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Research paper

Pressure broadening and narrowing due to oxygen and nitrogen gas mixtures at 1270 nm band: part III

Muhammad Ahmad Al-Jalali *

A former Prof. Dr. at the Department of Physics, Faculty of Science, Taif University, Kingdom of Saudi Arabia
**Corresponding author E-mail: maljalali@gmail.com*

Abstract

Collisional effects on the absorption spectrum of oxygen and nitrogen gas mixtures at 1270 nm band appear as a broadening or narrowing at the component of Lorentzian width in the Voigt profile. The deconvolution method of spectral lines reveals two kinds of bands, the first centered at 1264 nm, and the second centered at 1268 nm. At high pressures, the Gaussian full width may be fixed, and Voigt's full width at half maximum (FWHM) nearly equals to Lorentzian full width. Partial pressures of each gas were between 1-10 bar at 298 K. New results highlight the effect of nitrogen as a collisional partner on the oxygen spectrum.
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Keywords: *Partial Pressure; Voigt FWHM; Lorentzian Width; Gaussian Width.*

1. Introduction

At high pressures, gases may be found as a mixture of monomers, dimers [1-3], and dimols [4-6]. Oxygen and nitrogen mixtures contain free O_2 monomers, free N_2 monomers, O_2-O_2 or N_2-N_2 homodimers/homodimols, and O_2-N_2 heterodimers/heterodimols as dimolecular complexes. Dimers spectrum is usually attributed to Van der Waals dimers under low-temperature conditions [7-8], while the dimols spectrum frequently ascribes to collision-induced absorption (CIA), where the broad spectral maximum characterizes this absorption [9-13]. It is expected that the dimers and dimols will contribute to the spectrum, but at high temperatures and pressures they will compete with each other, and overlapping between the two spectrums may happen with the CIA spectrum's domination [14-18]. The absorption spectrum at 1270 nm band comes only from the oxygen spectrum, because of the nitrogen spectrum outside this region [19]. The role of nitrogen in the mixtures of oxygen and nitrogen is defined as a collisional partner or a collisional mediator to increase the absorbance without spectral interference between oxygen and nitrogen at 1270 nm band [20]. Deconvolution analyses of overlapping spectra at 1270 nm band indicate the existence of two kinds of absorption bands, the first is a discrete absorption band that arises from O_2 monomers, and the second is a continuous absorption band that comes from O_2-O_2 homodimers/homodimols, or O_2-N_2 heterodimers/heterodimols. In this paper, a continuous absorption band which is characterized by O_2 dimol, and O_2-N_2 heterodimols spectrum will be considered a consequence of the CIA by O_2-O_2 or O_2-N_2 in the electrically excited states, because dimers spectrum may disappear at high pressures, or their ratio may be much lower than dimols spectrum, so that, Voigt FWHM was deduced as a whole value of two kinds (dimers/dimols) to avoid big errors. The O_2 monomer ground state $({}^3O_2(X\,{}^3\Sigma_g^-(\upsilon=0), m_s=0,\pm1))$ is a paramagnetic triplet ground state, and the N_2 monomer ground state $({}^1N_2(X\,{}^1\Sigma_g^+(\upsilon=0))$ is a diamagnetic singlet ground state, whereas the ground state of oxygen binary is $({}^3\Sigma_g^-(\upsilon=0)+{}^3\Sigma_g^-(\upsilon=0))$, and the ground state of O_2-N_2 is $(X\,{}^3\Sigma_g^-(\upsilon=0)+X\,{}^1\Sigma_g^-(\upsilon=0))$. The absorption equations of three states may be given as follows [21-25]:

$$\begin{aligned}
&a^1\Delta_g(\upsilon=0) \Leftarrow 1268\,nm + X^3\Sigma_g^-(\upsilon=0)\\
&X^3\Sigma_g^-(\upsilon=0)+a^1\Delta_g(\upsilon=0) \Leftarrow 1264\,nm + X^3\Sigma_g^-(\upsilon=0)+X^3\Sigma_g^-(\upsilon=0)\\
&X^1\Sigma_g^-(\upsilon=0)+a^1\Delta_g(\upsilon=0) \Leftarrow 1264\,nm + X^3\Sigma_g^-(\upsilon=0)+X^1\Sigma_g^-(\upsilon=0)
\end{aligned} \tag{1}$$

$\left(a^1\Delta_g(\upsilon=0)\right)$ is the lowest singlet electronic excitation energy in all the above equations (1). Theoretically, it is supposed that excitation levels have the same absorption wavelength, but experimentally, the peak intensity typically shifts to lower wavelengths for the CIA contribution, because the CIA profile is asymmetric around its origin by a Boltzmann factor. So, monomers and dimols spectral lines appear as two separated transitions and occur at different absorption wavelengths (1268 nm for O_2 monomer and 1264 nm for O_2 dimol). At high pressures, virial theory has to take into account because total pressure has a complicated state, according to the relation (1), which it consists of monomers and dimols partial pressures:

$$p_{tot} = p_{\text{dimols}}(= p_{O2-O2} + p_{O_2-N_2} + p_{N_2-N_2}) + p_{\text{monomers}}(= p_{O2} + p_{N_2}) \tag{2}$$

Where 98% of total pressure comes from dimols, and nearly 2% or less belongs to monomers [45]. It is difficult to separate and calculate partial pressures in these mixtures, so data analysis will depend on the experimental partial pressure of each gas, and at the total pressures for both gases. At low pressures and for pure gas, the Voigt FWHM equation for every spectral line may be given as follows [26]:

$$voigt\ FWHM \cong 0.5346\,\Gamma_L + (0.2166\,\Gamma_L^2 + \Gamma_G^2)^{1/2} \tag{3}$$

Where Γ_L is the Lorentzian width, and Γ_G is the Gaussian width (Doppler broadening full width). The Voigt FWHM of the spectral line depends linearly on the perturber density or pressure either in the case of self-broadening in pure gas or in the case of broadening by foreign gas [27-40]. The effect of foreign gas such as N2 is assumed to be additive, and the Voigt FWHM contribution may be weighed by the mole fractions (χ_i) of gas mixtures, then the total Voigt FWHM may be written according to this equation (Dalton's law):

$$\begin{aligned}&\text{monomers}\, Voigt\ FWHM\ (1268\, nm\, band) = \chi_{O_2} Voigt\Gamma_{O_2} + \chi_{N_2} Voigt\Gamma'_{O_2} \\ &\text{dimols}\ Voigt\ FWHM\ (1264\, nm\, band) = \chi_{O_2\text{-}O_2} Voigt\Gamma_{O_2\text{-}O_2} + \chi_{N_2\text{-}O_2} Voigt\Gamma_{O_2\text{-}N_2}\end{aligned} \tag{4}$$

Where $Voigt\,\Gamma_{O_2-O_2}$ (or $Voigt\,\Gamma_{O_2-N_2}$) denotes to the dimols Voigt FWHM of the oxygen line in pure oxygen dimols (or in the $O_2 - N_2$ heterodimols), and $Voigt\Gamma_{O_2}, Voigt\,\Gamma'_{O_2}$ denotes to the monomers Voigt FWHM of the oxygen line in pure oxygen monomers (or in the O_2, N_2 monomers mixture). $Voigt\Gamma_{O_2-N_2}, Voigt\,\Gamma'_{O_2}$ could be calculated if the pure oxygen term was known by another experiment. The aim of the present work, in addition to our previous works on pressure broadening and narrowing behavior [41-46], is to analyze the experimental spectral lines of O_2+N_2 mixtures at 1270 nm band by an advanced nonlinear curve fitting method, and conclude the FWHM (Voigt, Lorentzian, Gaussian) by changing the partial pressures in both gases at 298 K.

2. Data analysis

Experimental procedures and analysis methods of spectral lines were described in our recent papers on pure oxygen gas [42,45], and this part is devoted to spectra of O_2+N_2 mixtures in the near infrared region (1200-1300 nm) at 298 K. Two types of experiments were performed, the first is when the partial pressure of nitrogen in absorption cell is held constant and the oxygen pressure increases in between 1-10 bar, and the second is when the partial pressure of oxygen is held constant and the nitrogen pressure increases in between 1-10 bar. The constant partial pressures of nitrogen or oxygen gas were changed from 1 bar to 10 bar. Table 1 shows a summary of the values of N_2 partial pressure (bar), O_2 partial pressure (bar), and total pressure of the O_2+ N_2 mixtures that were used in the absorption cell to get about 100 spectral lines.

Table 1: Numbers of Spectral Lines with Partial Pressures and Total Pressures of Gaseous Mixtures of Oxygen and Nitrogen

100 spectral lines	N_2 partial pressure(p_{N2}) / bar									
O_2 partial pressure (p_{O2}) / bar	1	2	3	4	5	6	7	8	9	10
	The total pressure of the N_2-O_2 mixture/bar									
1	2	3	4	5	6	7	8	9	10	11
2	3	4	5	6	7	8	9	10	11	12
3	4	5	6	7	8	9	10	11	12	13
4	5	6	7	8	9	10	11	12	13	14
5	6	7	8	9	10	11	12	13	14	15
6	7	8	9	10	11	12	13	14	15	16
7	8	9	10	11	12	13	14	15	16	17
8	9	10	11	12	13	14	15	16	17	18
9	10	11	12	13	14	15	16	17	18	19
10	11	12	13	14	15	16	17	18	19	20

Figure 1 shows a comparison between some spectral lines from Table 1 and the pure oxygen spectra under various pressures. The spectral lines of O_2-N_2 gas mixtures were treated by using Voigt deconvolution in order to separate the band spectrum of 1264 nm from 1268 nm band envelope [45-46]. Gaussian width was fixed at 298 K according to its famous relation:

$$\Gamma_G(T) = 7.7\times10^{-7}\lambda_c\sqrt{\frac{T}{m}}\quad nm \tag{5}$$

Where m is the atomic mass of the absorber and λ_c is the central wavelength. Figure 2 represents all the steps that are needed to separate the two bands from each other for two experimental spectral lines (as an example for all lines), the first line (Figure 2a) as a result of the gas mixture consists of oxygen partial pressure equals to 10 bar and nitrogen partial pressure equals to 1 bar (p_{N2}=1bar, p_{O2}=10 bar), and the second line (Figure 2 b) was with an oxygen partial pressure equals to 1 bar and nitrogen partial pressure equals to 10 bar (p_{N2}=10 bar, p_{O2}=1bar).

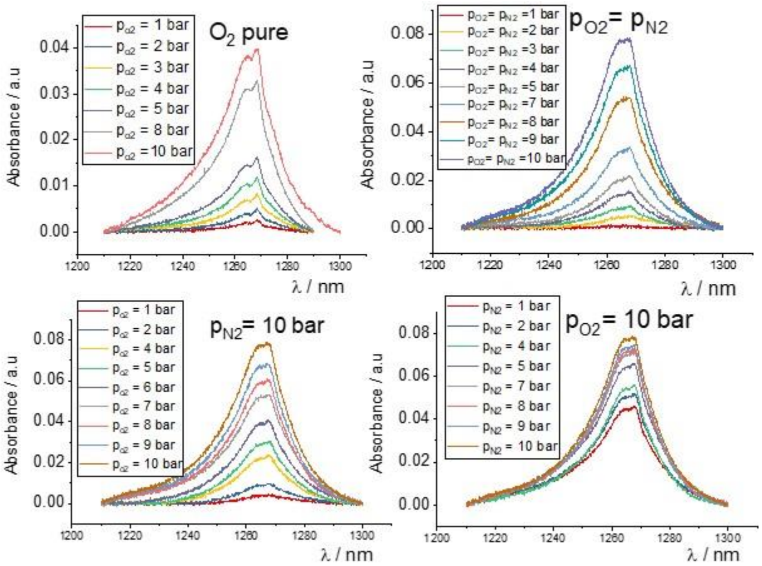
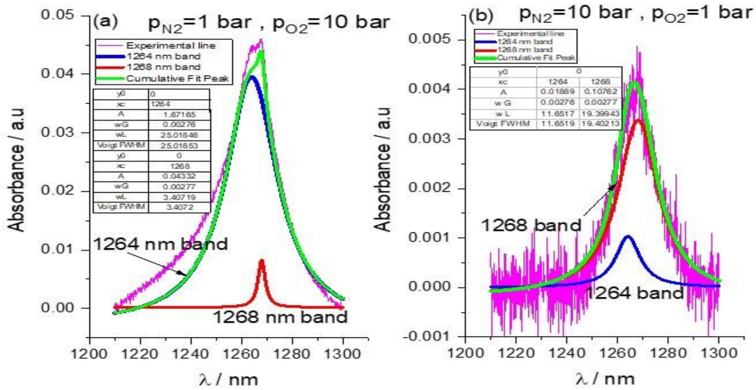


Fig. 1: Represents Some Examples of Raw Absorption Spectral Lines That Were Arbitrarily Taken from Table 1 For Some O_2+N_2 Mixtures, in Addition to the Raw Pure Oxygen Spectrum [45].

The Voigt FWHM, Lorentzian width, fixed Gaussian width, and integrated absorbance for two bands (1264 nm and 1268 nm) were deduced according to the steps in Figure 2. Initial results denote a big difference between Figure 2a and 2b, and FWHM could be controlled by changing nitrogen partial pressures or oxygen partial pressures. Voigt FWHM values with a fixed Gaussian width were nearly equal to Lorentzian width values for each line (Figure 2), where according to equation (5), Γ_G = 0.00276 nm for 1264 nm band, and Γ_G =0. 00277 nm for 1268 nm band at 298 K.



y0	0
xc	1264
A	1.67165
wG	0.00276
wL	25.01846
Voigt FWHM	25.01853
y0	0
xc	1268
A	0.04332
wG	0.00277
wL	3.40719
Voigt FWHM	3.4072

y0	0	
xc	1264	1268
A	0.01889	0.10762
w G	0.00276	0.00277
w L	11.6517	19.39943
Voigt FWHM	11.6519	19.40213

Fig. 2: Deconvolution Method of Spectral Line and Comparison between Two Spectral Lines (As an Example), A) P_{n2}=1bar and P_{o2}=10 Bar. B) P_{n2}=10 Bar, and P_{o2}=1 Bar. Figures 2a and 2b Show the Procedures to Separate the O_2 Monomer (Red Line) from O_2 Binary (Blue Line) Absorbance Profile in the 1270 Nm Region, Green Lines Represent the Total Voigt Profile that Represents A Convolution of 1264 Nm Band, and 1268 Nm Band, Blue Lines Represent Asymmetric Voigt Profile of 1264 Nm Band Envelope, Red Lines Represent Voigt Profile of 1268 Nm Band Envelope and Magenta Lines Represent the Experimental Lines. Symbols Inside Figures Are Offset(Y_0), Central Wavelength (Λ_c=X_c), the Area Under the Spectral Line Which Nearly Equals the Integrated Absorbance Intensity(A), Gaussian Width (WG=Γ_G), Lorentzian Width (WL=Γ_L), and Voigt FWHM.

3. Results and discussion

In this study, it was found that the values of Voigt FWHM are nearly equal to the Lorentzian width of all spectral lines, and collisional effects will be responsible for Lorentzian width. A Lorentzian component of the Voigt profile, Gaussian width with a fixed component, and the integrated absorbance of two kinds of bands (1264 nm band and 1268 nm band) were well determined using the steps in Figure 2 for all spectral lines in Table 1. Three types of analysis were studied, and illustrated in the three following paragraphs:

3.1. Lorentzian component when nitrogen pressure is constant

The partial pressure of nitrogen is held constant and the oxygen partial pressure gradually increases from 1-10 bar. The two kinds of Lorentzian widths are plotted against oxygen partial pressure in Figure 3 (nitrogen pressures are from 1- 4 bar) and Figure 4 (nitrogen pressures are from 5-10 bar), in addition to pure oxygen Lorentzian widths (Figure 3a) for comparison at 298 K. Figures 3 and 4 (the blue points) represent the first kind of Lorentzian widths, which belongs to 1264 nm band envelopes, and nearly vary linearly with an oxygen partial pressure, where a pressure broadening coefficient (slope of the line) characterizes this behavior. The Dicke effect (narrowing in Lorentzian width) disappears in the first kind because of high pressures, or in practice, it may be masked by strong collisional broadening. Also, in the same Figures 3 and 4 (The red points) the second kind of Lorentzian widths, which correlate to 1268 nm band envelopes show a non-linear decay of widths as a function of oxygen partial pressure. It seems that the Lorentzian widths (1268 nm band) in gas mixtures are twice higher than that in pure oxygen, and going to decay at high pressures, where deviation from Dicke effect is very clear. Collision narrowing at 1268 nm band does not totally belong to Dicke effect, perhaps this is due to other factors like a competition between two bands. Figure 3 shows no significant differences between Lorentzian widths in pure oxygen and in its mixtures with nitrogen at 1264 nm band. However, Figure 4 shows two different behaviors between 1264 nm band and 1268 nm band, where the widths belonging to 1268 nm band become larger than the widths belonging to 1264 nm band at lower pressures of oxygen.

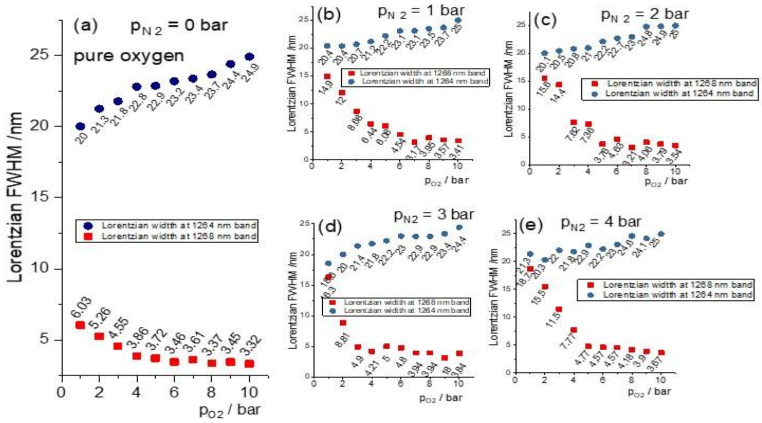


Fig. 3: Lorentzian Widths as A Function of Oxygen Partial Pressure with A Constant Pressure of Nitrogen (1-4 Bar). Figure 3a Belongs to Pure Oxygen for Comparison [45].

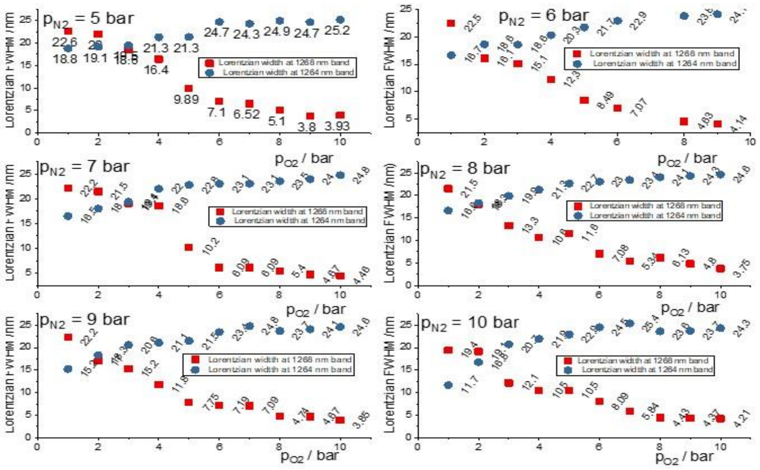


Fig. 4: Lorentzian Widths as A Function of Oxygen Partial Pressure with A Constant Pressure of Nitrogen (5-10 Bar) for 1264 Nm Band and 1268 Nm Band. It Seems the Competition between Two Bands at Lower Pressures of Oxygen Is Very Clear.

3.2. Lorentzian component when oxygen pressure is constant

The partial pressure of oxygen is held constant and the nitrogen pressure increases from 1 -10 bar. Figure 5 (constant pressures of oxygen are from 1 to 4 bar) and Figure 6 (constant pressures of oxygen are from 5 to 10 bar) show the behavior of two kinds of Lorentzian widths as a function of nitrogen partial pressures. Figure 5 shows both bands subject to broadening and narrowing in their widths, and it appears that Lorentzian widths of 1264 nm band in Figures 5a and 5b decrease, but tend to be constant in Figures 5c and 5d, whereas Lorentzian widths of 1268 nm band increase until a partial pressure of nitrogen reaches to 5 bar and after then decrease to the lowest values. Figure 5 breaks the rule which claims pressure broadening only belongs to 1264 nm band, where Figures 5a and 5b are also shown that pressure narrowing may come from the 1264 nm band, and Dicke effect may compete with pressure broadening at this band. Figure 6 shows Lorentzian widths of 1264 nm band are more stable with constant values, whereas Lorentzian widths of 1268 nm band increase linearly with the partial pressures of nitrogen, but tend to be constant at oxygen partial pressures more than 8 bar. From Figure 6, it seems that the nitrogen gas has a weak effect on both two bands at partial pressures of oxygen more than 5 bar. From Figures 5 and 6, It seems that pressure broadening and pressure narrowing do not obey the same rule as in the first type.

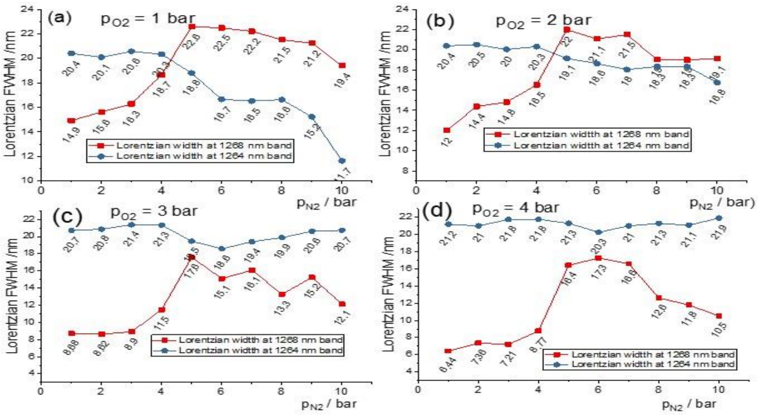


Fig. 5: Lorentzian Widths as A Function of Nitrogen Partial Pressure with Constant Pressure of Oxygen (P_{o2}=1,2,3,4 Bar).

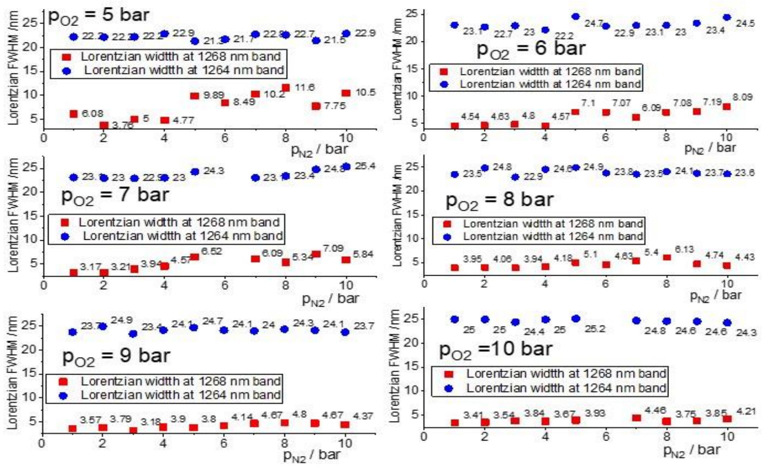


Fig. 6: Lorentzian Widths as A Function of Nitrogen Partial Pressure with Constant Pressure of Oxygen (P_{o2}=5,6,7,8,9,10 Bar).

3.3. Integrated absorbance intensity

Figure 7 shows a strong relationship between the integrated absorbance intensity of envelopes (area under the spectral line) and pressure. The values of the area under the two kinds of envelopes as a function of O_2 partial pressure were deduced from data analysis as in Figure 2 at 298 K. Figure 7a shows a quadratic relation between integrated absorbance intensity at 1264 nm band and a partial pressure of oxygen, where this dependency is a distinctive point to CIA behavior. Figure 7b shows a linear relation between integrated absorbance intensity at 1268 nm band with the partial pressure of oxygen. It seems from Figure 7 that integrated absorbance increases with the increasing of nitrogen partial pressures.

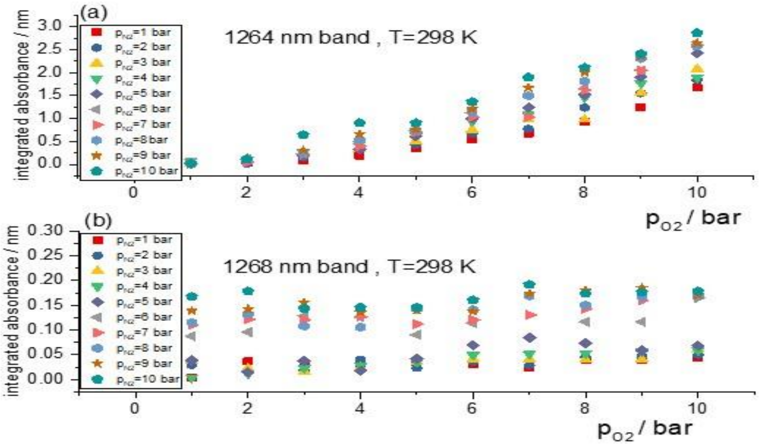


Fig. 7: The Relation between Integrated Absorbance(A) for 1264 Nm Band and 1268 Nm Band as A Function of Oxygen Partial Pressure.

4. Conclusion

Nitrogen plays an important role as a collisional partner and a sensor controller by increasing the absorbance in the oxygen and nitrogen mixtures, and its effect appears at 1268 nm band more than that in the 1264 nm band. Pressure broadening has sovereignty at the 1264 nm band, where the CIA contribution is a characteristic and an attractive point of this band, and pressure narrowing has dominion at the 1268 nm band. In addition, pressure broadening and narrowing may appear in both 1264 nm band and 1268 nm band. Integrated absorbance intensity as a function of oxygen partial pressure has the same behavior as in pure oxygen, but the role of nitrogen is to increase the area under the spectral line. The integrated absorbance intensity could be used to discriminate between dimols and monomer behavior.

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