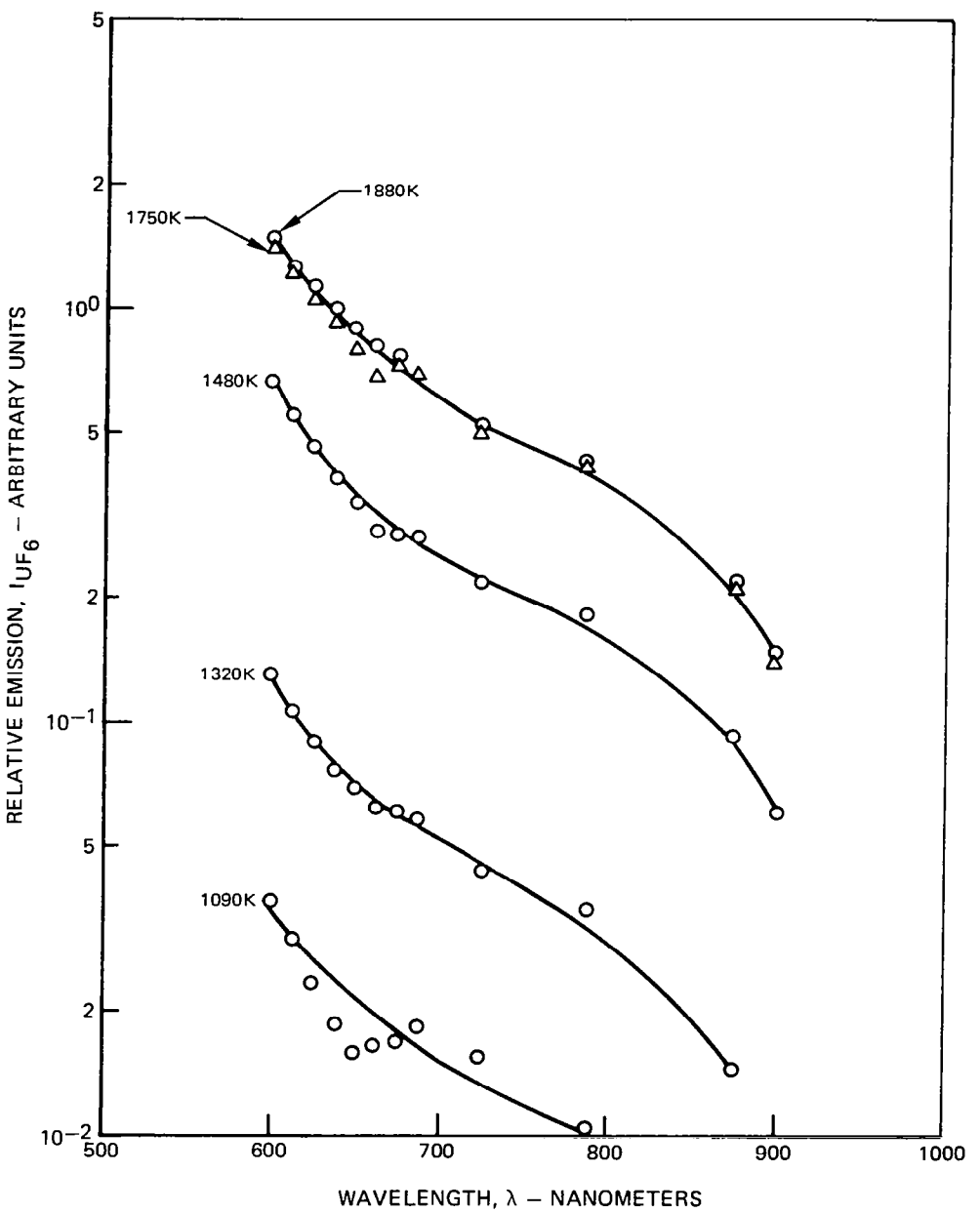


FIG. 8

RELATIVE EMISSION PER UNIT UF_6 PRESSURE BETWEEN 900 AND 1800 NANOMETERS

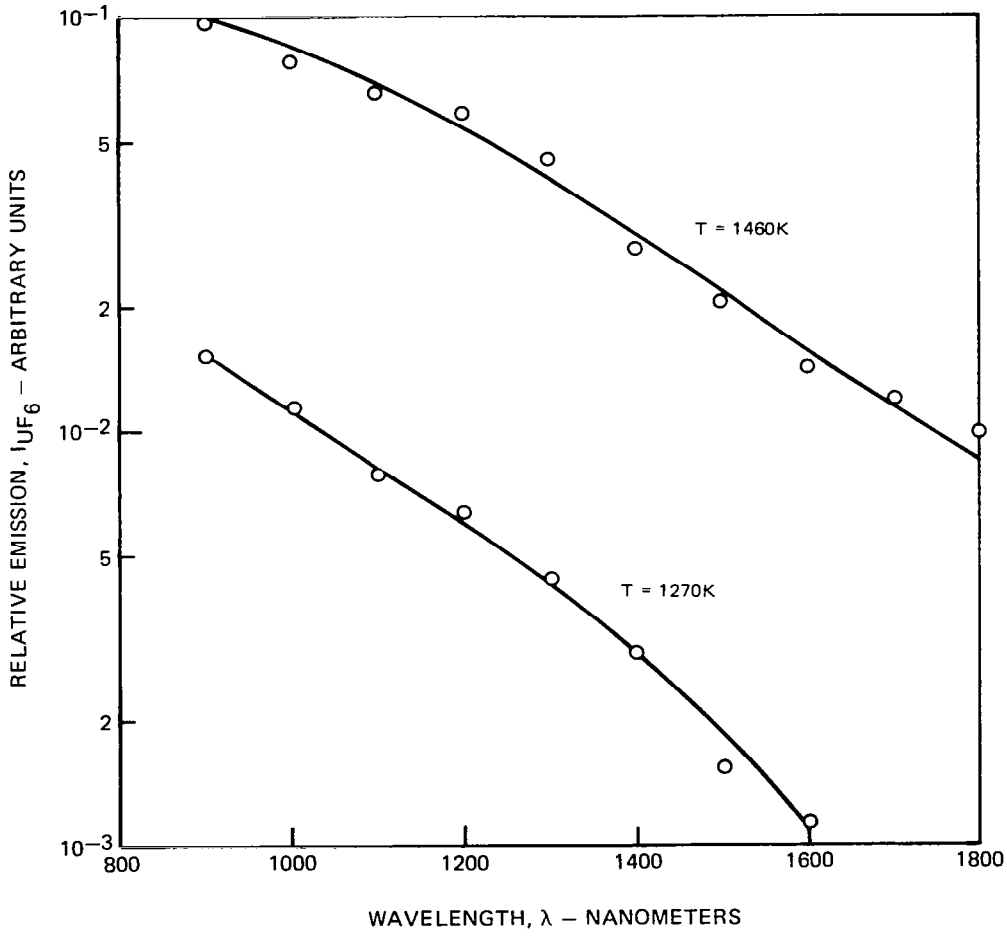
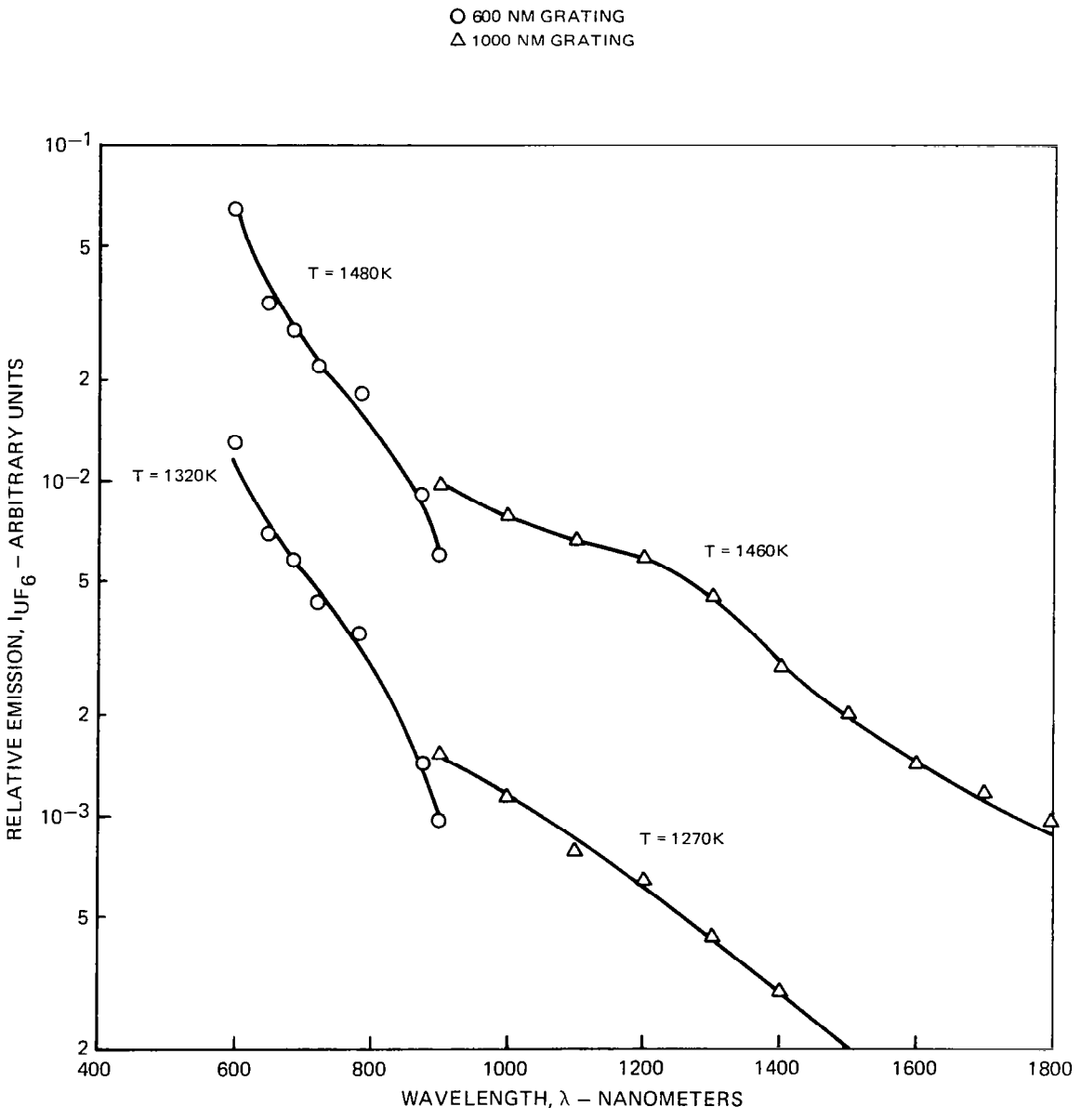
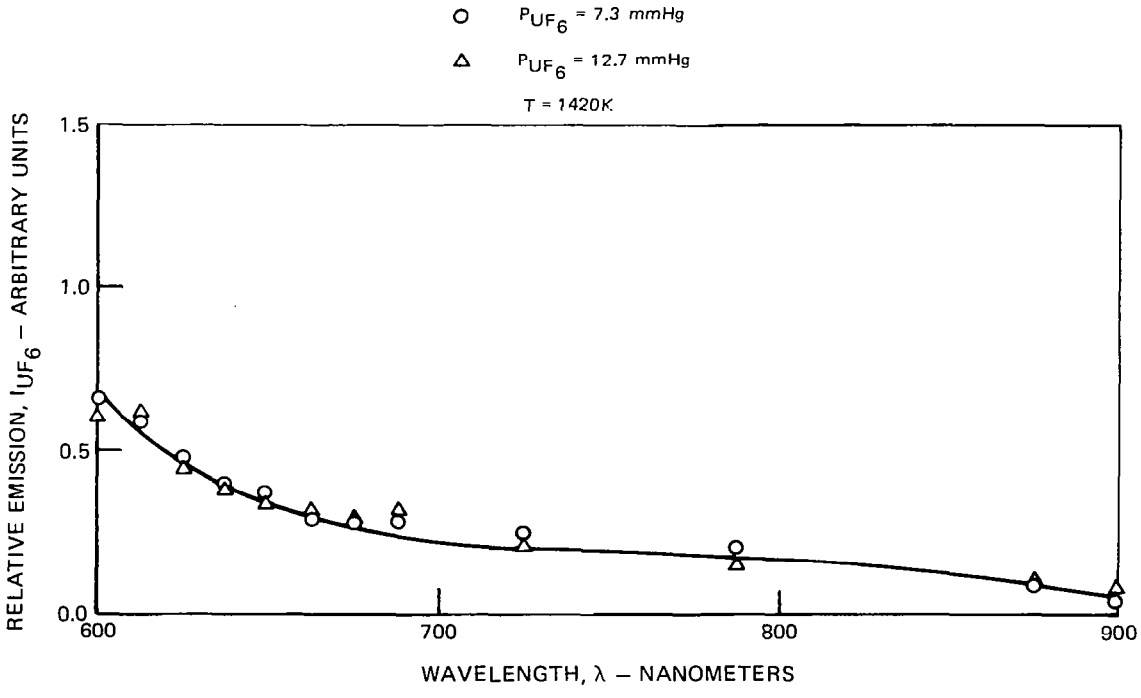


FIG. 9

COMPARISON OF EMISSION PER UNIT UF_6 PRESSURE FOR DIFFERENT GRATINGS IN THE WAVELENGTH REGION BETWEEN 600 AND 1800 NANOMETERS



COMPARISON OF RELATIVE EMISSION PER UNIT UF_6 PRESSURE AT DIFFERENT UF_6 PARTIAL PRESSURES IN THE WAVELENGTH RANGE BETWEEN 600 AND 900 NANOMETERS



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15. Supplementary Notes

Langley Technical Monitor: Frank Hohl
UTRC Technical Program Manager: J. S. Kendall
Topical Report

16. Abstract

An experimental study was conducted to determine relative spectral emission and spectral absorption data for UF_6-argon mixtures at elevated temperatures. These spectral data are required to assist in the theoretical analysis of radiation transport in the nuclear fuel-buffer gas region of a plasma core reactor (PCR). Current spectral data for UF_6 do not cover the wavelength range between approximately 600 to 5000 nanometers. In addition, no data exist at elevated temperatures of interest to the PCR concept.

Relative emission measurements were made for UF_6-argon mixtures over a range of temperatures from about 650 to 1900 K and in the wavelength range from about 600 to 5000 nanometers. All emission results were determined for a total pressure of 1.0 atm. Uranium hexafluoride partial pressures varied from about 3.5 to 12.7 mm Hg. Absorption measurements were attempted at 600, 625, 650 and 675 nanometers for a temperature of 1000 K. The uranium partial pressure for these determinations was 25 mm Hg. Generally, the results exhibit appreciable emission for hot UF_6-argon mixtures at wavelengths between 600 and 1800 nanometers. The relative emission increases strongly with temperatures. At wavelengths greater than about 1800 nanometers, no significant emission was observed at all temperatures. No measurable absorption was observed at wavelengths between 600 and 675 nanometers for UF_6 at a temperature of 1000 K. The absorption determinations indicate cross sections of less than 10^{-22} cm^2.

17. Key Words (Suggested by Author(s))	18. Distribution Statement
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