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Effect of Rare Gas Admixture on N_2/O_2 Gas Mixture Discharge

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Abstract: The effect of rare gas (He, Ne, Ar, Kr and Xe) admixture on the emission intensity of N_2/O_2 gas discharge has been studied aiming to apply the UV emission for sterilization. N_2/O_2 discharge emits intensive UV light in the 200-400 nm region which has germicidal effect and so it can be used for sterilization purpose. Experiments were carried out with He, Ne, Ar, Kr and Xe gases at different percentages of concentration to enhance the intensity of the emission from N_2/O_2 discharge. It has been observed that at low concentration <5%, these gases have no effect on the emission intensity whereas the emission intensity in both the UV and visible ranges decrease as the rare gas concentration is increased. Rare gases behave as buffer gases which cause collisions with the other co-existing molecules and decrease the emission intensity.

Key words: Rare gas, UV emission, nitrogen, oxygen, visible emission, microwave discharge, buffer gas

INTRODUCTION

UV radiations have many applications in biological, physical and chemical processes; such as disinfection of drinking water (Massachelein, 2002), sterilization of medical equipments (Nagatsu *et al*., 2003), photochemical synthesis (Legrini *et al*., 1993) and photo-enhanced chemical vapor deposition (Esrom and Kogelschatz, 1992). Now a days, UV irradiation of water has been established as a mature alternative to chlorination for disinfection of drinking water. UV light within the germicidal region from 200-280 nm results in the inactivation of the microorganisms of microbiological system by photochemically altering the DNA in the cell (Al-Shamma *et al*., 2001).

Mercury which is used today as filling element in the UV light source is highly toxic and potentially carcinogenic. These dangers, even in very small quantities can eventually lead to neurological system and brain damage in humans. Along with other harmful elements such as lead, cadmium and hexavalent chromium, environmental groups worldwide are calling for limit on the use of mercury in electrical and electronic equipment.

European Union's RoHS (Restriction of Hazardous substances) has already banned the use of lead, mercury, cadmium, chromium, polybrominated biphenyls and polybrominated diphenyl ethers from electronics sold in EU member states beginning 1 July, 2006. So, the replacement of mercury in conventional UV lamps by other components is highly desirable for environmental reasons. Demonstrated low pressure mercury free plasma light source using rare gas mixture (Uhrlandt *et al*., 2005). In gas discharge, admixture of rare gases with other gases is a common technique for realizing the desired condition for application. Examined the effect of rare gas admixture with oxygen discharge on the vibrational and rotational temperatures of the OH radical by Optical Emission Spectroscopy (OES).

They used helium, neon, argon, krypton and xenon as admixture components with oxygen (Sakamoto *et al*., 2006) studied glow discharges in mixtures of xenon with other rare gases aiming to use it for mercury free UV light source to replace the mercury UV lamp (Smith, 1936) measured the intensity distribution of continuous spectrum of hydrogen by admixture of rare gases (He and Ne).

The effect of these gases was to shift the maximum of intensity to longer wavelength (Fozza *et al*., 1998) investigated the VUV to near infrared emissions from molecular gas-noble gas mixtures (H_2-Ar and O_2-Ar) to obtain very intense VUV emissions to apply it for the photochemical treatment of polymer surface.

The effect of gas composition on spore mortality has been investigated by Lerouge *et al*. (2000) using gases such as pure O_2, O_2/CF_4 and O_2/Ar (Kitsinelis *et al*., 2004) studied of spectral output with a range of different buffer gases (neon, argon, krypton and mixtures of these gases). It was observed that near-UV output is maximized when argon is used.

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In this study, the effect of rare gas admixture with N_2/O_2 gas has been discussed. Experiment to study the effect of inert gases on the intensity of the emission was carried out. He, Ne, Ar, Kr and Xe gases were used at different percentages of concentration. It has been observed that at low concentration <5%, these gases have no effect on the emission intensity whereas the emission intensity in both the UV and visible ranges decrease as the rare gas concentration is increased.

MATERIALS AND METHODS

The schematic diagram of the experimental set-up for N_2/O_2 gas mixture microwave discharge is shown in Fig. 1. A one side closed quartz tube with 500 mm length, 15 mm outer diameter and 13 mm inner diameter was used for discharge. The tube was inserted into a discharge applicator with dimensions of 260 mm length, 100 mm width and 30 mm height. The open side of the quartz tube was connected to a gas supplying line for evacuating by using a oil rotary pump and for filling with different concentration of N_2 and O_2 gas (both N_2 and O_2 were 99.99995% pure). A magnetron power source operating at 2.45 GHz with power rating of up to 1.5 kW was used in the experiment.

After filling the discharge tube with gases, microwave power of 100 W was supplied to the applicator and discharge was produced with the help of Tesla coil. Reflected power was adjusted to 0 W by using the tuner. Optical emission spectroscopy was carried out by using two sets of fiber coupled spectrometer for UV (HR- 4000, Ocean Optics; Slit: 5, Bandwidth: 200-400 nm) and for visible light (USB-4000, Ocean Optics_GSlit: 25, Bandwidth: 200-850 nm) through a observation port consisting of a metal tube with 10 mm inner diameter and 40 mm length on the side wall of the microwave applicator. The optical fiber was set at the end of the port which was 10 cm away from the center of the discharge tube.

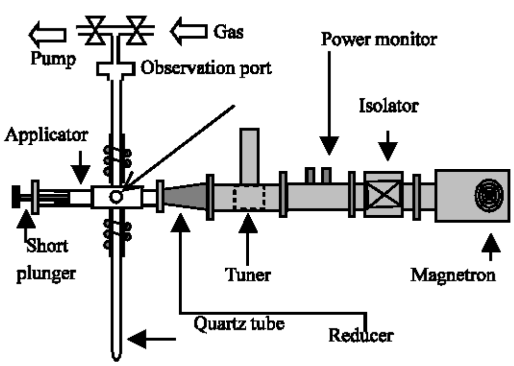

Fig. 1: Schematic diagram of the experimental setup

RESULTS AND DISCUSSION

Effect of admixture of Ar: $N_2/O_2/Ar$ discharge was produced at different concentration of O_2, N_2 and Ar gas to investigate the effect of Ar admixture on the UV intensity emitted from N_2/O_2 discharge. Figure 2a, b show the emission intensity at different gas composition in the UV and visible range, respectively. It can be seen from Fig. 2a that UV emission intensity changes with Ar concentration. However, the shape and peak position remain same in the UV region up to 50% of Ar concentration. At 2% O_2 and 90% Ar, no emission was observed in the UV region. From Fig 2b, it is seen that emission intensity in the visible range also changes with Ar concentration.

In the visible region, some additional peaks (at 698, 708, 739, 752 and 764 nm) from Ar were also observed at high concentration of Ar. As the concentration of Ar increased the peaks also increased. It was observed from both Fig. 2a, b that at low concentration <5%, Ar gas has no effect on the emission intensity. However, the emission intensity in both the UV and visible ranges decreased as Ar gas concentration was increased >5%. The reason for the decrease of intensity is that the UV intensity emitted from NO molecule is maximum at 80% N_2 and 20% O_2.

Below this concentration of N_2, the intensity decreased. When Ar concentration is increased, N_2 concentration decreased in the gas mixture and so the emission intensity also decreased with increasing Ar concentration. Emission in the 300-400 nm range is from 2nd positive system of N_2. It was also observed that the intensity in this range decreased with decreased concentration of N_2. In case of $N_2/O_2/Ar$ discharge as the concentration of N_2 decreased with the increase of Ar concentration, the emission intensity within this region also decreased.

The emission intensity in the visible region which is from 1st positive system of N_2 also decreased when N_2 concentration is decreased. So in $N_2/O_2/Ar$ discharge the intensity of emission also decreased for the same reason as mentioned before. The other reason for the decrease of intensity in both UV and visible range is that Ar gas behaves as buffer gas in the mixture. Buffer gases caused collisions with the other co-existing molecules and decreases the emission intensity.

Effect of Kr admixture: Figure 3a, b show the emission intensity from $N_2/O_2/Kr$ discharge at different gas composition in the UV and visible range, respectively. From Fig. 3a, it was observed that the shape and peak position remained same in the UV region although,

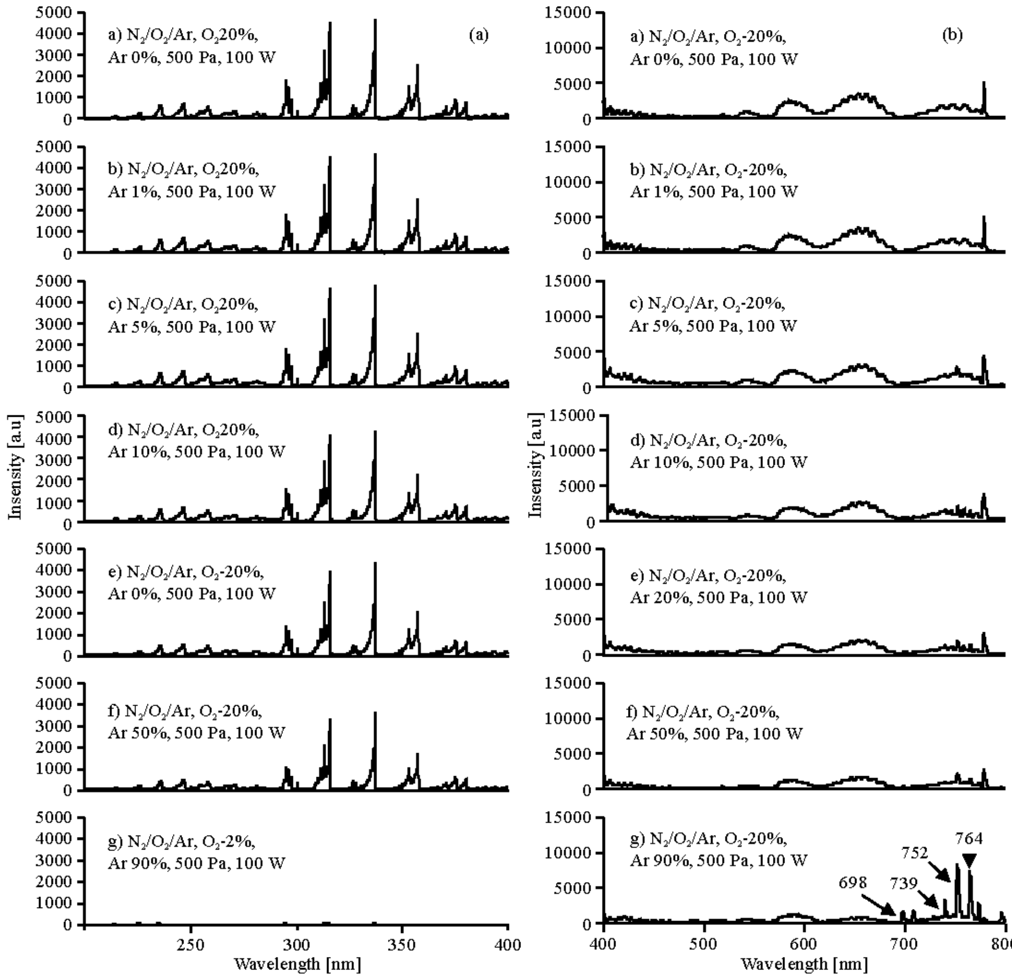

Fig. 2: Emission spectra from N_2/O_2 /Ar microwave discharge at various Ar concentration (a) in the UV range and (b) in the visible range

emission intensity in this range changed with Kr concentration. In the visible region as shown in Fig. 3b, emission intensity also changed with Kr concentration and some additional peaks from Kr appeared at high concentration of Kr. The admixture of Kr showed almost the same effect as of Ar. As <5% concentration, Kr has also no effect on the emission intensity. However >5% of concentration, the emission intensity in both the UV and visible ranges decreased. The reasons for the decrease of intensity are same as mentioned in case of Ar admixture.

Effect of Xe: The effect of Xe admixture on the emission intensity emitted from N_2/O_2 discharge in the UV and visible range is shown in Fig. 4a, b, respectively. From Fig. 4a, b, it has been observed that admixture of 1% Xe has no effect on the emission intensity in both UV and visible region. However at 5% and above this concentration the intensity gradually decreased. It is also observed that (Fig. 4b) at higher concentration of Xe some additional peaks from Xe at 467, 492 and 713 nm appeared in the spectrum.

Effect of admixture of He: Figure 5a, b show the effect of He admixture on the emission intensity emitted from N_2/O_2 discharge in the UV and visible range, respectively. Figure 5a shows that the emission intensity changed with He concentration but the peaks position remained same in the UV range. In the visible region, the intensity gradually decreased with increased He concentration at 90% He and 2% O_2 concentration, He peaks at 487 and 747 nm were observed. The one reason for the decrease of intensity is the decrease of N_2 concentration with the increase of rare gas concentration in the mixture and the other reason is the behavior of He gas in the mixture as buffer gas. Buffer

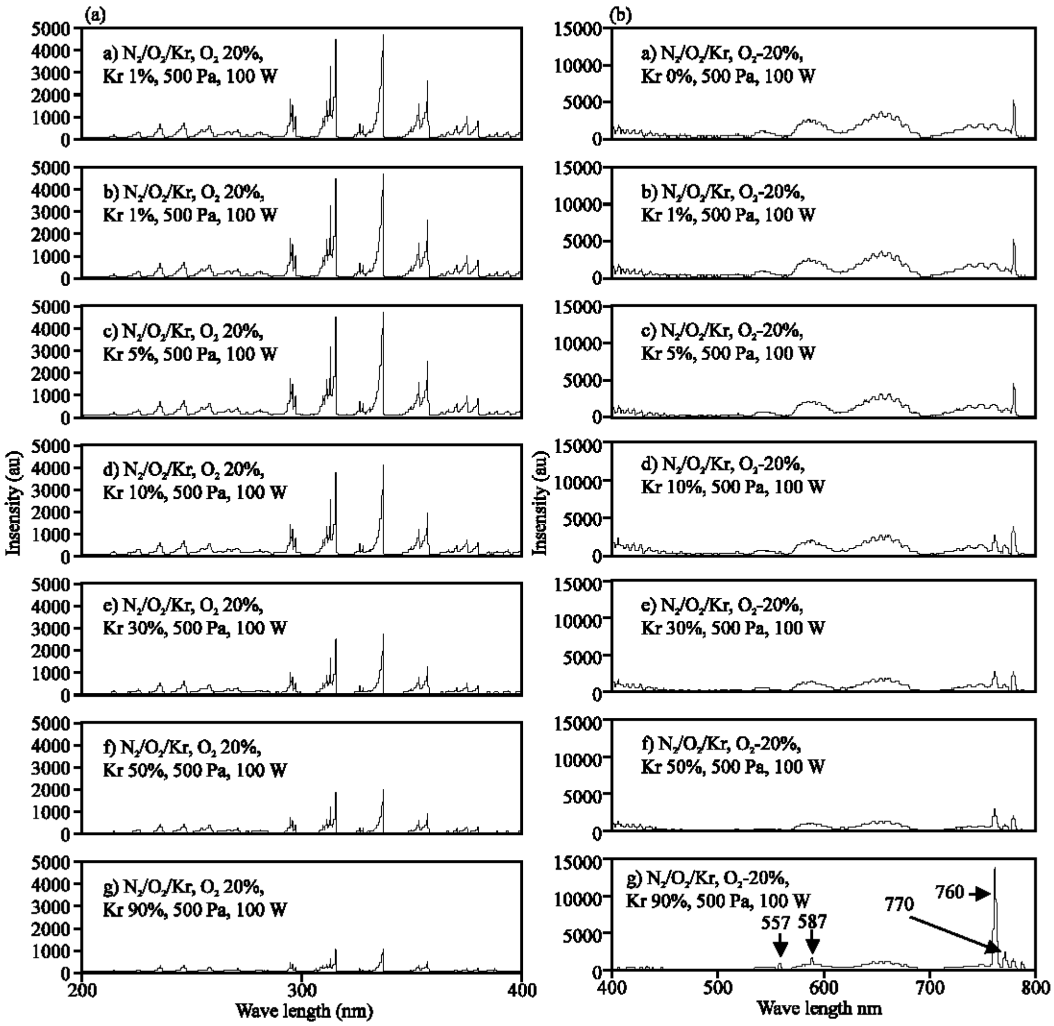

Fig. 3: Emission spectra from $N_2/O_2/Kr$ microwave discharge at various Kr concentration (a) in the UV region and (b) in the visible range

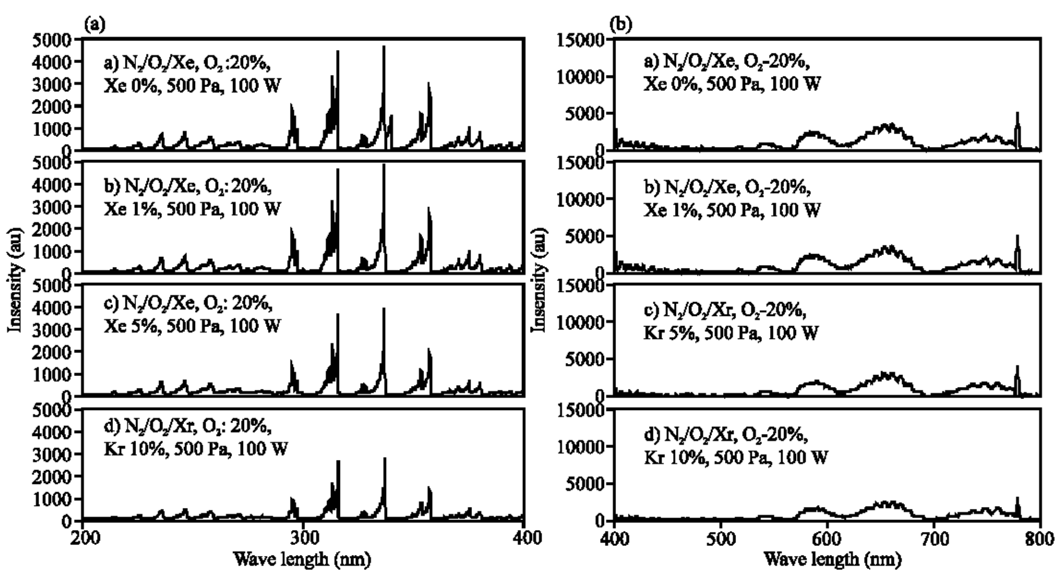

Fig. 4: Continue

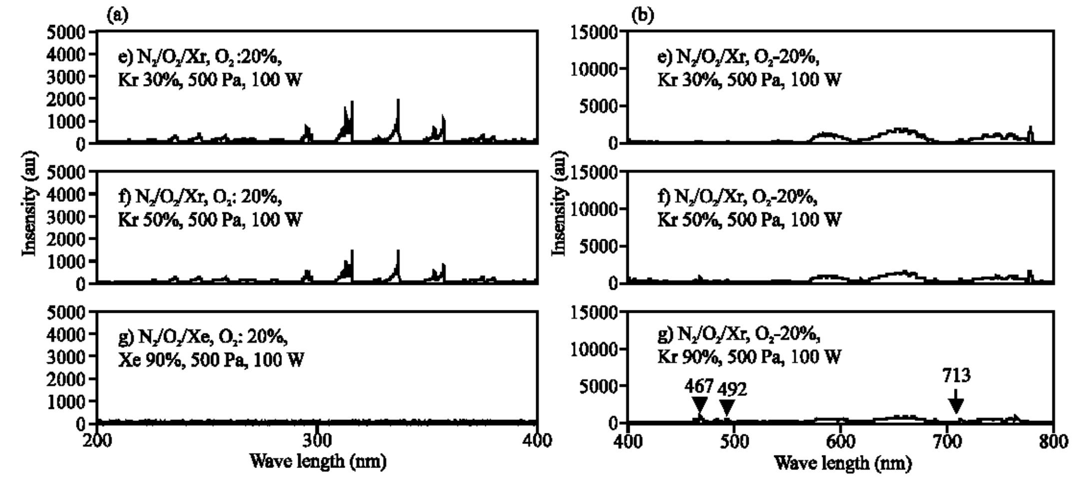

Fig. 4: Emission spectra from $N_2/O_2/Xe$ microwave discharge at different Xe concentration (a) in the UV region and (b) in the visible range

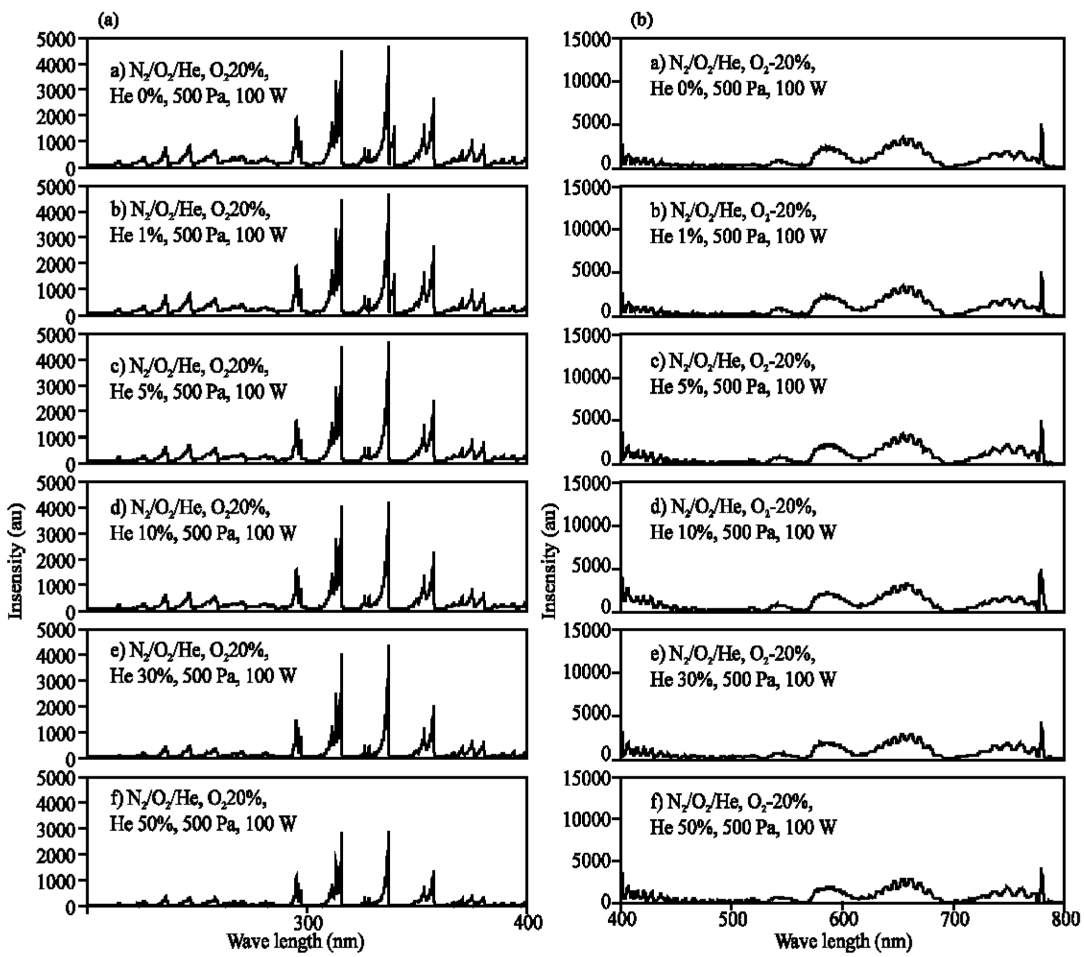

Fig. 5: Emission spectra from $N_2/O_2/He$ microwave discharge at various concentration of He (a) in the UV region and (b) in the visible range

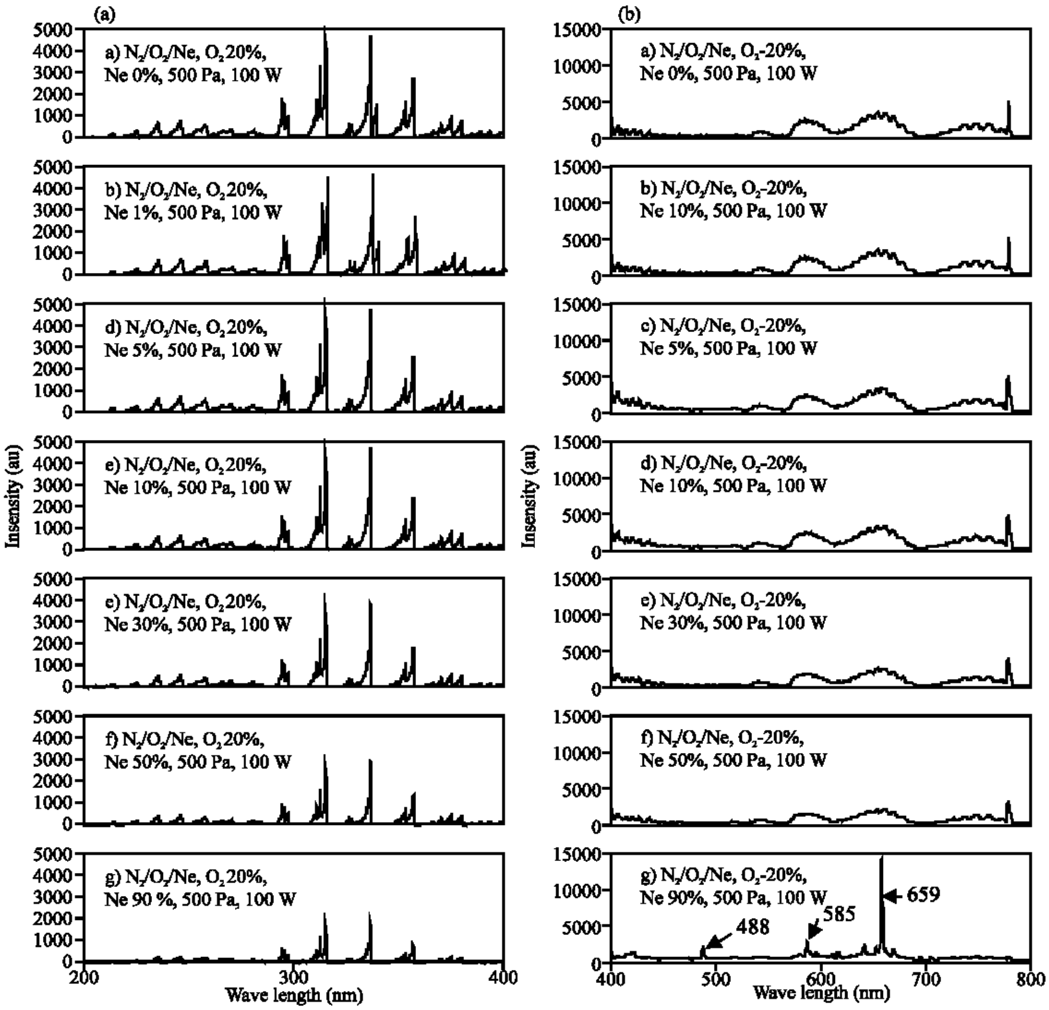

Fig. 6: Emission spectra from $N_2/O_2/Ne$ microwave discharge at various concentration of Ne (a) in the UV region and (b) in the visible range

gases caused collisions with the other co-existing molecules and decreases the emission intensity.

Effect of Ne admixture: Figure 6a shows the effect of Ne admixture on the emission intensity in the UV region and Fig. 6b shows the effect of Ne admixture on the emission intensity in the UV region emitted from N_2/O_2 discharge. It is seen from Fig. 6a, b that admixture of 1% Ne has no effect on the emission intensity in both UV and visible region.

However at 5% and above this concentration, the intensity gradually decreased. From Fig. 6, it is also observed that at 90% Ne and 2% O_2 concentration, Ne peaks appeared at 488, 585 and 659 nm.

CONCLUSION

He, Ne, Ar, Kr and Xe gases at different percentages of concentration was used in the experiment to examine the effect of theses gases on the emission intensity emitted from N_2/O_2 discharge. It was observed that at low concentration <5%, these gases have no effect on the emission intensity whereas the emission intensity in both the UV and visible ranges decreased as the rare gas concentration was increased. The gases behave as buffer gases. Buffer gases cause collisions with the other co-existing molecules and decreases the emission intensity. The other reason for the decrease of intensity is the decrease of N_2 concentration with the increase of rare gas concentration in the mixture.

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