Study and optimization of mercury free fluorescent signs

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Short pulse excitation and analysis of low pressure Ne/Xe plasma in cold hollow electrode fluorescent tube is reported. A very significant enhancement of light output, lamp efficiency and lifetime has been achieved by optimizing the pulse duration and the gas mixture pressure. The time resolved spectroscopy of radiating species - xenon atoms, xenon ions, neon atoms and (XeNe) molecules – in combination with the voltage and current measurement provide a comprehensive description of the glow ignition and development, and of the afterglow characteristics.

1. Introduction

This work deals with the development of mercury free low pressure glow discharges likely to be used for publicity lighting. There exists today an important need for alternative solution to mercury based fluorescent lamps used for general lighting and many other applications [1]. Rare gas plasma, including xenon as an ultraviolet emission element, is one of the main promising and environmentally friendly solution, providing that the energy efficiency of such new lamps, could be enhanced to an economically viable level. Low pressure rare gas discharge systems have been recently investigated [2,3] showing evidence for the severe constraints for an industrial lamp realization. In this work, experimental work has been performed to investigate rare gas hollow cathode discharge potentialities. Both AC driven and pulsed powered plasma characteristics have been measured. Time resolved vacuum ultraviolet (VUV) and visible emission spectroscopy together with electrical measurements are the main diagnostics used to obtain a comprehensive description of the plasma kinetic processes whose analysis is at the base of the sign performance optimization. Besides the previously reported four times enhancement of both output luminous flux and efficiency of xenon pulsed discharge in comparison with conventional AC hollow cathode fluorescent tubes[4], the use of pulsed excitation also indicates that high luminous fluxes can be obtained for higher mixture pressures excited by short duration electrical pulses. A brief experimental set up description and the main result on the influence of gas mixture pressure on VUV production and sign operation, including ageing study, is proposed in section 2. The next paragraph deals with the description of the active phase of the discharge, i.e. glow phase, and the analysis of the plasma excitation mechanisms. The section 4 enlighten the crucial role of post discharge phase on the tube performance and the possibilities to try to achieve the higher efficiency in pulsed neon-xenon lamps.

Summary of the results and perspectives are then proposed in the last paragraph of the paper.

2. Experimental setup and gas mixture pressure analysis

The Fig. 1 presents a schematic diagram of the experimental set up used for the rare gas plasma production and diagnostics. The experimental discharge vessel is a T-shaped fluorescent tube, equipped with two conventional hollow electrodes. The results presented in this work have been obtained using iron/nickel cylindrical electrodes, 40 mm in length, 10 mm in diameter. The inside of these electrodes is covered with an electron emitter barium fluoride layer. The 3 mm thick ceramic ring is used to prevent from strong sputtering at the plasma facing electrode edge. Three internal diameters (10, 13 and 18 mm) of the tubes have been used, leading to very similar behaviour, the discharge length being typically of 50 cm. The inside surface of the sign is covered with a blue or green phosphor, used in mercury based fluorescent tubes.

The T branch of the sign is first connected to the gas handling system, including gas flow controllers, pressure gauges and a vacuum pump to monitor the gas mixture composition. Before filling the tube with high purity gases, the vessel is heated up to about 350 °C, via the neon tube bombarder to remove the impurities trapped in the glass, phosphor and metallic electrodes of the sign. Finally, the T branch is optically connected to a VUV-UV-Visible (110-900 nm) 0.4m focal length spectrometer equipped with two gratings (300 and 1200 grooves/mm respectively). Both photomultiplier tubes and an intensified CCD camera are connected to the spectrometer for the measurement of time resolved emission spectra of the discharge. In this work, the VUV measurement of the xenon resonance line radiation at 146.9 nm has been performed through a vacuum evacuated glass capillary equipped with a MgF_2 window. The VUV window is set at the same position than the phosphor layer, through the T branch. In this way, the radiation trapping in the xenon gas is included in the measurement and corresponds to the phosphor excitation condition.
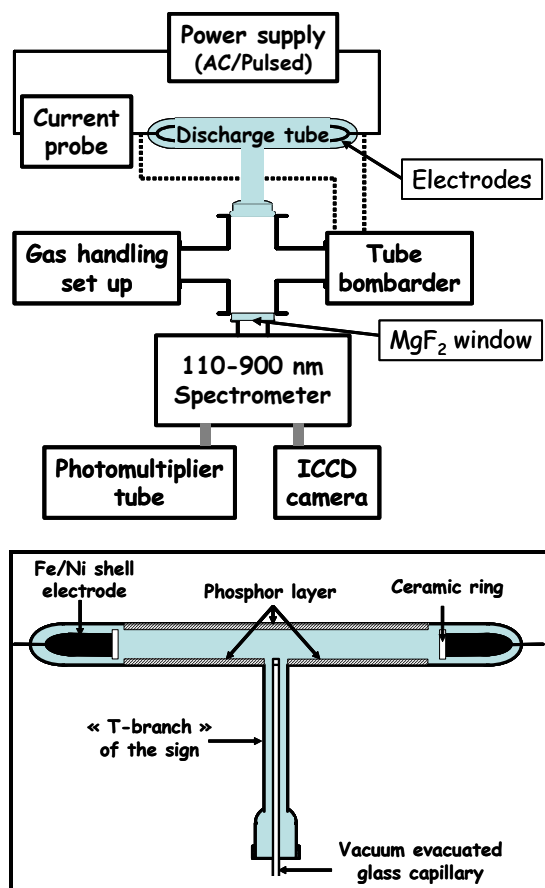
Fig. 1 Experimental setup schematic and detail of the fluorescent tube configuration.

In our work, AC excitation stands for whether 50 Hz or 25 kHz sinusoidal waveforms, and pulsed excitation corresponds to the application of microsecond duration high voltage (about 1 kV), high peak current (a few A) pulses, delivered at repetition rate in the kHz range.

The figure 2 presents the evolution of illuminance of a Ne/Xe (100/1) filled sign as a function of the xenon pressure both for AC and pulsed excitation. For AC driven lamps, the higher illuminance is obtained for the lower Xe content. Unfortunately, this has a negative influence on the ageing performance of the rare gas fluorescent lamp. It has been previously reported that one the main limitation for low pressure xenon lamp is the fast trapping of xenon atom during the discharge operation [2,4,5].

For pulsed excitation, the best lamp operation occurs for a mixture pressure around 70 mbar. The variation of the mixture pressure from 10 to 70 mbar leads to a 50 % increase of the illuminance level and to about a twenty times higher lifetime. The illuminance obtained with 50 mbar Ne/Xe pulsed lamps range from a few hundreds of Lux to 6000 Lux when varying the electrical power input. The illuminance of a mercury based signs powered by an AC 25 mA ballast was measured to be of 6000 Lux for the same tube length and diameter, same electrode characteristics and the same green emitting phosphor

coating as those used for the mercury free, Ne/Xe filled sign. Lamp operation ageing studies have been successfully performed during time period of about 4000 hours. No evidence for cathode sputtering, xenon trapping, chromatic coordinates changing have been measured.
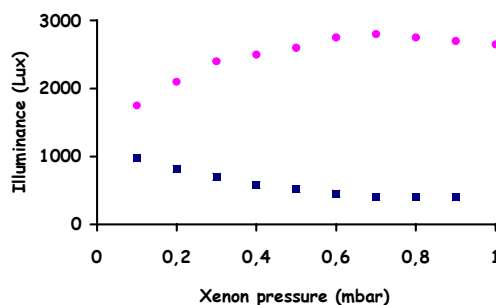

Fig. 2 Evolution of illuminance versus xenon pressure in a 1% Ne/Xe mixture powered by : AC signal - squares and pulsed signal - dots.

The Fig. 3 presents the VUV spectra of 10, 50 and 90 mbar Ne/Xe mixtures containing 1 mbar of xenon. The measurements are performed at the position of the phosphor layer, to include the resonance line trapping effect at this position. It appears that with increasing the mixture pressure, the 147 nm line of xenon is much more intense and that a significant fraction, estimated to about 10%, of the VUV excitation spectra consists in the (XeNe)* radiation, which is recorded as a blue wing of the Xe resonance line. The inset in figure 3, provides the 90 mbar spectrum in a logarithmic scale to have a more detailed view of the blue wing structure. The (XeNe)* spectrum is quite identical with that reported in [6].
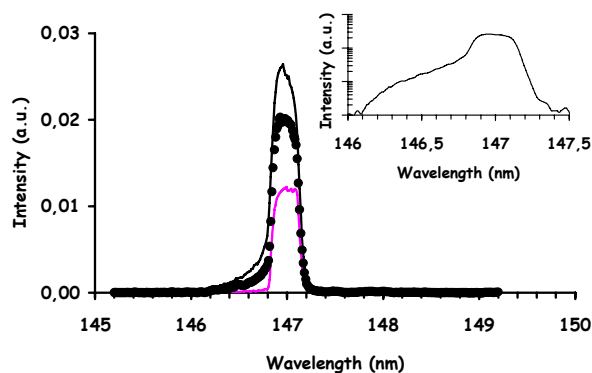

Figure 3 VUV spectra of Ne/Xe mixtures containing 1 mbar of Xe with: 10 mbar of Ne- thin grey curve, 50 mbar of Ne-dotted spectrum and, 90 mbar of Ne-black curve. The inset is a logarithmic plot of the 90/1 mbar mixture.

3. Glow phase analysis

The temporal behaviour of light emission for the duration of the pulse was studied in detailed in this work for the optimization of the rare gas based pulsed plasma. The pulse duration increase results in a larger electrical power input across the sign if all other parameters are kept constant. In our case, the main interest for fluorescent lamp development is to keep the electrical power input at a constant level and to investigate if the efficiency is influenced by the way this electrical power is injected. A very easy way to work at constant power input is to compensate the pulse duration variation by a corresponding repetition rate matching. This method was proven to be reliable for the sign performance measurement, but one has to keep in mind that changing the pulse repetition rate has an inherent influence on the glow re ignition at the instant of the voltage pulse application. Higher repetition rate correspond to higher residual electronic density [7] and an earlier current increase across the electrodes. Nevertheless, for any pulse duration and repetition rate experienced in this study, the active phase of the discharge, i.e. glow, always exhibit two distinct phases.

In Fig. 4, the pulse duration is 5 µs and the repetition rate is of about 2 kHz, the power input is close to 30 W. In this case, the transition between the two glow phases is observed 2 µs after the pulse onset. The transition between the two glow phases is shifted to earlier or later time as the repetition rate is increased or decreased while keeping a five microseconds pulse duration.

In the first 2 µs, the voltage signal is of large amplitude, that is the electric field is intense and the main excitation goes in the Xe lines, specially the 147 nm resonance line. The current amplitude during this first phase is small, a few hundreds of mA.

Then as the current density rises, the glow turns in the second phase during which a linear and strong increase of the current is measured up to the end of the pulse. This current rise is closely related with the population of neon excited state. It has been measured that the 147 nm line temporal behaviour during the glow is quite similar with the voltage waveform, indicating that the resonance level is excited by high energy electrons during the high field phase. The neon temporal behaviour is much more similar with the current waveform. Penning reaction plays an important role leading to a significant increase of the Xe ions density. At the beginning of the second phase, the electronic temperature starts to decrease, depopulation of the resonance level at the benefit of higher lying xenon levels [7] is measured (not shown in figure 4). The production of Xe ions and excited neon states finally lead to the population of xenon ion excited states as illustrated by the 529,2 nm line in figure 4.

4. Afterglow analysis

At the end of the current pulse, the electron temperature falls rapidly, and the plasma turns into its relaxation phase. The afterglow emission essentially results from the recombination of the xenon ion produced during the pulse. Both the collisional radiative (reaction (1)) and dissociative (reaction (2)) recombination reactions [8] can play a role in the plasma decay:

$$Xe^+ + 2\ e^- \rightarrow Xe^* + e^-, \qquad 2,8 \times 10^{-27}\ Te^{-9/2}\ cm^6 s^{-1} \quad (1)$$

$$Xe_2^+ + e^- \rightarrow Xe^* + Xe, \qquad 2,3 \times 10^{-6}\ Te^{-0,7}\ cm^3\ s^{-1} \quad (2)$$

In our experimental conditions, the strong current amplitude at the end of the pulse is connected with the production of a low pressure plasma having a rather high electronic density and moderate electronic temperature. These discharge characteristics are in favour of the collisional radiative recombination reaction. At moderate pressure, 50 mbar in the case of figure 4, the formation of Xe_2^+ molecular ions through the three body reaction involving one neon atom, is of large importance in the plasma kinetics and the successive dissociative recombination reaction appears as the main plasma recombination path. As described in the preceding paragraph, the density of the xenon ions is strongly connected with that of the neon through penning reaction. The figure 5 illustrates the influence of the pulse duration on the xenon ion kinetics. As the pulse duration is increased, the current amplitude is larger, the xenon ion density increase together with that of neon excited levels leading to a very pronounced rise of the excited ionic state of xenon which will first radiated their energy and produce a high xenon ion density.
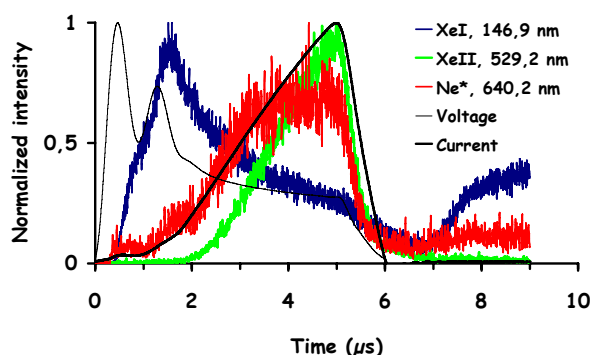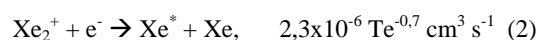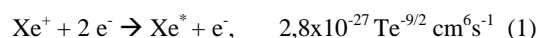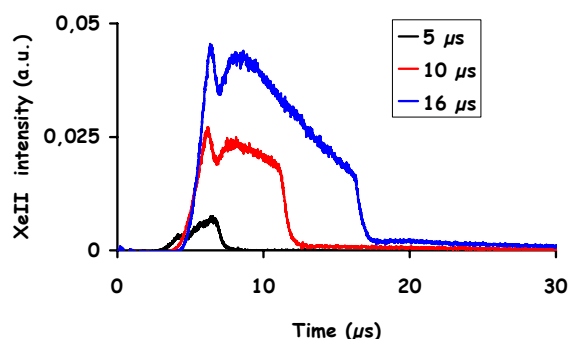

Fig. 4 Temporal profile of : thin line-voltage, thick line-current, black signal-147 nm line, grey signal-640,2 nm neon line and light grey signal-529,2 nm xenon ion line. Ne/Xe mixture excited by 5 µs duration pulses.

Fig. 5 Temporal profile of the XeII line at 529,2 nm for 5, 10 and 16 microsecond pulse durations.
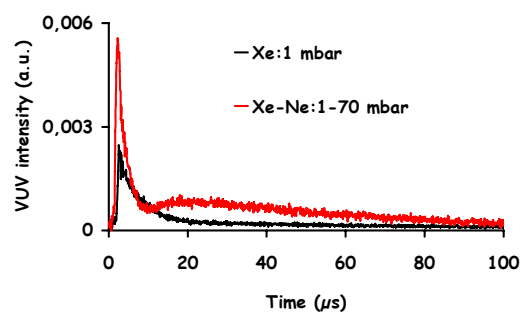

Fig. 6 Xe, 147 nm temporal profile for : black line- one mbar pure xenon sign and with the admixture of 70 mbar of neon in xenon-grey trace.

It has been measured that the afterglow signal is more and more predominant as the pulse duration is increased. The balance between the VUV pulse intensity and the VUV afterglow contribution has a direct influence on the sign efficiency. For the shortest pulse durations, the current is not enough intense to produce a sufficient xenon ion density and consequently a very small afterglow signal is measured. Even with very high repetition rate operation, the short pulse excitation leads to a less efficient regime. On the other hand, for pulse duration larger than 10 µs, the current and electronic density amplitude are to high, leading to the discharge constriction around the longitudinal axis of the plasma. Best results were obtained for pulse durations ranging from 2 to 10 µs.

After the pulse duration optimization, the measurement of the afterglow amplitude as a function of the mixture pressure has been performed. The figure 6 presents the 147 nm signal measured in a sign filled with only one mbar of pure xenon and with the adjunction of 70 mbar of neon. In both case, the pulse duration (2 µs) is kept constant while a very limited pulse repetition rate matching is realized to achieve a constant input electrical power.

The figure 6 indicates that the VUV radiation is much more intense during the glow phase for Ne/Xe mixtures than for pure xenon discharge and that the recombination amplitude during the afterglow is also much more important with neon admixture. This confirms the crucial role of neon in the production of high xenon ion density plasma. It must be noticed that pure xenon discharge can not be homogeneously produced for xenon pressure higher than about 20 mbar in cold hollow cathodes discharge conditions. The excitation of binary mixtures, especially Ne/Xe mixture, is a convenient way to obtain a high VUV emission in stable and homogeneous discharge.

5. Conclusion

The pulsed excitation of Ne/Xe plasma has been performed in fluorescent tubes equipped with conventional cold hollow electrodes and covered with phosphor usually used in mercury based sign industry. It has been measured that pulsed excitation, with microsecond pulse duration and kHz repetition rate, offers very large benefits in comparison with AC driven lamps. With pulsed excitation, the illuminance amplitude can be increased to level suitable for fluorescent publicity lighting applications, by increasing the xenon pressure in a Ne/Xe mixture plasma. This pressure increase offers also the possibility to achieve a continuous few thousand hours of operation of the sign without any evidence for cathode sputtering or lamp colour variation, as in AC drive mode where xenon trapping occurs, during the ageing experiment. The VUV spectra show evidence for a significant contribution of (XeNe)* radiation in the phosphor excitation for the higher pressure mixtures.

The time resolved spectroscopic measurement of xenon, xenon ion and neon lines together with the recording of voltage and current waveforms enable us to get a comprehensive description of both the glow and afterglow characteristics. The plasma ignition at the pulse onset is strongly influenced by the pulse repetition rate and the glow signal reveals two distinct phases. In a first step the excitation occurs with high electric field condition at the benefit of xenon lines, then the second phase occurs with a few microsecond of delay. This second part of the glow development is characterized by a strong current amplitude increase, the excitation of neon excited levels and at the end of the pulse by the population of xenon ionic excited states. This leads to the production of a low pressure, high electronic density and low electronic temperature plasma. The optimization of the pulse duration and mixture pressure indicate the very strong importance of the afterglow signal in comparison with the VUV signal produced during the active phase of the discharge. Pulse duration ranging from 2 to 10 µs, and gas mixture pressure ranging from 50 to 80 mbar appears as the best conditions for the development of the highest

efficiency mercury free signs. Longer pulse duration or higher pressure mixture were shown to lead to less homogeneous plasma due to discharge constriction along the longitudinal axis. Work is still in progress to try to achieve higher efficiency, in comparison with those obtained in mercury based systems.

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