Ultraviolet discharges from a radio-frequency system for potential biological/chemical applications

Joseph Ametepe[1,†], Sheng Peng[2], and Dennis Manos[3]

[1] *Georgia Gwinnett College, School of Science and Technology, Lawrenceville-GA 30078, USA*
[2] *EXFO, Mississauga, on Canada L5N 6H7*
[3] *College of William and Mary, Department of Physics, Williamsburg, VA 2385, USA*

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In this work, we describe a new electrode-less radio-frequency (RF) excitation technique for generating excimers in the vacuum ultraviolet (VUV) and ultraviolet (UV) spectral regions for potential biological/chemical applications. Spectra data of Xe_2^*, XeI^*, and KrI^* generated by this new technique are presented. Optical efficiency of the lamp system ranges from 3% to 6% for KrI^*, 7% to 13% for XeI^*, and 15% to 20% for Xe_2^*. Also, results of irradiating E-coli with XeI^* discharge from this lamp system is presented to show one of the promising applications of such electrode-less apparatus. This new RF lamp system offers an interesting addition to the already existing technologies for generating VUV and UV light for various biological, physical, and chemical processes especially those requiring large area for high productivity.

Keywords: radio-frequency, ultraviolet light, Xe_2^*, XeI^*, KrI^* excimers, escherichia coli

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1. Introduction

Excimer lamps[1–5] are new ultraviolet (UV) light sources capable of delivering high power, high efficiency, and narrow-band radiation from 354 nm to 126 nm. These lamps allow for selective, intense UV radiation at a specific wavelength useful for many industrial biological, chemical, and photo-physical processing applications.[6–15]

In the last decade, many research groups have studied different excitation techniques[16] for generating excimers. These include dielectric discharge (DBD), UV pre-ionization, microwave discharges, pulsed/continuous longitudinal discharges, nuclear excitation, and hollow cathode discharges. Not many of these excitation techniques used a 13.56-MHz radio-frequency (RF) for the purpose of efficiently generating excimers, although Alexandrovich *et al.*[17] described a capacitively coupled RF discharge for electrode-less subminiature fluorescent lamp with Hg–Ar gas mixtures. Furthermore, not many of the previous studies focused on using the excimer sources for destroying bacteria and viruses.

This paper is an experimental work describing a 13.56-MHz RF excimer source that is electrode-less and uses Hg-free gas mixtures for generating intense UV light for potential biological and chemical applications. This excitation technique takes a very different coupling approach from any of the previous excimer systems and is efficient enough to not only offer an interesting addition to the already existing technologies of generating vacuum ultraviolet (VUV) and UV light but also has potential for various practical research and industrial applications. The paper is organized in the following format. In Section 2, we give a description of the lamp system and the RF matching network. The spectra results are given in Section 3. In Section 4, we present some of biological applications, and the conclusion is given in the last section.

2. System description

2.1. Lamp system

Figures 1(a) and 1(b) show the electrode-less 13.56-MHz lamp schematics and discharge respectively. The lamp consists of an RF generator, impedance matching network, tuner (coupling means), and lamp bulb in a grounded cylindrical cavity. The RF generator (ENI Power System, Inc., model ACG-10) delivers up to a maximum of 1000-W power at 13.56-MHz into a 50-Ω load. The impedance matching network transforms the lamp and cavity electrical load impedance

$$Z = R + jXc, \tag{1}$$

into a constant 50 Ω of pure resistance. The tuner is a cylindrical conductor of length 7.5 cm–12.5 cm and diameter 5 cm–7.5 cm (see Fig.1(a)), placed concentric to the discharge cavity. This tuner provides an adjustable capacitance between the plasma discharge, which acts as the center conductor of a coaxial transmission line, and the outer grounded conductor of the cylindrical cavity. The tuner, therefore, forms both a part of the cabling to the power supply as well as an integral component of the cylindrical cavity itself. This coupling arrangement is unique to this system and is very different from any of the previous RF systems.

†Corresponding author. E-mail: jametepe@ggc.edu
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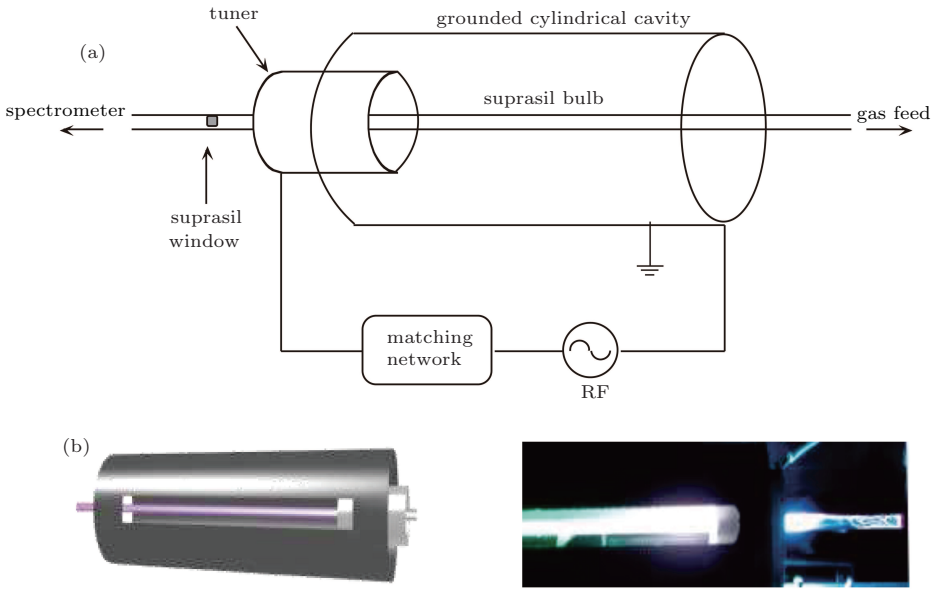


Fig. 1. (color online) (a) Schematic diagram of the 13.56-MHz RF lamp: the discharge tube, tuner, and grounded cylindrical cavity are all concentric to each other. (b) Actual discharge from 13.56-MHz lamp system. The system has been used to produce, Xe_2^*, KrI*, and XeI* discharges and optical efficiency ranges from 6% (for KrI) to $\sim$ 20% (for Xe_2^*).

Figure 2 shows the electrical circuitry equivalent of the system. The lamp arrangement is housed in a Faraday cage to reduce RF leakage.

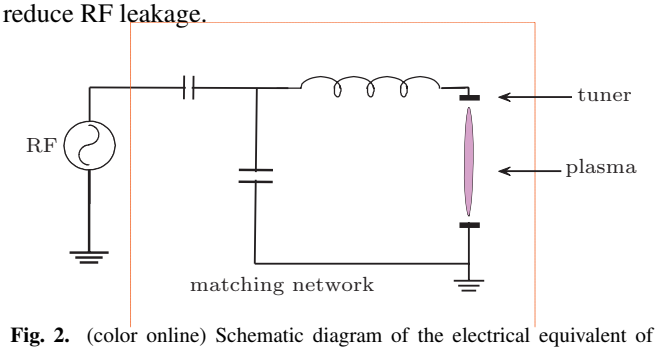


Fig. 2. (color online) Schematic diagram of the electrical equivalent of Fig. 1(a). The tuner forms both a part of the cabling to the power supply as well as an integral part of the cylindrical cavity itself.

2.2. Matching network

The Wheeler's formula,

$$L = \frac{N^2R^2}{9R + 10l}, \tag{2}$$

where L is the inductance in Henrys, R the radius of the coil in inches, l the length along the coil axis, and a Smith Chart were used to predetermine the values of the inductance (L) and capacitance (C) of the matching network components necessary for maximum power transfer at 13.56-MHz (RF generator) into a reactive plasma load from a 50-Ω source impedance. For simplicity, we used a one variable capacitor with a range from 5 pF to 250 pF and can withstand a voltage of up to 15 kV. We used a single layer inductive coil with $R = 1.75$ inch (1 inch = 25.4 mm), $l = 5.4$ inch, and a total of $N = 12$ turns. These parameters of the inductive coil yielded an inductance L between 5 μH–6.5 μH. The single layer coil has advantage of low self-capacitance and high self-resonance frequency. Using a Grid-dip and L–C meters, the above values of L were confirmed to be within ±0.5 μH. The total L and C providing the best matching conditions for our system were found to vary between 4 μH–7 μH and 100 pF–150 pF respectively.

2.3. Advantages of lamp system

No previous RF excimer lamp exists in which the tuner is both a part of the cabling to the power supply as well as an integral part of the lamp cavity allowing for a simpler and compact design. Two key advantages of a simpler and more compact system is the ease of handling the equipment and ease of maintenance. Another advantage is that this design allows for large sample area processing without speckle or interference fringes. Previous RF designs configured to avoid electrodes internal to the plasma require precise tuning. In such systems, the dominant capacitively or inductively coupled designs employ external tunable inductors and/or capacitors to create an impedance match between a tuned cavity and power supply. The present design eliminates this precise tuning requirement, resulting in a simple and compact system. Furthermore, the system is electrode-less and thus eliminates sputtering of electrode materials that tends to blacken the inside wall of the discharge vessel, altering light transmission in an arc lamp. This is particularly important for spectroscopic light sources and for plasma-assisted chemical processing and in most cases for other biological applications. Gas contamination by adsorbed/absorbed and reactive particles from the electrodes is eliminated resulting in prolonged bulb lifetime.

One of the major limitations of photo processing is the lack of sufficient intense UV sources. To date, one of the commonly used UV sources is the low pressure Hg lamp for large area UV processing. However, in most of the cases using Hg lamps, there is the need to photo-sensitize the reaction with Hg. This presents the drawback effects of health

hazards and trace quantities of contamination in the deposited films, limiting the types and quality of film deposited. This new electrode-less 13.56 MHz lamp system could provide an alternate high powered, intense, Hg-free UV source for processing of thin films since there is growing interest in the deposition and processing of thin films at low temperatures to eliminate the problems associated with high temperature processing. Furthermore, the design flexibility allows for both cylindrical and flat panel lamp geometries lending itself to other applications such as biological processes, photolithography processes, and material surface treatment.

3. Results

3.1. Spectra

Below we present figures of Xe_2^*, XeI*, and KrI* spectra data and a table (Table 1) showing the main discharge features observed from the 13.56-MHz system. Our spectral results and observed features are consistent with previous[14–16,18–21] studies of Xe_2^*, XeI*, and KrI*. A deuterium lamp (Oriel BJ2775, NIST-traceable, secondary calibration source) was used for intensity calibration according to the procedure detailed in works of Ametepe *et al.*[16] & El-Habachi *et al.*[18]

Table 1. The main emission features observed in our spectra (Xe_2^*, XeI*, and KrI*) using the 13.56-MHz RF lamp.

Discharge features	Transition	Wavelength/nm	Discharge efficiency
Xe_2^*	$^3\Sigma_u^+ \rightarrow {}^1\Sigma_g^+$	172	Xe_2^* (15%–20%)
XeI*	$B_{1/2} \rightarrow X_{1/2}$	253	XeI* (7%–9%)
I_2^*	$B_{1/2} \rightarrow A_{1/2}$	320	
I^2	$P_{3/2} \rightarrow {}^2P_{1/2}$	206	
KrI*	$B_{1/2} \rightarrow A_{1/2}$	225	KrI* (3%–6%)
I_2^*	$D_{1/2} \rightarrow X_{1/2}$	342	
I^2	$P_{3/2} \rightarrow {}^2P_{1/2}$	206	

3.2. Spectra data

3.2.1. Xe_2^*

Figure 3 shows a typical Xe_2^* second continuum at 172 nm.

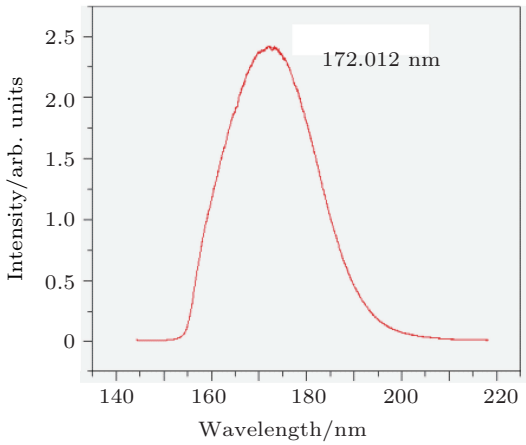


Fig. 3. (color online) This Xe_2^* emission, similar to those observed from other discharge techniques, is the second continuum transition between the lowest vibrational ($^3\Sigma_u^+$) excited state to the repulsive ground ($^1\Sigma_g^+$) state.

This continuum is a transition between the lowest vibrational ($^3\Sigma_u^+$) excited state to the repulsive ground ($^1\Sigma_g^+$) state. Integrating the total light output from 150 nm to 220 nm, and comparing the power emitted to the input RF power, we obtain a lamp conversion efficiency of about 20% at 600 Torr (1 Torr $= 1.33322 \times 10^2$ Pa) and 140-W RF input power for the xenon discharge. These lamp efficiencies compares favorably to previous studies. The lamp efficiency for the Xe_2^* discharge varied between 15% and 20% depending on operating gas pressure and input RF power.

3.2.2. XeI*

Figure 4 shows the XeI* emission at 253 nm, a $B_{1/2} \rightarrow X_{1/2}$ transition. With increasing pressures, we observed the $B_{1/2} \rightarrow X_{1/2}$ transition dominating the spectra emission. Our XeI* emission is similar to other XeI* data obtained from other[2,19,20,22] systems and optical efficiency ranged between 7% and 13% depending on the operating gas pressure and input power. We also observed the 320-nm, $B_{1/2} \rightarrow A_{1/2}$ transition band. The bound-free emission spectra for XeI have been investigated extensively by many groups[25,26] and more recently by Qion-Rong *et al.*,[19] Shuaibov *et al.*,[20] and Zhang *et al.*[21] Also, a detailed theoretical interpretation of these spectra has been achieved successfully by Tamagake,[24] and independently by Tellinghuisen *et al.*[27] and Radzykewycz *et al.*[28] The intense emission band observed at 253 nm, in our study, has potential antibacterial applications since certain DNA chains have a strong absorption band around 250 nm.

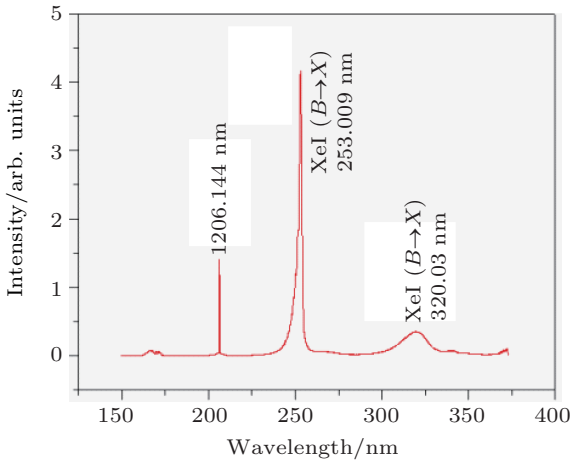


Fig. 4. (color online) The XeI* emission is the transition $B_{1/2} \rightarrow X_{1/2}$. With increasing pressures, we observed the $B \rightarrow X$ transition dominating the spectra emission. The emission is similar to other XeI data obtained from other systems including dielectric barrier discharge.

3.2.3. KrI*

In previous KrI* studies,[23,25,29–32] different reagents and methods were employed to generate KrI* emissions. But most of these studies were limited to low pressure (< 100 Torr) discharges where excimer states are not well populated. As

such, most observed excimer emissions in these low pressure regimes were usually too weak to be useful for obtaining information of molecular potential curves. A recent, relatively higher pressure study of KrI*, performed by Zhang *et al.*[31] demonstrated strong emissions at 191 nm (KrI* $B_{1/2} \rightarrow X_{1/2}$), 225 nm (KrI* $B_{1/2} \rightarrow A_{1/2}$), and 342 nm ($I_2^* D_{1/2} \rightarrow A_{1/2}$).

In this study, we present high resolution KrI spectra data over 160 nm to 360 nm and over the pressure range of 200 Torr to 1500 Torr. Figure 5 shows the KrI* excimer emission in the 13.56-MHz RF lamp at 511 Torr. The 225 nm (KrI*, $B_{1/2} \rightarrow A_{1/2}$) and 342 nm ($I_2^*$, $D_{1/2} \rightarrow A_{1/2}$) emission bands dominate the spectra at 511 Torr. We also observed the 206-nm iodine atomic line $^2P_{3/2} \rightarrow {}^2P_{1/2}$. Optical efficiency of up to 6% was recorded in the KrI* discharges, which had not been reported in previous KrI* studies. Experimental high pressure KrI* spectra data are scarce and the analysis and a kinetic model for this discharge has been presented elsewhere.[33] These high pressure KrI spectral data are valuable to determine accurate potential energy curves for the KrI* excimer.

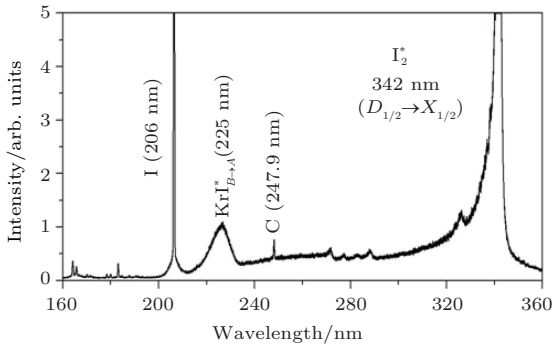


Fig. 5. KrI* excimer emission in the 13.56-MHz RF lamp at 511 Torr. The 225-nm KrI* and 342-nm I* emissions dominate the spectra at 511 Torr and correspond to the $B_{1/2} \rightarrow A_{1/2}$ and $D_{1/2} \rightarrow X_{1/2}$ transitions respectively.

4. Biological application

We performed a series of preliminary UV exposure experiments on E-coli cultures that were prepared by the Biology Department of Hollins University in Roanoke, Virginia. These agar-plate cultures were irradiated using XeI* discharge from the RF lamp system operating at around 1000 W.

Figure 6 shows non-irradiated (left) and irradiated (right) cultures of E-coli. Irradiation times ranged from 5 s to 60 s and at irradiation distances of 5 cm to 20 cm from bulb surface. We limited the exposure to distances greater than 5 cm to minimize substrate heating which could confuse the experiment with thermal damage to the organisms. At all the irradiation distances used in this experiment, from 5 cm to 20 cm, more than 99.9% of the bacteria were inactivated. At 5 cm from bulb surface, using these relatively low colony counts, we obtained an apparent 100% inactivation within 5 s. At 20 cm, we found that the same intensity required 30 s of exposure to yield 99.9% inactivation. These high percentages of inactivation are not entirely unexpected since the XeI* lamp has more than 90% of its UV emission within 5 nm of 253 nm, which gives it a very large overlap with the known primary UV absorption band of DNA. Future, systematic investigation will be required to test the efficacy of this style of lamp against not only E-coli, but other major pathogens to replicate and extend the results of this study. The results of this study, however, do hint at, the effectiveness of this lamp in eradicating pathogens in applications such as food services, clinical practices, defense, and counter-terrorism.

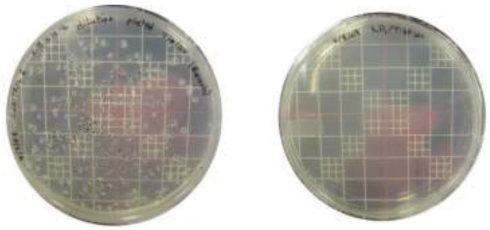


Fig. 6. (color online) Photos of unirradiated (left) and irradiated (right) cultures of Escherichia coli. Irradiations were done at distances of 5 cm to 20 cm from bulb surface. In all cases we obtained over 99.9% inactivation corresponding to disinfection coefficient of 4.3.

5. Conclusion

We have demonstrated the potential use of an electrodeless 13.56-MHz RF system for excimer generation and various discharges including Xe_2^*, XeI*, and KrI* have been investigated and reported. Additionally, optical efficiencies of up to $\sim 6\%$ (for KrI), 13% (for XeI), and 20% (for Xe_2^*) have been recorded. The high optical efficiencies in this lamp make it an attractive candidate for not only laser media, but also for other physical, chemical, and biological applications. We have demonstrated the use of XeI* discharge from this lamp systems for destroying E-coli, hinting the effectiveness of this lamp in eradicating pathogens in applications such as food services, clinical practices, defense, and counter-terrorism. The excitation technique employed in this study offers an interesting addition to the already existing technologies of generating VUV and UV light for various processes. This system is configured to avoid electrodes internal to the plasma and employs mercury free gas mixture, thus allowing longer bulb life.

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