AIP Applied Physics Letters

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Citation: Applied Physics Letters **105**, 013505 (2014); doi: 10.1063/1.4887379
View online: http://dx.doi.org/10.1063/1.4887379
View Table of Contents: http://scitation.aip.org/content/aip/journal/apl/105/1?ver=pdfcov
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Ultraviolet-B radiation enhancement in dielectric barrier discharge based xenon chloride exciplex source by air

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(Received 20 May 2014; accepted 25 June 2014; published online 7 July 2014)

A single barrier dielectric barrier discharge tube of quartz with multi-strip Titanium-Gold (Ti-Au) coatings have been developed and utilized for ultraviolet-B (UV-B) radiation production peaking at wavelength 308 nm. The observed radiation at this wavelength has been examined for the mixtures of the Xenon together with chlorine and air admixtures. The gas mixture composition, chlorine gas content, total gas pressure, and air pressure dependency of the UV intensity, has been analyzed. It is found that the larger concentration of Cl_2 deteriorates the performance of the developed source and around 2% Cl_2 in this source produced optimum results. Furthermore, an addition of air in the xenon and chlorine working gas environment leads to achieve same intensity of UV-B light but at lower working gas pressure where significant amount of gas is air. © *2014 AIP Publishing LLC*.
[http://dx.doi.org/10.1063/1.4887379]

The excimer and excilpex formations in plasma have great potential and can lead to develop plasma based incoherent UV light sources.[1,2] In fact, applications of ultraviolet light have become very important over the years and find diversified industrial and medical applications.[3,4]

In recent decades, interest in development of novel ultraviolet/vacuum ultraviolet (UV/VUV) sources with spontaneous emission has grown.[5] For the UV/VUV production, the plasma based dielectric barrier discharge (DBD) mechanism is the most simple and low cost technique and has great potential to produce high power and efficient incoherent UV radiations.[6] In photobiology, these are most efficient for photo-regulation of vital activity of microorganism, viruses, etc. In photomedicine, these finds huge applications for skin treatment, wound healing, etc.[7] UV-B phototherapy at wavelength 308 nm is most practiced for psoriasis curing.[8] Dmitruck and his team made a prototype of XeCl exciplex lamp and tested it for psoriasis decease and found quite effective.[9] In this paper, an effort has been made to develop a monohalide XeCl* excilpex UV-B source and to improve the radiation efficiency at wavelength 308 nm by air. The developed source is studied for the optimum use of air in the admixture of Xe and Cl_2 gases which can make the excilamp more cost effective.

A single barrier exciplex source has been developed for the experiment, which consists of a quartz tube as a dielectric barrier with the thickness 1.5 mm, inner diameter 19 mm, outer diameter 22 mm, and length 150 mm. Linear strips of Ti-Au with thickness 0.6 μm, width 3 mm, and length 120 mm are grown on the diametrical opposite portions of outer surface by using reactive sputtering system (TFSP-840). A helical tungsten electrode of thickness 0.75 mm and diameter 10 mm coated with ceramics has been inserted inside the quartz tube for the application of high voltage. The outer stripes of titanium-gold are grounded. The developed source is evacuated up to base pressure 1×10^{-5} mbar using rotary and turbo molecular pumps. The outlet of the vacuum system is connected with proper scrubber system to dilute the halide gases. The details of source development could be found elsewhere.[10] Different compositions of the gases (Xe and Cl_2) and (Xe, Cl_2, and Air) have been introduced in the exciplex source. The Xe and Cl_2 gases are controlled by Matheson mass flow transducer series 8272–0453 and D07, respectively. Air has been introduced using leak valve. Research grade xenon with a stated purity of 99.999% as a base gas and fractional chlorine has been introduced for exciplex formation.

The high voltage pulse power supply is used in the experiment to operate the source up to peak voltage 5 kV and frequency up to 30 kHz. The discharge voltage and the total current are measured by high voltage probe (Tektronix P6015A) and the fast response current transformer (Pearson 110). The oscilloscope (Tektronics DPO 4054) registers the current-voltage characteristics. The schematic diagram of the experimental setup is shown in Fig. 1.

The spectral analysis has been carried out using UV and VUV spectrometers (SP-2500i, Princeton Instruments and McPherson 234/302, respectively).We have fixed all the operating parameters of the spectrometer, and for all the electrical operating condition of the DBD source presented

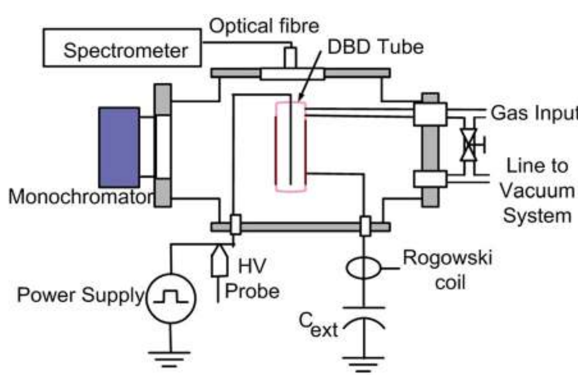

FIG. 1. Schematic diagram of experimental setup.

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in this work, the entrance and exit slit width, integration time, and voltage to the detector have been fixed in the measurements.

Figure 2 shows the typical current-voltage characteristic of the DBD discharge at sub-atmospheric pressure in the developed source. The discharge has been operated in the working pressure range 100–260 mbar at applied pulse voltage 4–5 kV with pulse width 2 μm and operating frequencies 10–30 kHz. The excimer of Xe_2^* (172 nm) and exciplex of XeCl*(308 nm) are promptly observed during the discharge as shown in Fig. 3.

The production of UV/VUV radiation from the excimer/exciplex starts with the excitation, ionization, and dissociation process of the rare gas atom and the halogen molecule. The following transition is responsible of excimer radiation:

$$Xe_2^* \rightarrow Xe + Xe + h\upsilon\,(172\,\text{nm}), \tag{1}$$

where Xe_2^* represents the rare gas excimer, which has been dominantly produced by 3-body recombination.[11]

The observed exciplex molecule XeCl* is dominantly produced by the so-called Harpoon reaction and 3-body reaction,[12,13] and transition is expressed by

$$XeCl^* \rightarrow Xe + Cl + h\upsilon\,(308\,\text{nm}). \tag{2}$$

The Harpoon reaction is expressed as

$$Xe^* + Cl_2 \rightarrow XeCl^* + Cl, \tag{3}$$

whereas 3-body reaction is expressed as

$$Xe^+ + Cl^- + M \rightarrow XeCl^* + M. \tag{4}$$

Here, M is collisional third partner, which is basically an atom or molecule of working mixture. There are large number of radiation exciplex associated with xenon chloride, $XeCl^*$ (D-X, B-X, B-A, and C-A transitions with transition wavelength 236 nm, 308 nm, 340 nm, and 345 nm, respectively). However, Xe_2^* (172 nm) excimer, triatomic rare gas–halide exciplex Xe_2Cl^* (490 nm), and Cl_2^* (259 nm) excimer with the binary gas mixture of Xe/Cl_2 are also possible. In our experiments, $XeCl^*$ and Xe_2^* radiation with 308 nm and 172 nm wavelength, respectively, are observed dominantly.[14,15]

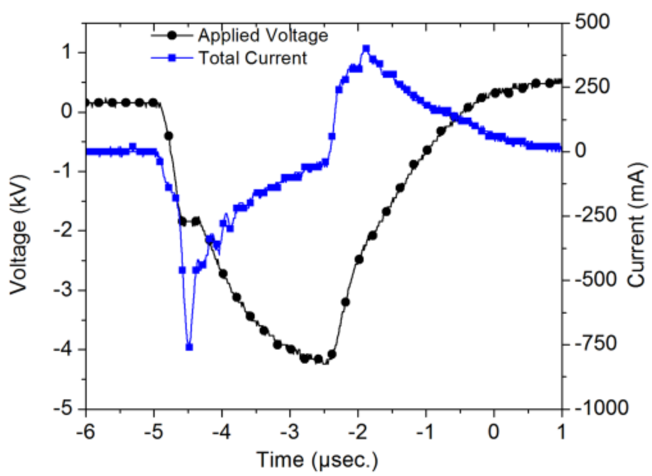
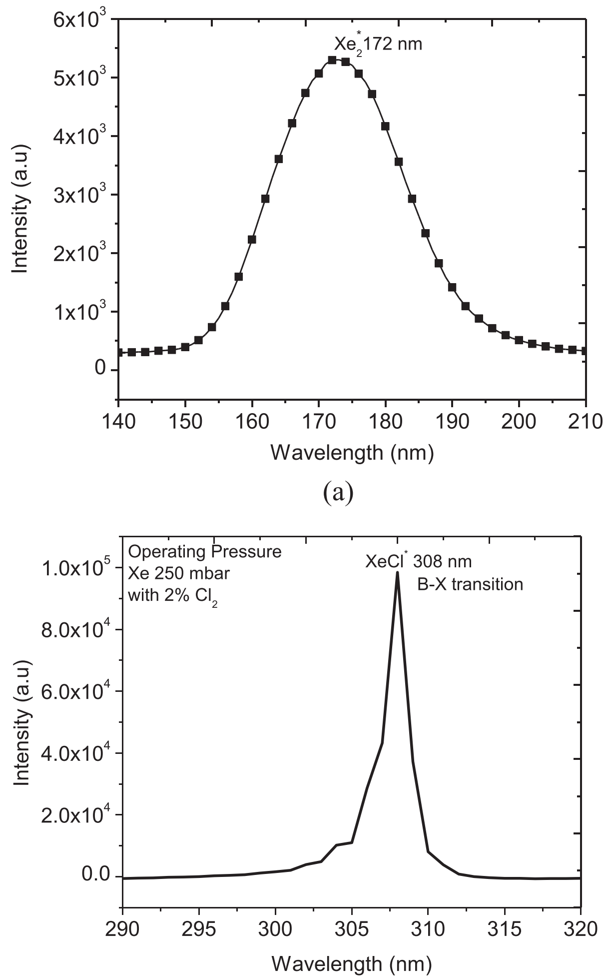


FIG. 2. V-I characteristics of DBD lamp at 4 kV, 25 kHz with a gas mixture of xenon at 250 mbar, and 2% of chlorine.



(b)

FIG. 3. Typical emission spectrum with a gas mixture of xenon at 250 mbar and 2% of chlorine (a) for Xe_2^* 172 nm measured using McPherson-302 (b) for the XeCl* (308 nm) measured using SP-2500i, Princeton Instruments.

To optimize the composition of the gas mixture of the rare gas and the halogen contents, the number of experiments has been carried out. The effect of frequency and pressure on the radiations of 308 nm and 172 nm wavelengths has been studied first. Initially, the xenon gas has been filled at working pressure 250 mbar, and later chlorine has been introduced continuously with concentration 2%, 4%, 6%, 8%, and 10% to record the intensity of 308 nm radiation (see Figure 4).

It has been observed that this radiation is maximum at 2% chlorine and with the increase in chlorine content, the UV-B radiation decreases continuously. It is because at higher concentration of the chlorine, quenching mechanism of XeCl* excimer becomes dominant.[11] In fact, in the intervening time, the ion transfer is enhanced from Cl^- to Cl_2 due to increase in chlorine concentration which ultimately reduces the Xe^+ ions, and is responsible for the formation of $XeCl^*$ via 3-body reaction. It has also been further seen that with the increase in concentration of the xenon there is decrease in UV intensity, which might be due to the collision-induced quenching reaction between XeCl* and Xe

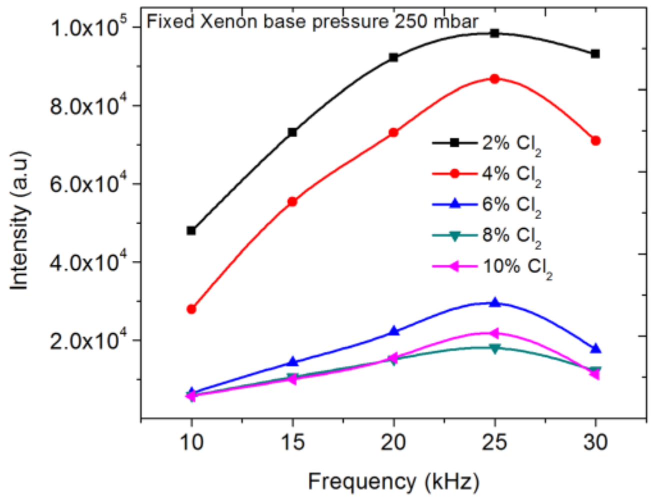


FIG. 4. Xe/Cl_2 intensity variation with different chlorine concentration at constant base pressure of Xenon 250 mbar.

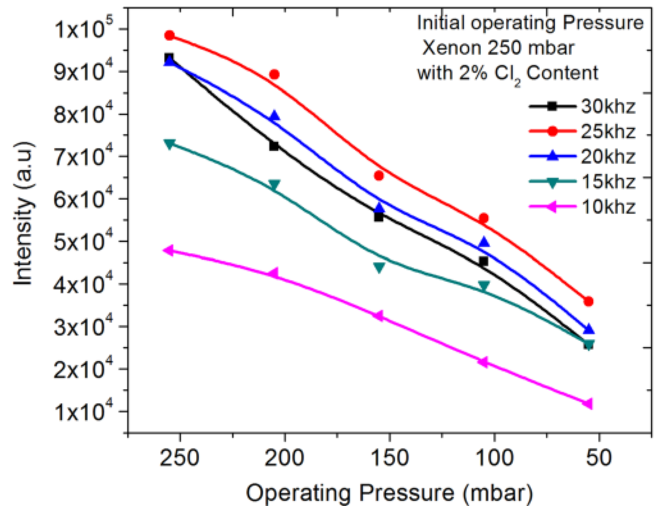


FIG. 6. Variation of 308 nm line with pressure at different operating frequency (Inverse X-axis).

and trimer (Xe_2Cl^*) can also be formed at higher pressure with the increment of Xe.[11] Baadj *et al.*[16] have done detailed analysis on the reactions involved in the xenon/chlorine mixture.

The effect of frequency for different chlorine concentrations with 250 mbar xenon working pressure is also shown in Figure 4. It depicts that the maximum intensity of the UV-B photon has been observed for every composition of the Xe/Cl_2 at 25 kHz frequency. The similar effect of frequency has been observed for 172 nm spectral line of Xe_2^* excimer (see Figure 5).

The effect of the operating pressure is also seen on the intensity of UV-B radiation. For this, the working pressure 250 mbar of Xenon has been fixed and then 2% Cl_2 gas has been added. Later, this total pressure has reduced to see the effect of frequency. The intensity of UV-B radiation decreases with the decrease in the admixture pressure as shown in Figure 6. It is clear that UV-B emission is greatly dependent on the gas mixture used for the generation of plasma and with the pressure 250 mbar of xenon and 2% of chlorine; our developed configuration produces maximum intensity.

Recently, Rodriguez-Mendez and his team have shown that with the increased concentration of the oxygen, an effective output of the DBD has been observed, for destruction of the different bacteria.[17] So, to elucidate the effect of air in our experiment, we added varied amount of air during the discharge. We have first set the pressure of DBD excimer lamp, with 250 mbar Xenon plus 2% chlorine and then continuously decreased the pressure up to 105 mbar, where minimum discharge of Xe/Cl_2 is seen. Later, atmospheric air is added stepwise in this binary mixture and the radiations are measured (see Fig. 7).

Figure 7 shows that in the observed spectrum 308 nm line radiation enhancement with the increment of air. It become maximum when around 60 mbar air pressure has been introduced, and later it is decreasing for further addition of air. It is significant to note that at around 47% of air admixture in the working pressure of Xe and chlorine keeping pressure ~165 mbar (see Fig. 7), the level of UV-B photon radiation is same as measured for binary mixture of xenon and chlorine keeping operating pressure ~250 mbar (see Fig. 3(b)). The enhancement in the radiation is mainly due to the hydroxyl molecular band peaking at wavelength 309 nm ($A^2\sum^+$, $\upsilon = 0$-$X^2\Pi$, and $\upsilon' = 0$) generated from the

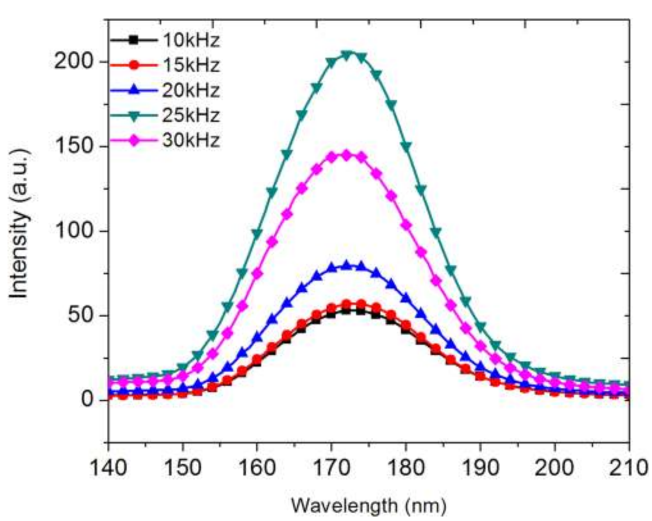


FIG. 5. Effect of frequency on Xe_2^* excimer (172 nm) in VUV range.

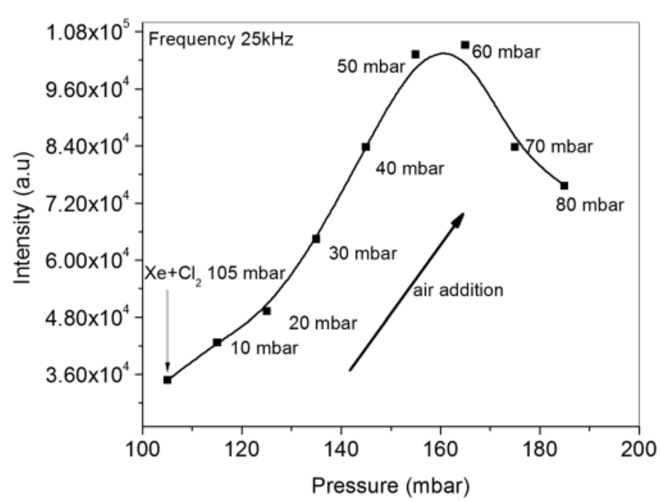


FIG. 7. Intensity variation of 308 nm line with air mixture.

moisture present in the air.[17,18] Shi and Kong have also shown the results of argon in DBD with air and they show significant hydroxyl line radiation at 309 nm.[19]

In fact, hydroxyl radicals are created mainly in the excited states and rapidly quenched for the particles present in the atmosphere and comes to the ground state by radiating photons at peak wavelength 309 nm in very short time. This radiation adds up in the UV-B 308 nm radiation because XeCl* exciplex radiation has about 5–10 nm spectral band.

In summary, we report here a study of XeCl* exciplex source with the emission of UV-B 308 nm line excited by a dielectric barrier discharge in different chlorine contents and also with the admixture of air. The developed source shows that at around 47% of air admixture in binary mixture of Xe/Cl_2 gives intense radiation of 308 nm wavelength even at lower working pressure than that of pure binary mixture.

One of the authors, Ms. Pooja Gulati, acknowledges CSIR for providing CSIR-SRF. The authors also wish to thank CSIR for providing the funds under Network program PSC0101.

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