



Highly sensitive photoacoustic multicomponent gas sensor for SF₆ decomposition online monitoring

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Abstract: A ppb-level photoacoustic multicomponent gas sensor system for sulfur hexafluoride (SF₆) decomposition detection was developed by the use of two near-infrared (NIR) diode lasers and an ultraviolet (UV) solid-state laser. A telecommunication fiber amplifier module was used to boost up the excitation optical power from the two NIR lasers. A dual-channel high-*Q* photoacoustic cell (PAC) was designed for the simultaneous detection of CO, H₂S, and SO₂ in SF₆ buffer gas by means of a time division multiplexing (TDM) method. Feasibility and performance of the multicomponent sensor was evaluated, resulting in minimum detection limits of 435 ppbv, 89 ppbv, and 115 ppbv for CO, H₂S, and SO₂ detection at atmospheric pressure.

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1. Introduction

Sulfur hexafluoride (SF₆) was widely chosen as the preferred dielectric gas for electrical insulation or interruption purposes in electric power systems, such as in the gas-insulated switchgear, gas-insulated circuit breaker, gas-insulated transformer, transmission line and substation, due to high dielectric strength, chemically inactive and environmentally acceptable property [1,2]. Normally, pure SF₆ is colorless, odorless, nonflammable and hard to be decomposed. However, several lower fluorides of sulphur SF_x ($x = 1, 2 \dots 5$) and F atoms can be generated through a corona (also called partial discharge), electric arc or spark in a gas-insulated electric power system. Although, most of the sub-fluorides will rapidly react with F atoms to regenerate SF₆ molecules again, a small portion of SF_x can further react with trace levels of unavoidable impurities (typical gases are H₂O, O₂ and N₂) or electrodes to produce numerous stable by-products [3–5]. The gaseous SF₆ decompositions (such as SO₂ and H₂S) are known to be corrosive, poisonous and chemically active, which can chemically attach the internal surface of solid insulating materials. The combination of the accumulated decomposition concentrations and the corrosive insulation materials degrade the system insulation capability, and may eventually lead to an unpredictable collapse of the electric power system. Moreover, the compositions and formation rates of SF₆ decompositions are related to the discharge insulating defect types, so that the internal status of power equipment can be identified by the composition and concentrations of SF₆ decompositions. In China, CO, H₂S and SO₂ have been recognized as indicators for the fault diagnosis of electric systems [6]. Therefore, it is critical to develop a robust, compact, and cost-effective gas sensor to monitor online the multicomponent concentration levels of SF₆ decompositions for the safety concern of personal security and electrical equipment.

As a powerful diagnostic tool for the internal conditions of the gas insulated equipment, many gas analysis techniques have been developed to detect SF₆ decompositions. These detection techniques are usually divided into chemical and optical methods. Chemical methods employ adsorption, separation and different physical and chemical properties of gaseous by-products to detect the concentration levels of analytes, such as gas chromatography [7], detection nanotubes [8,9] and electrochemical sensors [10]. However, gas chromatography has a high cost, a long response time and a larger size, and is just suitable for laboratory-based investigation rather than continuous online monitoring or portable detection. Detection tubes have poor accuracy since the concentration of decomposition products is determined by color changes. Electrochemical sensors are very cheap and can be connected to the equipment to achieve online monitoring, but with zero drift and cross talk between different gases. The optical methods are based on spectral absorption technology, which offers the unique advantages of fast response time, modest cost as well as high detection selectivity and sensitivity [11–17]. Additionally, the optical-based SF₆ decomposition detection can implement online monitoring while the power system is still energized. Such an online diagnostic tool provides significant economic benefits to power plants who cannot afford to take the system out of service for accident prevention. In recent years, a variety of optical multicomponent gas sensors have been reported in N₂ or air buffer gas [18–22]. The trace gas sensors used in the SF₆ buffer gas were demonstrated but with limited success, due to the different physical and spectral characteristics of SF₆ molecules with respect to N₂, in particular the heavy molecular weight, high density as well as the unidentified absorption spectrum in the infrared (IR) spectral range [23]. For example, H. Heise *et al.* [24] and X. Zhang *et al.* [25] employed Fourier-transform infrared (FTIR) spectroscopy technology to perform quantitative multicomponent gas analysis for the SF₆ decompositions. J. Luo *et al.* [26] employed a non-resonant photoacoustic spectroscopy sensor to detect SO₂, CO and CF₄ by using a broadband light source within 4–8 μm wavelength range. However, most by-products have cross-interference absorption with pure SF₆ in the mid-infrared (MIR) region. Such a limitation results in a degraded detection accuracy and cannot meet the detection requirement of < 1 ppmv for the SF₆ gas-insulation equipment [27,28]. Therefore, it is preferable to employ an excitation source emitting at the near-infrared (NIR) or ultraviolet (UV) spectral regions and a resonant photoacoustic cell (PAC) for the optical detection technology.

In this manuscript, we reported a sensitive photoacoustic multicomponent gas sensor for the online monitoring of the SF₆ decompositions in an electric power system. The detection and quantitative measurements of trace SF₆ by-products including CO, H₂S and SO₂ were carried out via a time division multiplexing (TDM) method. A custom made two-channel differential PAC was designed with a high-*Q* factor characteristic. Two NIR telecommunication distributed feedback (DFB) lasers were used as excitation light sources emitting at 1568.1 nm and 1582.1 nm to detect CO and H₂S, respectively. A commercialized compact erbium-doped fiber amplifier (EDFA) module was employed to boost the laser powers up to 1,600 mW and 1,300mW, respectively, which compensated the absorption line-strength loss compared to the MIR spectral region. The high-power NIR laser beam was directed to one of the PAC channels, while the other channel was used to detect trace SO₂ by means of a UV-band diode-pumped solid-state laser (DPSSL). The novel combination of designed differential PAC and detection optical resources resulted in a ppb-level detection limit for the SF₆ decompositions.

2. Selection of excitation wavelengths and optical sources

According to the *HITRAN* database, SF₆ molecule has the strong ν_3 band and the hot band (ν_4 and $\nu_4 + \nu_6 - \nu_6$) between 10 μm to 17.2 μm. However, the power system is filled with pure SF₆ as a dielectric gas, which means that the concentration of SF₆ is usually > 99.8%. The SF₆ absorption lines with weak line strengths cannot be ignored in the MIR spectral region,

although there are not listed in the *HITRAN* database. As shown in the upper part of Fig. 1, a SF_6 absorbance spectrum was experimentally achieved by using a FTIR spectrometer (Thermofisher Nicolet IS50) equipped with a 9.5-m multipass gas cell. The multipass gas cell was filled with pure SF_6 (>99.99%) at atmospheric pressure. Apparently, some uninterrupted SF_6 absorbance spectra were experimentally observed in the 3.3 μm - 10 μm spectral regions. The same experiments were performed, when the gas mixtures containing 50 ppmv CO , 50 ppmv H_2S and 50 ppmv SO_2 in N_2 buffer gas were fed into the gas cell. However, the experiment results showed that no absorption spectra for all the three gas mixtures were observed, since the limited detection sensitivity of the FTIR.

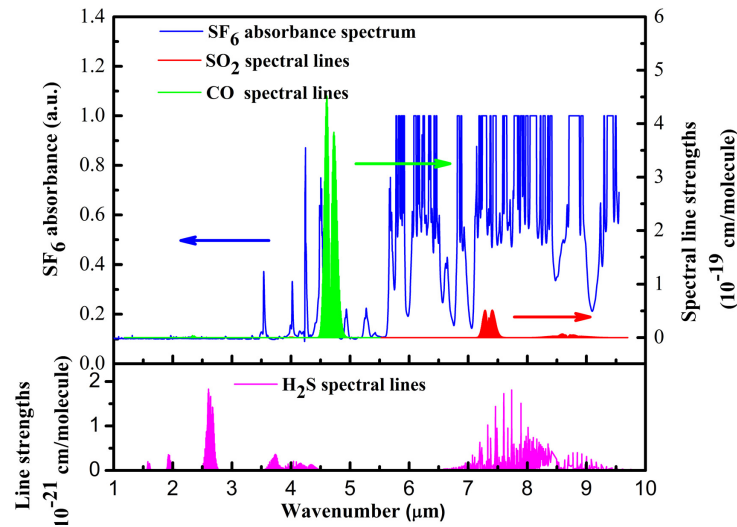


Fig. 1. Upper part: measured pure SF_6 absorbance spectra (blue line) observed by a FTIR spectrometer with a 9.5-m multipass gas cell. Simulated SO_2 (red line) and CO (green line) absorbance spectra according to the *HITRAN* database. Bottom part: simulated H_2S (magenta line) absorbance spectrum.

In order to better compare the excitation wavelengths between three target gases and SF_6 , the absorption line positions and the line strengths of CO and SO_2 molecules were also plotted in the upper part of Fig. 1, according to the *HITRAN* database. The H_2S absorbance spectrum was plotted in the bottom part of Fig. 1, due to the relative weak line strengths. Obviously, the fundamental bands of CO (located around 4.7 μm) and SO_2 (around 7.3 μm) cannot be selected for the highly sensitive quantity detection of the SF_6 decompositions, owing to interfering influence with the SF_6 absorbance spectrum. The H_2S molecules showed maximum absorption line strengths near 2.7 μm and 7.8 μm between 1 μm and 10 μm spectral regions, which were about one order of magnitude larger than the overtone absorbance band around 1.6 μm . But the spectral overlap between H_2S and SF_6 occurs at 7.8 μm and the laser sources emitting at 2.7 μm have a high cost with a low output power. Furthermore, CO molecules have also an overtone absorbance band located near 1.6 μm , which cannot be observed from the upper part of Fig. 1 due to the weak line strengths. These overtone absorbance bands of H_2S and CO are located in the NIR wavelength window for fiber-optic communications. The NIR telecommunication diode lasers are competitive for trace gas detection, because of their long life (>10 years), low cost, and low power consumption [29,30]. Moreover, they can be operated at room temperature and can be fiber coupled easily to a commercial EDFA. Due to the development of the telecommunications industry, the fiber-optic amplifiers are currently able to boost the laser power up to 3~4 orders of magnitude. Thus, the amplified power compensates the weaker absorption line strengths in the overtone band. More importantly, an L-band EDFA can be used to amplify the optical

powers in the laser wavelength ranges from 1565 nm to 1610 nm. Such a wide wavelength range can cover several laser sources when the TDM method was used, which provides many advantages for the sensor such as a small size, low cost and portable operation.

The line selection was carefully implemented to avoid the interferences from H₂O and CO₂, since H₂O and CO₂ played an important role during the SF₆ decomposition process. As shown in Fig. 2, the molecular absorption line positions and line strengths for CO, H₂S, H₂O and CO₂ were plotted within the wavenumber range of 6319.5 cm⁻¹ to 6379.0 cm⁻¹. Obviously, the two selected interference-free target absorption lines (star marks) are at least two orders of magnitude higher than the neighboring lines of the other two gases. For this work, two butterfly-type DFB lasers (Sichuan Tengguang Electronics and Technology Co., China) with central wavenumbers of 6377.4 cm⁻¹ and 6320.6 cm⁻¹ were selected as the excitation source to detect CO and H₂S, respectively. In order to obtain a miniaturized sensor size, a compact L-band EDFA (Connect laser technology, China) with small dimensions (20 × 13 × 5 cm³) was utilized to amplify the excitation power up to ~1,500 mW.

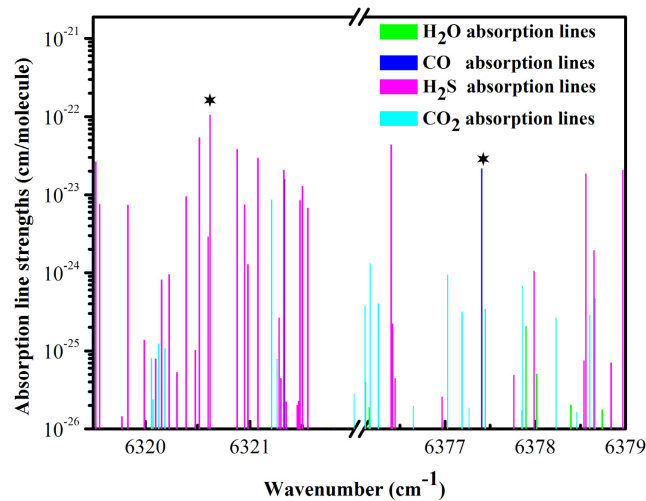


Fig. 2. Absorption line positions and line strengths of H₂O (green), H₂S (magenta), CO (blue) and CO₂ (cyan). Star marks indicate two selected interference-free target absorption lines.

According to the HITRAN database, there is no significant absorption for SO₂ molecules in the wavelength region between 1 μm and 3.5 μm. However, SO₂ has two strong absorption cross sections in the UV wavelength region due to electronic transitions, which provides an option for concentration measurements of SO₂. Moreover, the UV spectral region between 250 nm to 400 nm is also transparent for SF₆ molecules [6]. As shown in Fig. 3, the cross section curves of SO₂ were plotted for the second allowed band (¹B₂ ← ¹A₁ 165 nm-240 nm) and the first allowed band at (¹A₂, ¹B₁ ← ¹A₁ 240 nm-340 nm). The maximum cross section of the second allowed band is ~36 times higher than that in the first allowed band. However, an inconspicuous absorption band of H₂S exist with a center wavelength of 195 nm, which has a cross-talk with SO₂ and hence degrades the detection limit. Therefore, the absorption band in the wavelength range of 240 nm-340 nm was chosen as the characteristic wavelengths.

Recently, a small-size and cost-effective UV laser became commercially available, due to the development of Pr³⁺-doped based crystal lasers. It offers laser transitions in the visible spectral range, which opens a new access to the UV or even deep-UV light generation [31]. By using intra-cavity frequency doubling of a Pr:YLF-based laser and a LiB₃O₅ (LBO) crystal, a 5 mW solid-state laser emitting at 303.6 nm was achieved as the laser source to detect SO₂ in SF₆ buffer gas. The dimensions of the laser were 25 × 8 × 7 cm³ and the spectral

linewidth is ~ 0.15 nm. A quality factor value of M^2 was measured to be 1.51 using a beam profiler (Thorlabs BP209-VIS/M). The dimensions of laser spot in the x and y directions were 4.5 mm and 2.5 mm at a distance of ~ 15 cm from the laser beam exit, respectively.

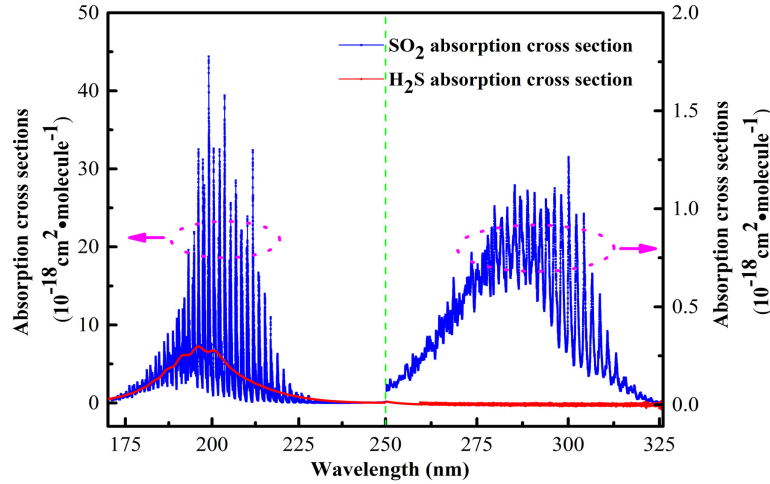


Fig. 3. SO₂ (blue line) and H₂S (red line) absorption cross section between 170 nm-325 nm.

3. Experiment setup of sensor system

3.1 Dual-channel photoacoustic detection module

The photoacoustic spectroscopy gas analysis is based on the detection of the sound signal (acoustic wave), which is generated due to localized transient heating and expansion through gas molecular collisions. The spatial size and shape of the acoustic source depend on the excitation optical beam geometry and on the absorption length in the gas. For a given excitation optical beam, the use of a PAC with high- Q factor is an effective way to increase the cell constant, since the photoacoustic signal is proportional to the cell constant. In general, the resonator Q -factor describes the energy losses during one period in the acoustic wave propagation [32,33]. For a longitudinal resonance, the contribution of the surface losses to Q -factor can be given as:

$$Q = \frac{R}{d_{vis} + (\gamma - 1)d_{th} \left(1 + \frac{2R}{L} \right)} \quad (1)$$

where γ is the ratio of the specific heats, R and L are the radius and length of resonator, d_{vis} and d_{th} are the viscous and thermal boundary layers thicknesses. By substituting the physical constants of SF₆, a background-gas-induced high- Q ($Q > 50$) PAC with a dual-channel was realized for multicomponent gases detection.

As shown in Fig. 4, the stainless steel made PAC consists of two identical cylindrical resonators, each of which have a radius of 4 mm and a length of 90 mm. The inner surfaces of the resonators were polished to facilitate the formation of standing wave. In the middle of each resonator, an electret condenser cylindrical microphone (Primo, EM158) with a sensitivity of -32 dB was embedded via a 1-mm diameter hole. The two selected microphones with a dimension of $\Phi 6 \times 10$ mm have a similar response below 1.5 kHz. The signals from the two microphones were fed into a custom trans-impedance differential preamplifier with an amplification factor of 13. The gas-inlet and gas-outlet holes were

mounted in the buffer volumes, which were connected to the both ends of the resonators with a radius of 11 mm and a length of 10 mm. Two 25.4 mm × 5 mm quartz windows are used to insulate the PAC from the surrounding environment. In order to remove the light path alignment difficulty, an optical collimator (Thorlabs, F230FC-1550) is screwed onto one of the resonators in front of the entrance window, which can be used for a high power laser up to 15 W. The entrance of the other resonator is open with an 8-mm diameter through-hole for the UV laser. After passing through the resonators, both NIR and UV lasers are collected and absorbed by a black-out rough beam dump. The dual-channel photoacoustic detection module will be used for the SF₆ multicomponent decomposition analysis described in the next section.

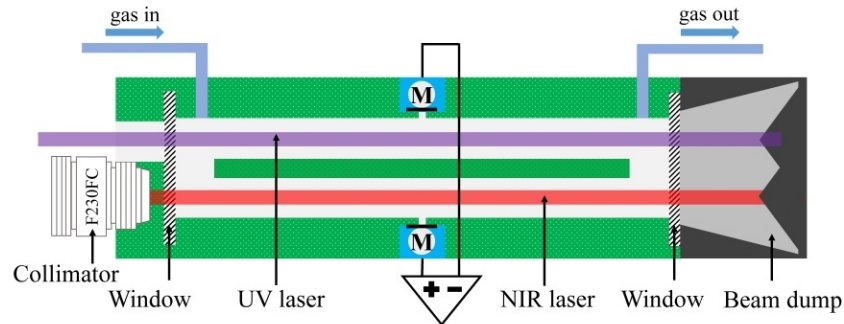


Fig. 4. Schematic of the dual-channel photoacoustic detection module.

3.2 Sensor system

A schematic of the online multicomponent gas monitoring system for the detection of the SF₆ decompositions is shown in Fig. 5. Two fiber coupled DFB lasers emitting at 1568.1 nm and 1582.1 nm were used for the detection of CO and H₂S, respectively. Wavelength modulation and second harmonic detection were employed for sensitive trace gas detection. Two 50 mHz swept ramp signals and two sine waves were generated by a digital function generator, and then added by two custom-made electric adders, respectively. The modulation frequencies of the two sine waves were set to one half of the PAC resonance frequency ($f = f_0/2 = 343.3$ Hz). The two added signals were fed into two commercial diode laser controllers (Wavelength Electronics LDTC-0520), which can sweep the laser wavelengths back and forth across the target absorption lines. The temperatures of the two DFB lasers were also controlled by the laser controllers. Both the modulated laser beams were directed to an optical switch, which can be controlled by a TTL voltage (0-5 V) for the selection of two output beams. L-band fiber amplifier module was used to boost the selected laser beam power up to 1,500 mW. The fiber amplifier module was operated in the constant power mode. An internal power meter was installed inside the fiber amplifier for a long-term output optical power calibration. After an optical collimator, a standard TEM₀₀ Gaussian laser beam with a $1/e^2$ beam diameter of 0.9 mm was obtained to pass through one of the two resonators (8mm diameter). A standard square wave was generated by the function generator to modulate the solid-state laser at the resonance frequency (686.5 Hz) of the PAC. The output UV laser beam was directed at the center line of the other resonator for the detection of SO₂. A beam dump was placed behind the PAC for the collection and absorption of both the NIR and UV laser beams. The differential amplified signals from the PAC were fed into a lock-in amplifier board (FEMTO, LIA-BV-150-L), which can also be controlled by a TTL voltage signal to select the 2- f (CO and H₂S) or 1- f (SO₂) demodulation mode. Three synchronizing signals from the function generator were directed to the lock-in amplifier. A 12 dB/oct filter slope and 1 s time constant were set for the amplifier, which corresponded to a detection bandwidth of 0.25 Hz. A touch-screen computer was used for the data acquisition, logic control and signal processing via a LabVIEW routine for three target gas detection. All experiments were carried out at

atmospheric pressure and room temperature. A gas flow rate of 100 sccm was selected, which results in a gas exchange time of ~ 6 s between the PAC and the outside, considering a PAC volume of ~ 9.8 cm³.

As shown in the inset of Fig. 5, the frequency characteristics of the PAC were experimentally investigated. At atmospheric pressure and room temperature, the fundamental longitudinal vibration frequencies of 686.5 Hz and 1772.1 Hz were achieved when filled with SF₆ and N₂ buffer gas, respectively. By calculating the ratio of resonance frequency to the half-width value of the resonance profile, a high- Q factor of 84 was obtained for SF₆ while a Q -factor of 22 was obtained for N₂, which are in excellent agreement with the calculated values of 81 and 38 for SF₆ and N₂, respectively, using Eq. (1). A detailed principle explanation for the background-gas-induced high- Q PAC can be found in our previous publication [27].

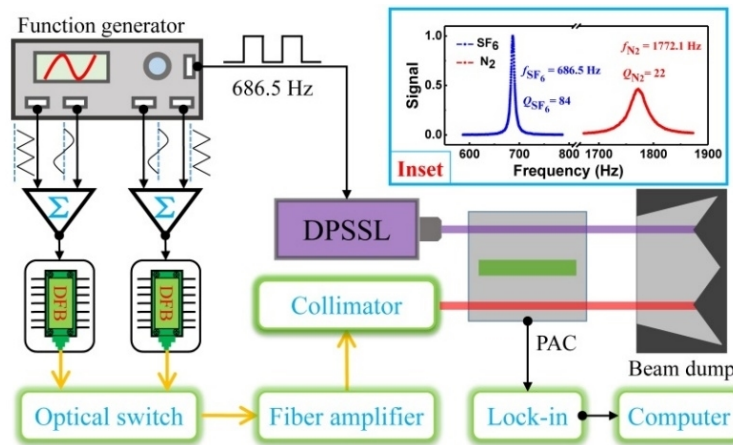


Fig. 5. Schematic of the online multicomponent gas monitoring system for SF₆ decompositions based on two NIR DFB lasers, an UV diode-pumped solid-state laser (DPSSL), a 1.5 W fiber amplifier, and a dual-channel photoacoustic cell (PAC). Inset: Resonance frequency response curves of the PAC in SF₆ (blue) and N₂ (red) buffer gases.

4. Results and discussion

The excitation power level is important for a photoacoustic gas sensor, due to the fact that the photoacoustic signal is proportional to the power. However, a saturation effect may occur with an increasing optical excitation power, so that the signal will not benefit from a higher optical power. Moreover, the erbium-doped optical fiber amplifier operating in the L-band has different power gains for different laser wavelengths. In order to achieve the gain characteristics of the selected lasers, the wavelengths of the two DFB lasers were locked at the target absorption lines (1568.1 nm and 1582.1 nm). A power meter (Ophir Optonics Solutions, 3A-ROHS) was placed behind the PAC to monitor the actual excitation power. As shown in Fig. 6(a), the actual powers of the two lasers increased linearly with the increase of the set power levels. But the laser power amplification rate was different for the two laser wavelengths. When setting the power of the fiber amplifier module to 1,500 mW, an actual power level of 1,724 mW was obtained at the wavelength of 1568.1 nm, while a power level of 1,351 mW was obtained at 1582.1 nm.

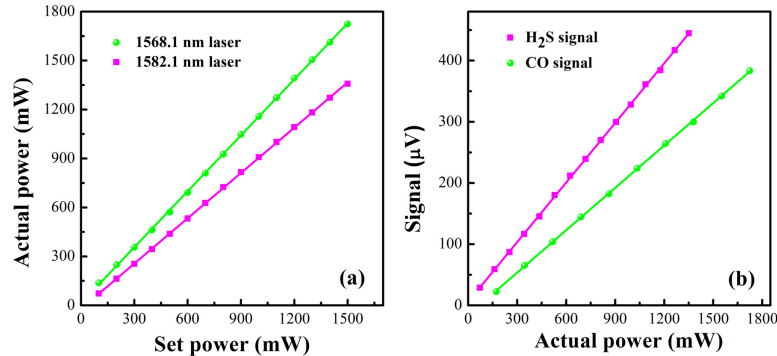


Fig. 6. (a) The actual amplified optical power levels as a function of the set power for two laser wavelengths of 1568.1 nm (green) and 1582.1 nm (magenta). (b) CO (green) and H₂S (magenta) photoacoustic signals as a function of the actual powers measured after the photoacoustic cell.

The gas mixtures containing 160 ppmv CO and 50 ppmv H₂S in SF₆ buffer gas were fed into the PAC in order to study the saturation effect. After the optimization of the modulation depths, the maximum 2-*f* signal amplitudes were recorded in Fig. 6(b). The linear fitting analysis was implemented. The obtained R-square values of 0.999 for both CO and SO₂ signals indicate that no saturation behavior with the excitation power occurred. Further experiments were implemented with the actual output mean powers of 1,724 mW and 1,351 mW for the CO and H₂S detections in order to obtain the best minimum detection limits.

In order to evaluate the detection sensitivity of the multicomponent gas sensor platform for SF₆ decompositions, the different concentrations of three target gas mixtures (CO, H₂S and SO₂) were fed into the PAC, respectively. The gas mixtures were generated by a gas dilution system (EnviroNics EN4000). A cross-talking behaviors between three target gases was not observed due to an implementation of a good line selection. The signal data points were recorded continuously for 100 s after the signal amplitude became stable. The noise levels of the sensor were defined as the standard deviations (1 σ) of the stable output signals. A 1 σ standard deviation of 1.16 μ V was obtained with pure SF₆ gas, which was used as the sensor noise level. As shown in Fig. 7(a), the CO/SF₆ signal amplitudes were plotted ranging from 0 ppmv to 300 ppmv. A signal amplitude of 53.4 μ V with 20 ppmv CO/SF₆ gas mixtures was obtained, which resulted in a signal-to-noise ratio (SNR) of 46 and a minimum detection limit of 435 ppbv. Taking into account the laser power and the detection bandwidth, a normalized noise equivalent absorption (NNEA) coefficient of $3.8 \times 10^{-8} \text{ cm}^{-1} \text{ WHz}^{-1/2}$ was determined. As shown in Fig. 7(b), the H₂S/SF₆ signal amplitudes were plotted ranging from 0 ppmv to 8.3 ppmv. For the 1 ppmv H₂S/SF₆ gas mixtures, a detection limit of 89 ppbv and a NNEA coefficient of $2.9 \times 10^{-8} \text{ cm}^{-1} \text{ WHz}^{-1/2}$ was achieved. The SO₂/SF₆ signal amplitudes ranging from 0 ppmv to 25 ppmv were plotted in Fig. 7(c). A minimum detection limit of 115 ppbv and a NNEA coefficient of $1.8 \times 10^{-9} \text{ cm}^{-1} \text{ WHz}^{-1/2}$ were obtained with the data of 5 ppm SO₂/SF₆ gas mixtures.

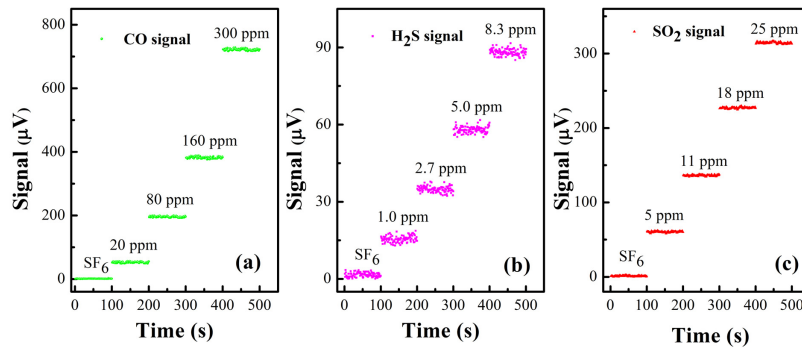


Fig. 7. (a)-(c). Photoacoustic signal amplitudes as a function of the different CO/SF₆ (green), H₂S/SF₆ (magenta) and SO₂/SF₆ (red) gas mixtures for the sensor performance evaluation.

To verify the linear concentration response, the 1000 ppmv CO/SF₆ gas was diluted with pure SF₆ to generate different concentration levels of CO/SF₆ gas mixtures. The measured maximum 2-*f* signal amplitudes as a function of the CO concentrations were plotted in Fig. 8(a). The calculated R-square value was equal to 0.9998 based on a linear fitting analysis, which indicates that the sensor system has an excellent linearity response to the CO concentration levels. The similar experiments were carried out when the 50 ppmv H₂S/SF₆ gas mixtures and the 50 ppmv SO₂/SF₆ gas mixtures were diluted with pure SF₆. As shown in Figs. 8(b) and 8(c), both the H₂S and SO₂ signals show linear responses to different concentration levels.

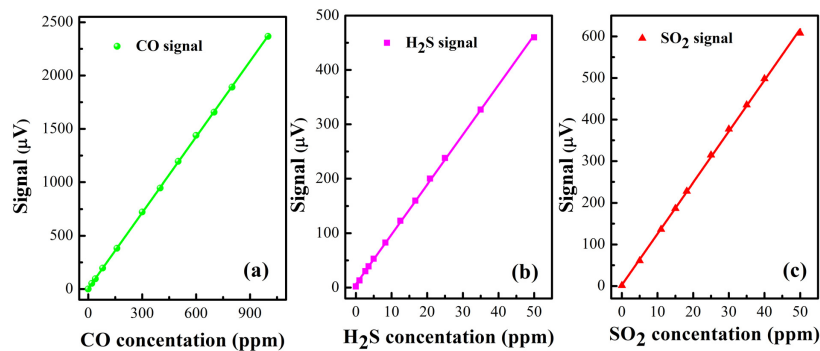


Fig. 8. (a)-(c). Response linearity of the photoacoustic F spectroscopy-based CO (green), H₂S (magenta) and SO₂ (red) sensor system for the multicomponent SF₆ decomposition analysis.

5. Conclusion

A sensitive multicomponent gas sensor was developed for the online monitoring of SF₆ decompositions in an electric power system. By using the TDM method, a dual-channel and background-gas-induced high-*Q* PAC was designed. The selection of the excitation wavelengths and optical sources were investigated in detail, since most by-products have cross-interference absorption with SF₆ in the MIR spectral region. A commercialized L-band fiber amplifier module was used to boost the optical power of two NIR lasers for the detection of CO and H₂S, which compensated the absorption line-strength loss relative to the MIR spectral region. A novel compact DPSSL laser emitting at 303 nm was used to detect SO₂ in SF₆ buffer gas. After the optimization, the sensing capacity was validated by experimental measurements of CO, H₂S and SO₂ with the minimum detection limits of 435 ppbv, 89 ppbv and 115 ppbv, respectively. With the operating pressure of the sensor at atmospheric pressure,

an automatic gas handling system including the pressure relief for sampling and the pressurization for refilling must be employed to work together with the online multicomponent gas sensor for field applications, due to the fact that the SF₆ pressure in an electric power system is 6-8 times higher than atmospheric pressure. The optimal design of the excitation optical sources and the detection module offers a sensitive, small-size and cost-effective SF₆ decomposition sensor, which is very practical for online monitoring of an electric power system.

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