UF_6 THERMAL PLASMA AND ASSOCIATED DIAGNOSTIC RESEARCH

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ABSTRACT

Research is being conducted in developing some of the technology applicable to uranium plasma core reactors. Included is the transfer, flow control, and exhaust gas systems required to permit injection of pure UF_6 into an rf plasma test chamber and subsequent recovery and reconstitution of the uranium compound exhaust products. Results to date indicate techniques have been developed for fluid mechanical confinement of high-temperature, UF_6 plasmas for long test times while simultaneously minimizing uranium compound deposition on the walls. Associated diagnostic techniques for characterizing the plasma, exhaust gases, and residue have been successfully developed and applied.

INTRODUCTION

Fissioning uranium plasma core reactors based on the utilization of fissile nuclear fuel in the gaseous state could be used as a prime energy source for many space and terrestrial applications. Refs. 1, 2 summarize various reactor concepts, designs, principles of operation, applications, and associated performance estimates. Figure 1 is a sketch of one unit cell. The reactor would consist of one or more such cells imbedded in beryllium oxide and/or heavy-water moderator and surrounded by a pressure vessel. In the central plasma fuel zone, gaseous uranium injected in the form of UF_6 is confined by argon buffer gas injected tangentially at the periphery of the cell. The thermal radiation from the plasma is coupled to a separate working fluid channel. A long-range plan for establishing the feasibility of fissioning gaseous UF_6 and uranium plasma reactors has been formulated by NASA[3]. The plan includes demonstration of an argon/UF_6 injection, separation, and recirculation system to efficiently separate UF_6 from argon in a form adaptable to subsequent recycling in the uranium plasma experiment. Past fluid mechanics nonplasma experiments[4] have indicated that a radial inflow vortex is applicable for preferential containment of a heavy gas within a light gas. Other experiments[5] have been conducted on the confinement of argon rf plasmas. These tests resulted in development of a high-intensity plasma energy source (equiv. $T_{BB} > 6000$ K).

CONFIGURATION

Different test chamber configurations were tested before selecting the configuration and flow scheme which demonstrated the best confinement characteristics[6]. Figure 2 is a sketch showing a cross section of the test chamber configuration selected for use in the UTRC 1.2 MW rf (5.4 MHz) induction heater facility. This chamber was designed for power levels up to 100 kW and pressure levels up to 20 atm. A single endwall driven vortex injection scheme and a combination of on-axis exhaust thru-flow and axial bypass exhaust on the opposing endwall are used. A feeder system to provide a controlled and steady flow of UF_6 at temperatures up to 500 K was developed. The system was designed to provide UF_6 mass flow rates up to 5 g/s and pressure levels up to 20 atm. Monel was used because of its resistance to chemical attack by hot, pressurized UF_6.

DIAGNOSTICS AND SUPPORTING ANALYSES

An optical diagnostic system was developed for performing simultaneous plasma emission and absorption chordal scan measurements. Species or uranium concentration was measured by absorption of the strong transition at $\lambda = 591.54$ nm using a tunable cw single-frequency dye laser system. Previously developed analytical techniques for absorption/emission coefficient determination for optically-thick plasmas were adapted to the UTRC data acquisition system[7,8]. The absorption coefficient was determined from the attenuation of the intensity of the probing dye laser beam traversing the plasma source. Ref. 9 describes a method utilizing an iterative procedure to find the radial distribution of the emission coefficient. The radial temperature profile was determined via Kirchhoff's law without knowledge of particle densities or transition probabilities. This is particularly important for uranium where much data are still unknown. Typical centerline temperatures were 9700 K. To aid in the determination of the total uranium atom number density distribution within the rf plasma, UF_6/Ar equilibrium composition data were calculated[10] for the current range of test parameters (partial pressure from 10^{-4} to 1 atm and $300 \leq T \leq 10^4$ K - see Fig. 3). Calculations have been completed on the variation of the total number density of uranium atoms and ions (i.e., UI+UII+UIII) with UF_6 injection partial pressures and equilibrium temperature. The partition functions for Uranium I and II as a function of temperature[11], the radial distributions of temperature and transmission, and the variation of uranium neutral atom number density with UF_6 partial pressure, were used to calculate the total uranium atom number density (UI+UII+UIII) as a function of radius. The total uranium atom number density reached a maximum of approximately 10^{16} atoms/cm^3 at the centerline of the plasma. X-ray absorption equipment and methods were also developed[6] for measuring the amount of uranium confined in the uranium rf plasma. Estimates indicated that absorption of 8 KeV x-rays from a copper target can be used to determine amounts of uranium within the density range of interest in these experiments (3×10^{16} to 3×10^{18} atoms/cm^3). The basic system consisted of an x-ray generator, rf uranium plasma test chamber, and the x-ray detection components. The emitted x-rays from the

target passed through a collimator and slit assembly and were reflected at the Bragg angle for 8 KeV ($\lambda = 0.154$ nm) x-rays from a magnesium oxide (MgO) crystal monochromator before passing through the test chamber. The test chamber contained x-ray viewing windows (0.125-mm-thick Mylar) which were essentially transparent to the 8 KeV x-rays. In these tests, the fraction of x-rays transmitted were compared with the transmission calculated for UF_6 using Beers' law. At comparable test conditions to those used in the optical chordal scan measurements ($\dot{m}_{UF_6}=3.2\times10^{-2}$ g/s) and assuming a 23-cm-diagonal path length, the corresponding uranium total number density was 2.9×10^{16} atoms/cm^3. The agreement is reasonable considering that the x-ray measurements provide an upper limit and include contributions due to all species (including cold UF_6, etc. in the boundary layer). Other x-ray measurements at higher UF_6 injection mass flow rates resulted in uranium densities up to approximately 4×10^{17} atoms/cm^3 (see Fig. 4). Experiments using a dc plasma torch system were also conducted to investigate the spectral properties of UF_6 and its thermal decomposition products in the VUV wavelength range. Also included were measurements of the spectral emission and absorption for UF_6 and possible thermal decomposition products of UF_6 at elevated temperatures ($800 \leq T \leq 1800$ K)[12]. In general, the results show that relatively little emission occurs at wavelengths $\lesssim 300$ nm over the temperature range investigated. The absorption cross sections determined from transmission measurements ($420 \leq \lambda \lesssim 580$ nm, $1000 \leq T \leq 1800$ K) ranged in value from about 8×10^{-19} to 9×10^{-20} cm per molecule of UF_6. The degree of wall coating incurred on the fused-silica peripheral walls after long run time tests with pure UF_6 injection was also investigated. The maximum operating test time in the experiments was 41.5 min. at a UF_6 flow rate of 2.2×10^{-2} g/s; in this case 30.4 mg of residue was deposited on the fused-silica tube. This included approximately 10 mg of silicone grease initially deposited on the ends of the tubes for sealing the O-rings. In other tests, greater than an order of magnitude increase in flow rates were achieved (2.1×10^{-1} g/s) while still maintaining the rf plasma in a confined steady-state mode. To permit a detailed analysis of the samples of residue collected from the various components of the test chamber after rf plasma tests with pure UF_6 injection, the following UTRC instrumentation was employed: (1) Profilometer, (2) IR Spectrophotometer (2.5-40μm), (3) Scanning Electron Microprobe, (4) X-Ray Diffractometer, (5) Electron Microprobe using selected area diffraction, (6) Ion Scattering Spectrometer (ISS)/Secondary Ion Mass Spectrometer (SIMS). The results from the x-ray and electron diffraction analyses indicated traces of several oxides of uranium. This was verified by the IR spectrophotometry absorption measurements. In addition to traces of water vapor, silicon dioxide, uranium dioxide, and uranyl fluoride were also present. Results of measurements using the profilometer indicated the peripheral wall coating to be approximately 1-μm-thick. TABLE I summarizes some results[16] of the research directed toward minimizing the deposition of uranium compounds on the test chamber peripheral wall, endwall surfaces, and exhaust ducts. Traps and getters were required at the inlet gas locations to aid in reducing the impurities.

The use of a specially developed porous (5 μm pore size) sintered Monel duct with transpiring argon flow resulted in an approximate 2 orders of magnitude reduction in residue wall coating.

IN-PROGRESS EXHAUST GAS COMPOSITION AND REPROCESSING STUDIES USING PURE UF_6

Other complementary current research included investigations using both off- and on-line exhaust gas composition diagnostic techniques[13,14]. Both IR and mass spectrometric T.O.F. techniques have been employed to quantitatively determine the concentration of UF_6 and other gaseous constituents. Tests in progress indicate the feasibility of applying a specially developed ruggedized T.O.F. mass spectrometer, data acquisition and sampling probe system for on-line quantitative measurement of the UF_6 concentration at various sections of the exhaust system. Current emphasis is being placed on development of test equipment, operating techniques, and associated diagnostic equipment directed toward demonstration of a UF_6/Ar injection, separation, and recirculation system for use with the flowing rf-heated uranium plasma confinement tests. Included is the use of a static F_2 batch-type regeneration test apparatus for converting the uranium compound post-test residue back to UF_6. Test results to date indicate 100% conversion efficiencies are achievable; this result is considered significant because it demonstrates the feasibility of chemically converting on a batch basis all the nonvolatile exhaust products to UF_6 using a single reactant. Research is also in progress to develop a flowing preheated fluorine/UF_6 regeneration system. This includes providing additional information relative to the corrosion resistant aspects of various materials under extended exposure to hot, pressurized fluorine and UF_6.

ACKNOWLEDGMENT

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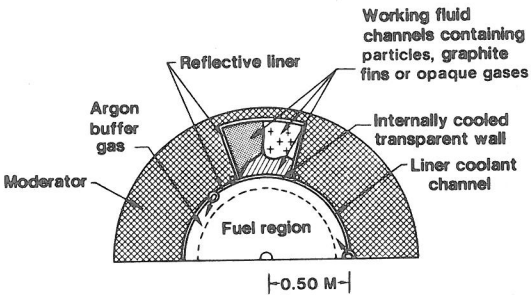

Fig. 1 - Plasma Core Reactor Unit Cell

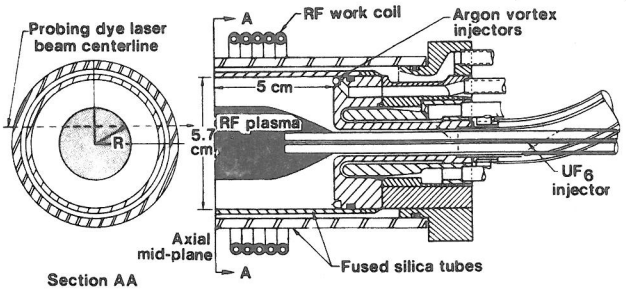

Fig. 2 - RF Plasma Test Chamber

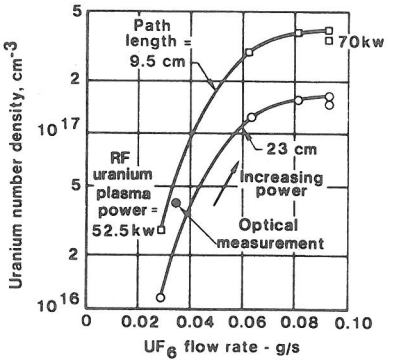

Fig. 4 - X-ray and Optical Absorption Results

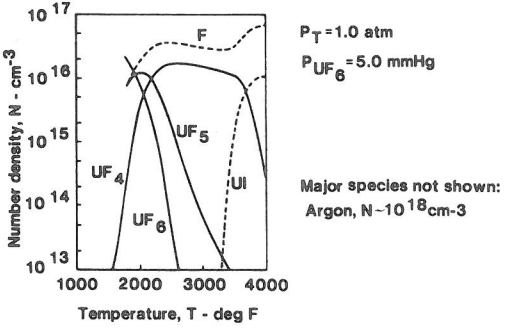

Fig. 3 - Equilibrium composition of UF_6/Ar

COMPONENT	EARLY TESTS		TESTS TO DATE	
	RESIDUE (mg)	MAJOR CONSTITUENT	RESIDUE (mg)	MAJOR CONSTITUENT
PERIPHERAL WALL	200 - 600	UO_2F_2, UF_4 UO_2, γUO_3 O-RING DECOMPOSITION	4 - 50	UO_2F_2
INJECTOR END WALL	90 - 200	UO_2F_2, UF_4 UO_2	4 - 20	UO_2F_2, UF_4
EXHAUST END WALL	25 - 100	UO_2F_2, UF_4, UO_2	0.1 - 3	UO_2F_2, UF_4
UF_6 INJECTOR	200 - 400	UO_2F_2, UF_4 UO_2, αUO_3,	10 - 50	UO_2F_2, UF_4, UO_2
INTERSTAGE EXHAUST DUCT	100 - 800	UO_2F_2, UF_4 U_3O_8	25 - 275	UO_2F_2, UF_4
INTERSTAGE EXHAUST DUCT WITH FLOW INJECTION AUGMENTATION	--	--	0.15 - 2.5	UO_2F_2, UF_4
POROUS MONEL EXHAUST DUCT WITH FLOW AUGMENTATION	--	--	0.05 - 0.3	UO_2F_2, UF_4

Table I: Summary of Residue Analysis