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Sulfur Hexafluoride (SF6) Trace Gas Sensing using Modulated CO_2 Laser Beam

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Abstract: Sulfur hexafluoride gas (SF6) is a very important in high voltage equipment as a spark arrestor medium for switch gear assemblies. Laser-based Photo-Acoustic (PA) technique has been used to monitor the traces of SF6 leaks in the surrounding atmosphere. This technique is based on inducing acoustic waves in the gas which is stimulated by a modulated (CW) CO_2 laser beam of 10.6µm wavelength, 1.0 to 2.5W power and power modulation frequency of 1365 to 10920Hz (1^{st} to 8^{th} harmonics). A cylindrical acoustic resonator of 50mm length and 4.5mm diameter is used, with a miniature microphone mounted at its midpoint to detect the sound waves. A sound monitoring computer software (*Sound Card Oscilloscope V4.1*) has been used to capture and record the sound waves and their frequency spectrums. Experimental results revealed that significant acoustic signal can be detected even at trace concentration, this is attributed to the high absorption of the CO_2 laser radiation by SF6 gas. Also, it conforms very well to the laser modulation frequency applied to the resonator, with a tendency to oscillate at higher harmonics when higher concentrations of SF6 exist within the gas mixture. The strongest line of oscillation has been statistically found to be the 4^{th} harmonic (5460Hz).

Keywords: *Laser photoacoustic spectroscopy, gas monitoring, Sulfur hexafluoride.*

Introduction

The photoacoustic effect has been discovered by Alexander Graham Bell. This effect is featured by the generation of sound from the interaction of light and matter [1]. Photo-Acoustic Spectroscopy (PAS) has been used in a diversity of applications in the fields of atmospheric researches, combustion processes, medical diagnostic, food industry, pollution monitoring, and trace gases sensing. Reliable trace gases sensors has acquired increased importance and interest in the recent years. Various fields of applications require a precise monitoring of a large variety of species in different gas mixtures and at various concentration levels ranging from parts per billion (ppb) to several hundreds of parts per million (ppm) [2].

Extremely high purity gas mixtures are usually utilized in Industrial processes, where the level of impurities and contaminants must be precisely monitored, controlled and kept at trace levels. Ammonia, acids such as (HF, HCl, HBr) in semiconductor clean room applications, moisture contamination in the manufacturing of electronics or optical devices such as laser diodes, and also other types of impurities need to be monitored. PAS offers a viable means for monitoring of such gases [2]. SF6 has been used by Nike in sport shoes to minimize leaks from the air cushion in the heel because of its large molecule, and also used as a tracer for underground water leaks and burst water pipes [3]. Laser-based spectroscopic technology is one of the attractive methods used for trace gas monitoring, this technology exhibits both high sensitivity and selectivity during detection [4].

In 2002, a photoacoustic spectrometer based detection of SF6 leaks in high power insulated switchgear systems has been developed by **M. A. Gondal** et. al. [5], where a CO_2 laser tuned to operate at 10.55µm wavelength, and a mechanical chopper to modulate the laser beam, with an acoustic resonator, and a sensitive microphone tailored with a Lock-In Amplifier are used. The optimum modulation frequency

for trace SF6 in nitrogen has been found experimentally to be 5200Hz. In 2009, **X. Yuanzhe** et. al. [6] designed a photoacoustic spectrometry based experimental system for detecting SF6 gas concentration. The photoacoustic technique was used to survey the photoacoustic characteristic of SF6 molecule at 10.6μm wavelength. The detection sensitivity is 0.12×10^{-9} under standard atmospheric pressure.
This work is aimed to presents an experimental approach to analyze the response of SF6 gas using standard miniature microphone and sound recording software. A range of modulation frequencies of the CO_2 laser beam at different SF6 concentrations in ambient air have been experimented to feed the analyses.

Fundamentals of laser photoacoustic spectroscopy

The process of generation and detection of the PA signal in a gaseous sample can be divided into four steps, these are [4]:
Molecular absorption of photons which leads to the excitation of the absorbing molecules to a higher energy level.
Non-radiative relaxation of the excited state (collisional relaxation). The optical energy previously absorbed is totally released as heat in the sample which leads to an induction of gas expansion, i.e., a local modification of its pressure.
When the laser of intensity I, is modulated at a frequency $= \frac{\omega}{2\pi}$, so it is expressed as:

$$I = I_o e^{i\omega t} \quad (1)$$

Both thermal and acoustic waves are generated in the sample as a result of the production of a periodic heating of the sample, which leads to periodic pressure variations.
Detection of the acoustic wave using a microphone provided that the modulation frequency f falls within the audio range [7]. The steps of PA effect in gases is shown in Figure 1.

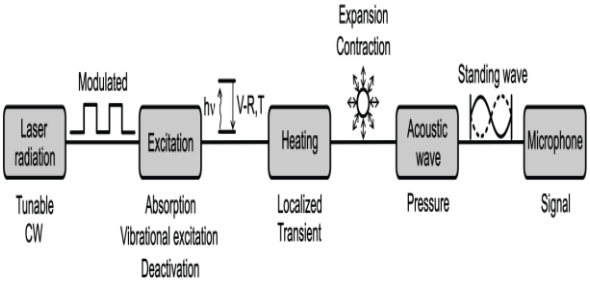


Fig. 1: Schematic of the physical processes occurring during optical excitation of molecules in photoacoustic spectroscopy [8]

Experimental procedure

The experimental arrangement which has been used to carry out the detection of photoacoustic waves generated by SF6 gas is shown schematically in Figure 2, while the real set-up is shown in Figure 3. It consists of a modulated CO_2 laser, a PA cell, SF6 gas supply and a personal computer. The CO_2 laser (10.6μm wavelength) and its power controller has been used with a signal generator which feeds the controller with the modulation frequency and the pulse shape. The PA cell is made of Brass, it consists of a cylindrical resonator 50mm long and 4.5mm diameter with two end buffers for gas entry and exit. A ZnSe window (to pass the modulated laser beam) and a rear mirror made of Brass are used to close the ends of the PA cell.
A miniature microphone is fitted in the middle of the resonator wall to pick up the acoustic waves. SF6 gas is supplied from a canister with the necessary connections and vales. The acoustic waves are recorded by a sound monitoring computer software (*Sound Card Oscilloscope V1.4*) installed on the personal computer.

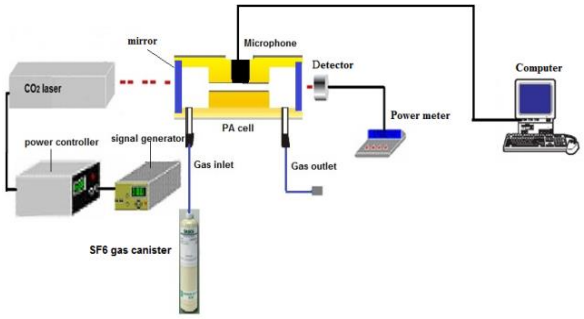


Fig. 2: Experimental layout schematic

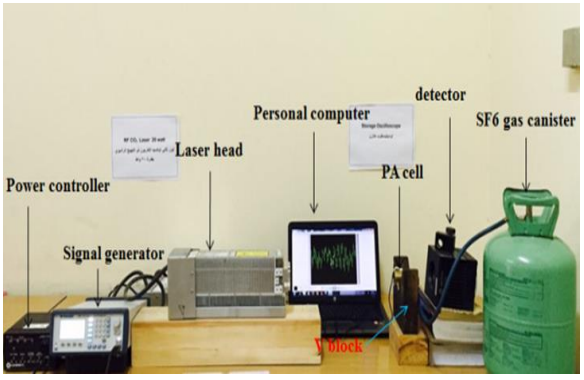


Fig. 3: Photograph of the experimental arrangement

The first signal is captured while SF6 gas is flowing through the bore of the resonator (i.e., both inlet and outlet gas valves are open), this is

to ensure that the injected gas is actually filling the whole resonator, i.e. 100% gas concentration. The inlet valve is then closed to enable gas exchange with atmospheric air, the gas concentration is reduced gradually with time, and acoustic signals are recorded after 1, 5, 10, 20, 40, 80 and 100 minutes from closing the inlet valve. This method has been followed because no standard premixed gas samples of known concentration are available at hands, therefore these waiting times shall denote less concentration of SH6, where the 100 minutes shall indicate a trace level of concentration.

Both time-domain waves and line spectrum are recorded at laser modulation frequencies which correspond to the fundamental and the higher harmonics of the acoustic waves which are generated by the SF6, and for the full gas concentration and the lower concentrations that occur due to the waiting times listed above.

The resonance frequency of SF6 has been calculated for the present resonator to be 1365Hz and that of atmospheric air is 3500Hz using Eq.(1) [9], and using SF6 & air properties, respectively, at normal conditions:

$$f_{kmn} = \frac{v_s}{2}\left[\left(\frac{k}{L}\right)^2 + \left(\frac{\propto_{mn}}{\pi r}\right)^2\right]^{1/2} \qquad (1)$$

where v_s is sound velocity in the sample gas, L and r are the length and radius of the resonator; k, m and n indices (non-negative integers) refer to the values of the longitudinal, azimuthal, and radial modes, respectively, and α_{mn} is the nth root of the derivative of the m_{th} Bessel function. The sound velocity is calculated using Eq.(2) [1, 10]:

$$v_s = \left(\frac{\gamma RT}{M}\right)^2 \qquad (2)$$

where γ refers to the ratio of the specific heat of the gas at constant pressure Cp to that at constant volume Cv, R is the ideal gas constant, T is the absolute temperature and M is the gas molecular weight.

Experimental Results

Acoustic signals and their corresponding spectrums have been recorded and analyzed for the range of laser modulation frequencies 1365 to 10920Hz. Because of the large sum of the collected data, only representative samples of results have been selected to be shown in this section which present the trends of variations of acoustical response of SF6 when its concentration is reduced in the gas mixture. Therefore, results at modulation frequencies of 1365 and 5460Hz and at selected delay times (after closing the gas inlet valve to the cell) will be presented hereafter, and as follows:

Results at 1365Hz modulation frequency

The acoustic signals and their frequency spectrums at the resonance frequency of the monitoring gas and full gas concentrations are presented in Figure 4. From Figure 4(a) it is clear that many harmonics have been generated when the resonator is filled with SF6 gas, some of them as peaks in amplitude. From Figure 4(b), it is clear that the frequency spectrum contains signal peaks at 1365Hz, 4100Hz, 8200Hz, 10900Hz and 13100Hz. The strongest peak has been recognized at 10900Hz i.e. the 8th harmonic frequency. Higher harmonics tend to be excited more than lower ones due to the strong absorption of laser radiation which is converted to more energetic acoustic frequencies, i.e., higher harmonics.

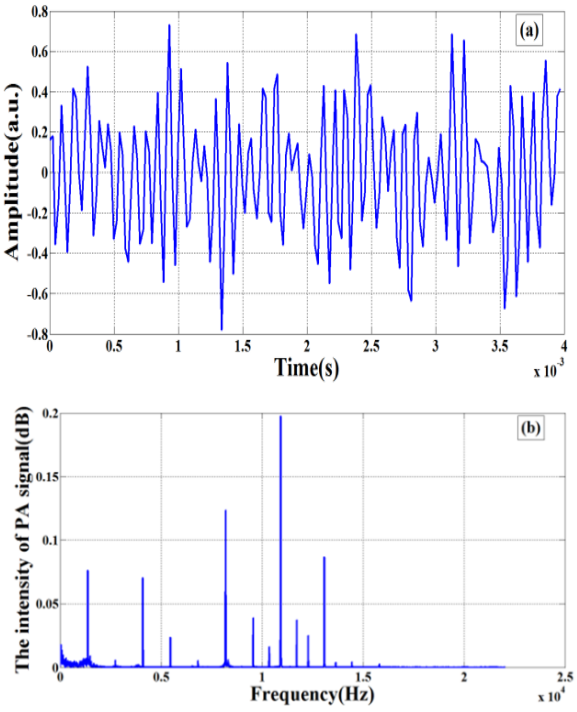


Fig. 4: Experimental results for SF6 at 1365Hz at full gas concentration, (a) PA signal amplitude (b) frequency spectrum.

Figure 5(a) presents the audio signal after 40 minutes, it reveals that fewer harmonics have aroused compared with the former case of full SF6 concentration. Figure 5(b) depicts the frequency spectrum of the recorded signal, the strongest peak is located at 1365Hz.

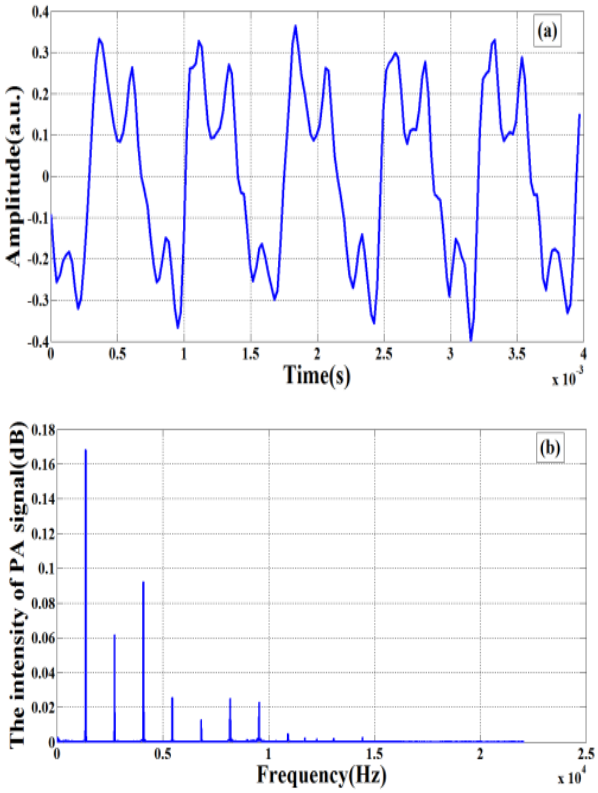
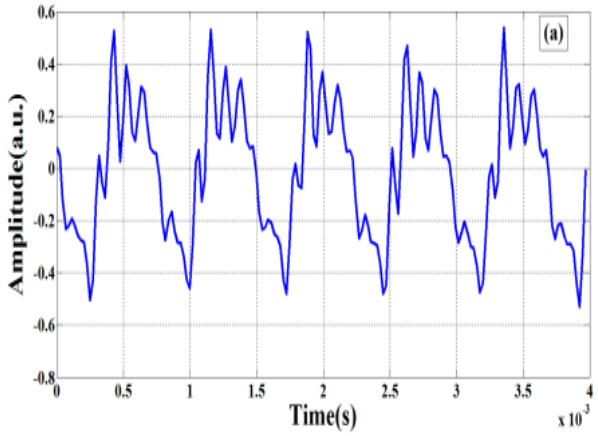


Fig. 5: Experimental results for SF6 at 1365Hz 40 minutes after closing the inlet valve (a) PA signal amplitude (b) frequency spectrum

From Figure 6(a) and (b) it is recognized that after 99 minutes from closing the inlet valve, the strongest peak has been detected at the resonance frequency of the monitoring gas, i.e. at 1365Hz. Higher harmonics have still been observed but with reduced line strength in contrast to that noticed at the previous states. An audible sound of acoustic wave can be heard even at this very low SF6 concentration, which indicates quite substantial energy coupling between laser photons and SF6 molecules and consequently conversion into acoustical oscillations.



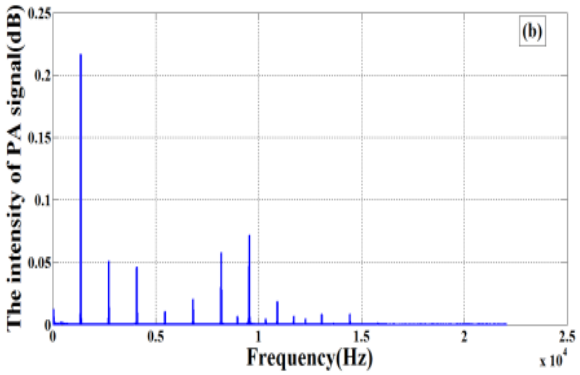


Fig. 6: Experimental results for SF6 at 1365Hz 99 minutes after closing the inlet valve (a) PA signal amplitude (b) frequency spectrum.

Results at 5460Hz modulation frequency

The behavior of the acoustic wave and its frequency spectrum have been monitored at the 4th harmonic frequency. Figure 7(a) illustrates the time domain detected signal at full gas concentration, the frequency of peak amplitude is at 10920Hz. Figure 7(b) shows the frequency spectrum of the recorded signal, strongest peak was at 10920Hz.

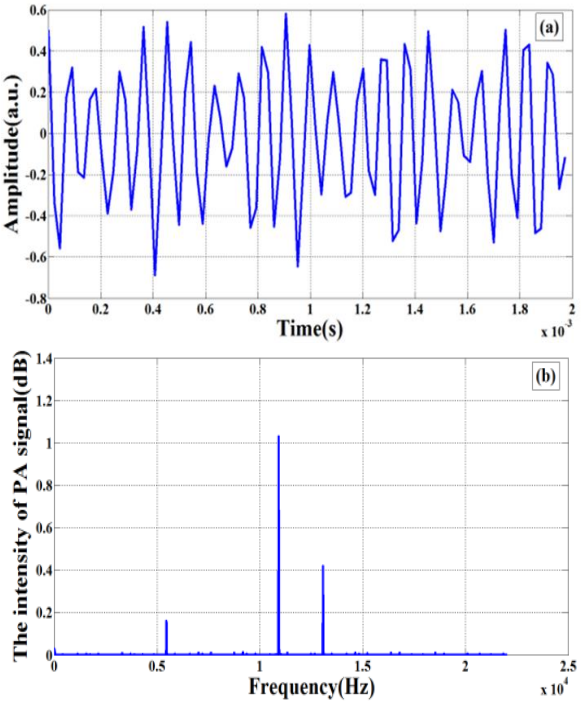


Fig. 7: Experimental results for SF6 at 5460Hz at full gas concentration (a) PA signal amplitude (b) frequency spectrum.

Figure 8 illustrates the behavior of the detected gas after 1 minute. The detected acoustic signal has been presented in Figure 8(a). Almost regular sinusoidal signal has been detected, this

is due to energy conversion between SF6 and atmospheric gas molecules (i.e. nitrogen and oxygen). The spectrum of this signal have been showed in Figure 8(b). There are two peaks, these peaks are located at 5460Hz and 10920Hz, where the strongest one is at 5460 Hz.

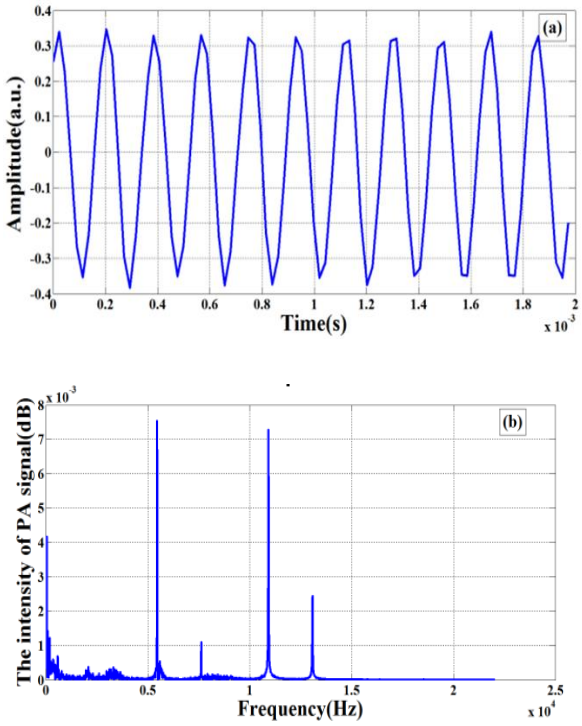


Fig. 8: Experimental results for SF6 at 5460Hz 1 minute after closing the inlet valve (a) PA signal amplitude (b) frequency spectrum.

Discussion

The photoacoustic behavior of SF6 gas has been tested for the range of exiting modulation frequencies of 1365 to 10920Hz, for the full concentration and at each of the times and the modulation frequencies, which means a large sum of data. To summarize these experimental results into concise formats for better understanding, two sets of graphs have been extracted which show the actual data points and the fitted curves.

Figures 8 and 9represents the line intensity of each harmonic at a specific modulation frequency and a specific waiting time (or full concentration). While the graphs of the second set are the concentrates of the first set, as each graph depicts the average of the line spectrum intensity values of each harmonic for all of the modulation frequencies for the full SF6 gas concentration and for the subsequent waiting times, as in sample Figures 10 and 11. Figure 8 is a representative sample that depicts line intensities of the fundamental and higher harmonic at 1365Hz modulation frequency and full SF6 concentration. This graph indicates two significant lines only, namely the 1st (1365Hz) and the 3rd (4095 Hz) harmonics. Figure 9, on the other hand, reveals the results at 5460Hz modulation frequency and full SF6 concentration, where two significant lines have been emerged, the 2nd (2730Hz) and the 4th (5460Hz) harmonics. Figure 9 findings can be viewed from another point, that is the significant lines are the modulation frequency (5460Hz) and its sub-harmonic (half harmonic 2730Hz). Both Figures indicate that the maximum line intensity lies in the modulation frequency lines, i.e. SF6 modes of oscillation conforms quite well to the modulation frequency applied by the exciting laser source. This procedure has been repeated for all of the modulation frequencies for collecting data which are to be used in the next step of calculating average values for the second data set.

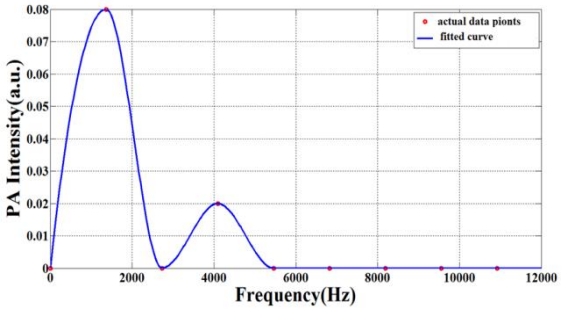


Fig. 8: PA intensity at 100% SF6 concentration and modulation frequency of 1365Hz

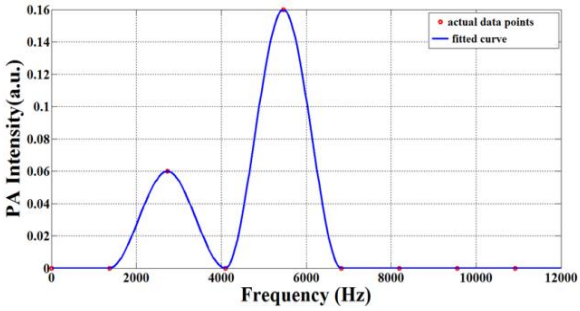


Fig. 9: PA intensity at 100% SF6 concentration and modulation frequency of 5460Hz

Figure 10 presents a comparison for the modulation frequencies 1365 to 10920Hz at full SF6 gas concentration. Each actual data point on the graph depicts the average of the line spectrum intensity values corresponding to a specific harmonic from all of the modulation

frequencies for the full SF6 gas concentration. This procedure has been repeated for the subsequent waiting times those stand for less SF6 concentration up to the trace level at 99 minutes. Results of the 99 minutes have been drafted in Figure 11. Figures 10 and 11 represent the two extremes concentrations of SF6 in atmospheric gas, i.e. the full and the trace concentrations. It can be clearly concluded from these two Figures that the strongest line of oscillation in both cases is 5460Hz (the 4th harmonic). This behavior has been noticed in the other waiting times, particularly longer waiting times, with few exceptions. This statistical result is very close to the direct experimental result found by Gondal et. al. [5], as they determined the optimum laser modulation frequency to be 5200Hz for the strongest acoustical signal.

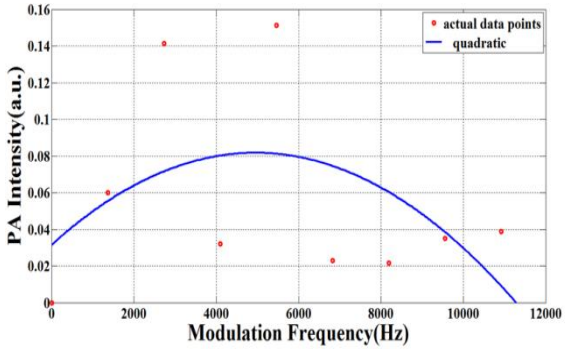


Fig. 10: PA intensity at 100% SF6 concentration for the resonance frequency and higher harmonics

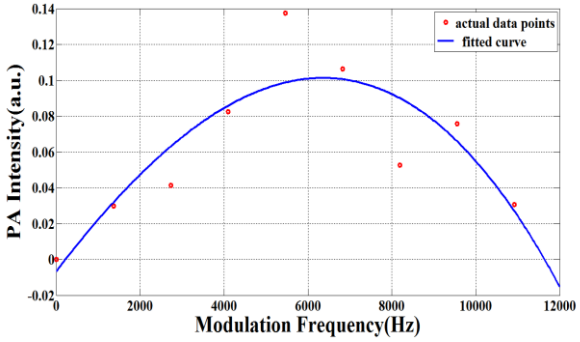


Fig. 11: PA intensity for the resonance frequency and higher harmonic frequencies after 99 minutes

Conclusions

The experimental and the statistical results have led to the following conclusions:

1. SF6 gas has a very good power to absorb the CO_2 laser radiation because of its large absorption cross section compared with other gases, and conforms well to the applied frequency. The prominent oscillating line has been determined by statistical analysis to be 5460Hz, which is the 4th harmonic corresponding the acoustic cell design employed in this work.
2. The waveform periodic patterns are resulting from the interference between the acoustic waves of the fundamental frequency and the higher harmonics. Consequently, the peaks on the line spectrums are being generated due to these interference patterns.
3. Significant acoustical signals have constantly been detected even at trace SF6 concentration level accompanied with audible sound. This would enforce the possibility of using simple, low cost arrangement to monitor this type of gas by using commercially available miniature microphone and computer software to record acoustic signals and frequency spectrums. This is in contrast to the complicated lock-in amplifier and special microphone used in other works.

References

[1] D.C. Dumitras, "CO2 Laser optimisation and application", In Tech, 2012.

[2] J.P. Besson, S.Schilt, L. Thévenaz, "Multi-gas sensing based on photoacoustic spectroscopy using tunable laser diodes", SPECTROCHIMICA ACTA PART A, vol.60, pp.3449–3456, 2004.

[3] P. H. Pedersen, "Ways of reducing consumption and emission of potent greenhouse gases (HFCs, PFCs and SF6)", Nordic Council of Ministers, 2000.

[4] S.Schilt , L.Thévenaz, M. Niklès , L. Emmenegger, C. Hüglin, "Ammonia monitoring at trace level using photoacoustic spectroscopy in industrial and environmental applications", Spectrochimica Acta Part A, vol.60, pp. 3259–3268, 2004.

[5] M. A. Gondal, M. A. Baig, M. H. Shwehdi, "Laser Sensor for Detection of SF6 Leaks in High Power Insulated Switchgear Systems", IEEE Transactions on Dielectrics and Electrical Insulation, Vol. 9, No.3, 2002.

[6] X. Yuanzhe, Z. Hongjing, Li Bo, Zhao Xin, "study on Detection of SF6 Gas Leakage Based on Photoacoustic Spectrometry", International Conference on Measuring Technology and Mechatronics Automation, pp.261-265, 2009.

[7] Y-h. Pao," Optoacoustic Spectroscopy and Detection", Academic Press, 1977.

[8] J. C. Lindon, G. E. Tranter, J. L. Holmes," Encyclopedia of Spectroscopy and Spectrometry", Academic Press, 2010.
[9] A. Tavakoli, M. Tavakoli, M. Taheri, H. Saghafifar, " Design, simulation and structural optimization of a longitudinal acoustic resonator for trace gas detection using laser photoacoustic spectroscopy (LPAS)", Optics & Laser Technology, Vol.42, No.5, pp.828-838, 2010.
[10] C. Kelly, J. Leis and D. Buttsworth, "Development of a Photo-Acoustic Trace Gas Sensor", Conference Proceedings of ACOUSTICS, No. 139, Australia, 2-4 November 2011
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الكشف عن اثار غاز سادس فلوريد الكبريت باستعمال حزمة ليزر ثنائي اوكسيد الكاربون المضمنه

محمد عبد الامير حسين سارة سامي احمد

قسم هندسة الليزر والألكترونيات البصرية ، جامعة النهرين ، العراق ، بغداد

الخلاصة: لأهمية غاز سادس فلوريد الكبريت في معدات الجهد الكهربائي العالي كوسط مانع للشرارة الكهربائية في مفاتيح التبديل، فقد تم إستخدام تقنية مطيافية الليزر الضوئية–الصوتية للكشف عن اثار هذا الغاز في الغلاف الجوي. تستند هذه التقنية على توليد موجات صوتية في الغاز، حيث تحفز الموجات بوساطة حزمة مضمنة من ليزر ثنائي اوكسيد الكاربون المستمر بطول موجي 10,6مايكرومتر، و بمستوى طاقة فعالة من 1,0 الى 2,5 واط، وبمدى تردد لتضمين طاقة الليزر من 1365 الى 10920هرتز (التوافقيات الأولى الى الثامنة). تم إستخدام مرنان صوتي أسطواني بطول 50ملم وقطر 4,5ملم، و تم التقاط الموجات الصوتية باستخدام مايكرفون صغير تم وضعه في وسط المرنان. وقد استخدم البرنامج الحاسوبي (*Sound Card Oscilloscope V4.1*) لضبط وتسجيل الموجات الصوتية و خطوط الأطياف التابعة لها. بينت النتائج التجريبية إمكانية التقاط إشارات صوتية جيدة حتى في التراكيز الضئيلة للغاز. ويعزى ذلك لإمتلاك غاز سادس فلوريد الكبريت قدرة امتصاص عالية جدا لإشعاعات ليزر ثنائي اوكسيد الكاربون. كما اثبتت ان هذا الغاز يتناسق بشكل ممتاز مع الترددات المضمنة لحزمة اليزر التي تمر عبر المرنان، مع ميله للاهتزاز عند التوافقيات العليا عند توافر تراكيز عالية منه ضمن مزيج الغازات. ولقد وجد إحصائياً بأن أقوى تردد للإهتزاز هو عند التوافقية الرابعة (5460 هرتز).