Quantitative detection of H$_2$S and CS$_2$ mixed gases based on UV absorption spectrometry

Xiaoxing Zhang,$^{a}$ Zhaolun Cui,$^{a}$ Zheng Cheng,$^{b}$ Yalong Li$^{a}$ and Hai Xiao$^{c}$

H$_2$S and CS$_2$ are the decomposition components of insulating gas SF$_6$. The detection of these two gases is significant for the online monitoring and fault diagnosis of SF$_6$ electrical equipment. In this study, an ultraviolet (UV) differential optical absorption spectrometry (UV-DOAS) platform is established for detecting the concentration of H$_2$S and CS$_2$ mixed gases. Based on the platform, we obtained the UV absorption spectra of the two gases. The linear relationship between each gas absorption spectra and the concentration was established by wavelet processing and frequency analysis. The interference between the two gases in the UV spectrum region was studied. H$_2$S at different concentrations had little effect on the UV absorption spectra of CS$_2$. The linearity ($R^2$) of CS$_2$ inversion formula was 0.9997. CS$_2$ above 50 ppb produces a great interference on H$_2$S concentration detection. CS$_2$ concentration has a linear relationship with the H$_2$S concentration inversion; hence, the H$_2$S correction formula with CS$_2$ concentration as the variable is proposed. With the correction formula, the linearity ($R^2$) of H$_2$S inversion formula reaches 0.9994 in mixed gas detection, which can meet the H$_2$S and CS$_2$ mixed gases quantitative detection.

1 Introduction

Sulfur hexafluoride (SF$_6$) has good electrical properties and excellent arc suppression performance. As an insulating gas, SF$_6$ is widely used in a variety of high-voltage electrical equipment. In the SF$_6$ insulation equipment, partial discharge (PD), partial overthermal faults (POF), and other failures may cause the decomposition of SF$_6$; this finding may result in CS$_2$, SO$_2$, H$_2$S, SOF$_2$, and SO$_2$F$_2$ decomposition components. These decomposition components on one hand will exacerbate the equipment faults, on the other hand may cause harm to the safety of personnel. Therefore, SF$_6$ decomposition components and the monitoring of insulation faults, such as PD, must be diagnosed.

H$_2$S occurs mainly in the SF$_6$ overheating decomposition like PD and POF with trace moisture involved. SF$_6$ decomposes and generates S$^{2-}$ after the collision, H$_2$O breaks and generates H$. S^{2-}$ and H$^+$ eventually combine to generate H$_2$S. The chemical formula of the reaction mechanism is described as follows:

$$ SF_6 + 2e \rightarrow S^{2-} + 6F \quad (1) $$

$$ S^{2-} + 2H^+ \rightarrow H_2S \quad (2) $$

In relevant studies, CS$_2$ occurs in PD and POF failures when the molecular structure of epoxy insulator deteriorates in relatively high temperature. Some active pieces like CH$_2$, CH and C will be generated and react with SF$_6$ or its decompositions. Generation of CS$_2$ is showed in Fig. 1.

Among them, C atoms are derived from organic insulating materials and stainless steel, and S atoms are derived from SF$_6$. Generation of S atoms requires SF$_6$ to break six S-F bonds, which correspond to a higher energy; hence, CS$_2$ can be detected at higher temperatures. Known from the Arrhenius law in the chemical reaction kinetics, the chemical reaction rate is exponentially related to the reaction temperature. When the local overheat temperature is low, the CS$_2$ generation reaction is slow; when the local overheat temperature is high, CS$_2$ generation reaction abruptly accelerates. CS$_2$ is an important characteristic component to judge the existence of solid insulation.

![Figure 1](image-url)  Generate path of CS$_2$. 

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defects. CS₂ detection is of great significance to the fault diagnosis of SF₆ insulation equipment.

At present, the detection of SF₆ characteristic decomposition components is achieved by two main methods: chemical detection methods and optical detection methods. Chemical gas detection methods include gas sensor method, detection tube method, and gas chromatography method; optical methods include infrared Fourier transform spectroscopy, photoacoustic spectroscopy, and ultraviolet (UV) absorption spectroscopy. In comparison with the optical detection method, the chemical detection method has the characteristics of long detection time and high detection precision. Long detection time makes it not suitable for online monitoring. The infrared detection time and high detection precision. Long detection spectroscopy. In comparison with the optical detection method, the chemical detection method has the characteristics of long detection time and high detection precision. Long detection time makes it not suitable for online monitoring. The infrared absorption spectrum can realize online monitoring. However, the infrared absorption characteristic peaks of SF₆ gas and various component gases are very close, and the overlapping of characteristic peaks easily occurs, which limits the detection accuracy. Photoelectric spectrum has a high sensitivity but is susceptible to ambient temperature, pressure, and external noise. The above methods have limitations in the online monitoring of the SF₆ characteristic decomposition component.

The external electrons in the molecules are excited by photons in different energy levels. The number of electron layers of different molecules and the energy levels of each layer are different; hence, difference in UV absorption spectra of different molecules is observed. One way to quantitatively study of a unknown gas can be realized by analyzing the peak position and peak of the absorption spectrum. UV differential absorption spectroscopy (UV-DOAS), by fitting the slow absorption part of the original absorption spectrum and subtracting it, can effectively eliminate the influence of spectral absorption caused by Rayleigh scattering and Mie scattering on the gas concentration measurement and has strong anti-interference ability.

H₂S and CS₂, as the important decomposition products of SF₆ in the PD and POF insulation failure under, have absorption peaks in the UV spectrum region. Among them, H₂S absorption peaks distribute in 180–230 nm, and CS₂ absorption peak distribute in 190–210 nm; an overlap between the two gases absorption peaks is found.

In 1963, Kleman et al. studied the absorption properties of CS₂ at 190–210 nm. CS₂ spectrum in the range of 285–340 nm was obtained by Ahmed and Kumr. Due to the weak absorption at 285–340 nm band, when the light path is short or the low UV lamp power is low, the absorption cannot be measured in the band; hence, this study on the CS₂ UV spectral concentrated in the 190–210 nm band. In 2004, Yu et al. constructed a gas pool with a length of 1400 m and increased the detection limit of CS₂ to 2 ppb using UV absorption spectroscopy. However, studies on the quantitative detection of H₂S gas UV spectroscopy are few. The optical detection methods of H₂S are mostly photoacoustic spectroscopy and infrared spectroscopy.

In this paper, based on the UV differential absorption spectroscopy, the optical detection platform for H₂S and CS₂ was constructed. First, the UV absorption spectra of the two gases were obtained according to the experiment. For each gas, the relationship between the absorption spectrum and the concentration was established by wavelet transform and frequency domain analysis. The cross-effects of the two kinds of gas absorption peaks at different concentrations were studied. The H₂S concentration inversion formula in the presence of CS₂ was obtained. The quantitative detection method of H₂S and CS₂ mixed gases was put forward, and the mixed gases with different mixing ratios can be detected in high accuracy. This method effectively utilizes the regularity of the absorption of two gases in the UV spectrum region, and realizes the quantitative detection of the two gases mixture, which is suitable for the quantitative detection of SF₆ decomposition components.

2 Theory

2.1 Principle of UV differential absorption spectrometry

One way of quantitative detection by UV-DOAS is to utilize the absorption characteristics of gas molecules in the UV spectrum region. The measurement principle, based on Beer–Lambert law, is as follows:

\[
c = A(\lambda)/(\sigma(\lambda) L)
\]

where, \(c\) is the concentration of the light-absorbing medium, \(A(\lambda)\) is the absorbance at the corresponding wavelength position, \(\sigma(\lambda)\) is the absorption cross-section, and \(L\) is the effective absorption path of the medium.

To eliminate the non-spectral noise signals, such as the dark current generated by the photodetector during the spectral acquisition process, and the background noise signal, such as the jitter of the optical fiber during the experiment, \(A(\lambda)\) can be expressed as:

\[
A(\lambda) = \ln \left( \frac{I_0(\lambda) - I_s(\lambda)}{I(\lambda) - I_s(\lambda)} \right)
\]

where, \(I_0(\lambda)\) is the transmitted light intensity through vessel without light-absorbing medium, \(I(\lambda)\) is the transmitted light intensity through the light-absorbing medium, and \(I_s(\lambda)\) is the dark spectrum measured when no incident light is found.

The differential absorption spectrum, as the fast-changing part, can be obtained by separating the slow-changing part from the original absorption spectrum. The fast-changing part characterizes the gas absorption spectrum information and the slow-changing part is caused by Rayleigh scattering, Mie scattering, and airflow quiver. This part is defined as:

\[
F(\lambda) = A(\lambda) - S(\lambda)
\]

where,

\[
S(\lambda) = \sum_i c_i \sigma_{ib}(\lambda) + \epsilon_R(\lambda) + \epsilon_m(\lambda) + E(\lambda) L
\]

\[
c' = F(\lambda)/(\sigma'(\lambda) L)
\]

$\sigma_b(\lambda)$ is the absorption cross section changing slowly with the wavelength; $\sigma'(\lambda)$ is the absorption cross section changing rapidly with the wavelength; $\epsilon(\lambda)$ is the Rayleigh absorption coefficient; $E(\lambda)$ is the parameter characterizing gas jitter, instrument jitter, and other influence factors; $F(\lambda)$ is the differential absorption spectrum; and $S(\lambda)$ is the slow-changing spectrum, which can be fitted according to the basic variation of original absorbance spectrum $A(\lambda)$.

2.2 Absorption peaks of the two gases

The UV absorption spectrum of H$_2$S can be obtained from the MPI-Mainz database. Fig. 2 shows the UV absorption peak data of H$_2$S. H$_2$S absorption peak is mainly concentrated in the range of 180–230 nm, which belongs to the deep UV region.

According to the CS$_2$ data provided by MPI-Mainz database, main absorption peaks in the UV 190–210 nm band and weak absorption peaks in the 285–340 nm band are found. As it’s shown in Fig. 3.

Therefore, due to the spectra superimposed in the wave-number domain, H$_2$S cannot be quantitatively analyzed by using the concentration inversion expression of single gas directly. However, given that CS$_2$ is not affected in the wave-number domain by H$_2$S, CS$_2$ can be quantified by the inversion formula of CS$_2$. Therefore, if we can find the influence rule of CS$_2$ on H$_2$S, then the H$_2$S quantification in the mixed gases can be realized. This finding is also the key to achieving simultaneous detection for mixing gas of CS$_2$ and H$_2$S.

3 Experiment

3.1 Experiment set

This platform mainly includes UV light source, gas absorption cell, spectrometer, host computer, gas sample compounder, vacuum pump, and other components, shown in Fig. 4. Among them, the UV light source (Ocean Optics D2000) can output 190–400 nm range of stable UV spectrum, a peak-to-peak stability of less than 0.005%, and per hour drift is within 0.5%. With regard the spectrometer (Ocean Optics maya2000pro spectrometer), the spectral range of 165–1100 nm, and the standard optical resolution is at 0.25 nm. Fibers (Ocean Optics QP600) can achieve 80% transmittance above 180 nm. The gas absorption cell is made of custom stainless steel. The internal light is reflected once, and the optical path length is 0.8 m. The inner wall of the cell is coated with Te$_2$On coating, which can effectively prevent the adsorption of test gases. The gas distributor is a gas sample compounder, which has the largest dilution ratio is of 300 : 1 and an accuracy of ±1% FS. Standard gases (New-radargas Co., Ltd., Wuhan) are high purity nitrogen, 2 ppm CS$_2$, and 50 ppm H$_2$S.

3.2 Experimental operation

We prepared different concentrations for the two kinds of single gas and the different ratios of mixed gases by gas sample compounder. The specific mix ratio is discussed in Section 3.3. The experimental gases were prepared from low to high concentration. Before the test, the gas cell was cleaned three to five times with high-purity nitrogen gas. Each time, the cell was vacuumed and inflated to atmospheric pressure with nitrogen, then left to stand for 3 min. After cleaning, the cell was inflated to atmospheric pressure with nitrogen. The dark spectrum is collected in the case of no-light, and the background spectrum

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**Fig. 2** UV absorption cross section spectra of H$_2$S.

**Fig. 3** UV absorption cross section spectra of CS$_2$.

**Fig. 4** Schematic diagram of the UV-DOAS detection system.
is collected in the case of with-UV-light, and the light source is confirmed to be stable by the spectrometer before collecting the background spectrum. Finally, the test gases are collected in a low-to-high-concentration order. The gas cell was evacuated and washed twice after each detection. Each group was collected with 10 spectral data; one of which was used for quantitative analysis, and the other nine groups were used to verify the concentration inversion expression. The obtained dark spectrum, the background spectrum, and the test gas spectrum were placed into the formula (4) to calculate UV absorption spectrum. Through the baseline deduction to remove the slow-changing part of the spectrum, differential optical absorption spectrum can be obtained as described in formula (5).

### 3.3 Filters of the spectrum

The UV differential absorption spectra directly obtained by experiments cannot obtain good detection accuracy. On the one hand, the existence of some noise that cannot be removed by UV-DOAS theory. On the other hand, the measurement of mixed gases will cause the spectrum to superimpose, which is not enough to achieve the effective separation of the two gas spectra in the wavelength domain; this finding means that quantitative detection of mixing components cannot be achieved. To solve the above problems, this paper combined the wavelet transform and FFT transform to extract further the feature quantity in the UV differential absorption spectrum and to eliminate the effect of the interference noise. In this paper, Meyer wavelet is used. The wavelet function and the wavelet’s scale function are both defined in the frequency domain and have a fast convergence rate. Through the experimental test, the optimal wavelet scale for CS₂ and H₂S filtering is 10 and 33, respectively. Interference noise in the spectrum was removed by Meyer wavelet filter, and the feature information is enhanced several times. The details are discussed in Section 4.1.

### 3.4 Gas distribution

Compared with CS₂, H₂S are more likely to produce and with a produce higher yield in POF fault 30,31 so go to the following concentration. For each single gas, H₂S test gas was prepared in 1, 2, 5, 10, and 20 ppm, CS₂ test gas were prepared in 10, 20, 50, 100 and 200 ppb. The mixtures of both gases prepared in nitrogen because SF₆ and N₂ neither has absorption in 190–400 nm and don’t have a interference for detection. The mixing gas for the two gases is shown in Table 1, where, “✓” indicates that the combination is prepared.

#### 4 Results and discussion

### 4.1 Single-gas inversion expression

#### 4.1.1 H₂S

Through a large number of experiments, the results are shown in Fig. 5. Fig. 5(a) shows the UV absorption spectra of H₂S at different concentrations. The overall absorbance increases with the increase of the concentration. UV absorption of H₂S ranges from 190 nm to 250 nm; however, the narrow band absorption is only around 190–210 nm.

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**Table 1** Mixture gases of CS₂ and H₂S

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<tr>
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</table>

**Fig. 5** UV spectrum of H₂S. (a) UV absorption spectrum. (b) Differential spectrum.
Concentration of H$_2$S of 5, 10, and 20 ppm showed a narrow band absorption, whereas the narrow band absorption is very weak for 2 and 1 ppm H$_2$S. Fig. 5(b) is the corresponding UV difference absorption spectra extracted from the UV absorption spectra of H$_2$S at different concentrations. Data are not found with regard the absorption in the rear band of 210 nm, and the H$_2$S at each concentration shows narrowband absorption. The narrowband absorption of H$_2$S gradually increases with the increase of the concentration. The UV differential absorption spectra of H$_2$S are affected by the high-frequency noise; the lower the concentration, the more serious the influence there is.

A bandpass filter algorithm for the extraction of UV differential absorption spectra of H$_2$S was constructed by using Meyer wavelet. The differential absorption spectrum after treatment is shown in Fig. 6(a), and the UV absorption spectrum after filtering is further transformed by FFT; the results shown in Fig. 6(b). Fig. 6(a) shows that (1) the characteristic information of the UV differential absorption spectrum is very concentrated, almost no high and low wave number of noise interference, (2) peaks of different concentrations appear at the same position (10 nm$^{-1}$), and the peak value of H$_2$S increases with the increase of H$_2$S concentration; this finding means that the concentration can be well characterized.

To avoid the randomness of single data point, the algebraic sum of the FFT values of three points (9 nm$^{-1}$, 10, and 11 nm$^{-1}$) is chosen as the FFT eigenvalues to characterize the trace H$_2$S concentration. The FFT eigenvalues at different concentrations were calculated. Least squares method was used to linearly fit the H$_2$S concentration and its corresponding FFT eigenvalues. The fitting results are shown in Fig. 7. The FFT eigenvalues and H$_2$S concentration have a high degree of linearity, which reaches 0.9999 ($R^2$). H$_2$S concentration can be obtained by inverse expression as:

$$y = 1.564x + 0.571$$

where, $y$ is the FFT eigenvalue of the H$_2$S, and $x$ is the H$_2$S concentration (ppm). The uncertainty of slope and intercept is $4.83 \times 10^{-3}$ and $4.98 \times 10^{-2}$.

4.1.2 CS$_2$. The obtained CS$_2$ UV absorption spectrum at each concentration is shown in Fig. 8(a). CS$_2$ has UV absorption at each concentration, and the absorption is increased with the increase of concentration. The interval between the narrow band absorption peaks is significantly smaller than that of H$_2$S. The UV differential absorption spectra at each concentration were extracted, and the results are shown in Fig. 8(b). The UV differential absorption spectra of CS$_2$ at different concentrations show similar characteristics. With the increase of the concentration, the differential absorption is enhanced. When
the CS₂ concentration is low, high-frequency noise in the differential spectrum is observed, and the spectrum is easily affected.

The UV differential absorption spectra of each CS₂ concentration were filtered using the Meyer wavelet; the results are shown in Fig. 9(a). In the figure, the differential absorption spectra of each concentration after filtering treatment are more obvious and the spectrum is very smooth. Almost no interference information is found. With the increase of concentration, the spectral characteristics of CS₂ are similar, and the absorption gradually strengthens. The FFT frequency spectrogram results are shown in Fig. 9(b). Fig. 9(b) shows that the characteristic information is concentrated after filtering treatment, and the peak appears at 30 nm⁻¹, which is different from H₂S. With the increase of CS₂ concentration, the FFT value increases simultaneously. Therefore, the UV differential absorption spectrum after filtering and its FFT value in the wavenumber domain have the ability to realize quantitative analysis of CS₂. The FFT eigenvalues of the UV absorption spectrum were used for CS₂ concentration inversion.

The algebraic sum of the FFT values (9, 30, and 31 nm⁻¹) is chosen as the FFT eigenvalues to characterize the concentration of CS₂, and the least squares method is used to linearly fit the concentration and FFT eigenvalues. In Fig. 10, results show a high degree of linear relationship between the FFT eigenvalues and CS₂ concentration, and the linearity ($R^2$) is as high as 0.9998. The inversion expression is:

$$y = 0.1434x + 0.1062$$  \hspace{1cm} (9)

where, $y$ represents the FFT eigenvalue of the CS₂, and $x$ represents the CS₂ concentration in ppb. The uncertainty of slope and intercept is $7.58 \times 10^{-4}$ and $7.76 \times 10^{-2}$. 

![Fig. 8](image) UV spectrum of CS₂. (a) UV absorption spectrum. (b) Differential spectrum.

![Fig. 9](image) UV difference absorption spectroscopy after filtering and its FFT spectrum. (a) Differential spectrum after filter. (b) FFT frequency spectrogram.
4.2 Mixed gas detection

In Fig. 1 and 2, the absorption coefficient of CS₂ is about two orders of magnitude larger than that of H₂S. To visually show the intersection of the two kinds of gases near UV 200 nm band, the spectra of 20 part per million (ppm) of H₂S and 200 part per billion (ppb) of CS₂ are detected, and the two are superimposed on Fig. 3.

In Fig. 11, the two gases in the UV spectrum region have a serious cross; this finding means that each absorption spectra of the two gases cannot be separated directly by UV differential spectroscopy, and the two gas concentrations cannot be obtained by inversion method. The slow-changing part of the absorption spectrum was removed by baseline deduction, and the differential absorption spectrum was obtained. The obtained differential spectrum was inverted into the frequency domain by FFT transform. The results are shown in Fig. 12.

Fig. 12 shows that the differential absorption spectra of the two gases are superimposed in the wavelength domain, and the UV differential absorption spectra of H₂S are all submerged in the spectra of CS₂. Fig. 4(b) shows that the eigenvalues of CS₂ in wavenumber domain are not affected by H₂S; however, the CS₂ contains characteristic information for H₂S quantification, which will have an effect on the quantitative detection of H₂S.

4.2.1 Inversion results. The mixed gases are prepared as discussed in Section 3.4. The spectral data of each mixture were collected in 10 groups to avoid experimental errors. The differential absorption spectra of the two gases were extracted by the filters, and the CS₂ and H₂S concentrations were calculated by using the concentration inversion expressions in Section 4.1. The results are shown in Table 2. The concentrations were calculated by taking the average after the minimum and maximum were removed in 1 of the 10 groups. Δ is the difference value between the calculation results and the actual concentration of H₂S. δ is the percentage of error. In Table 2, the relationship between the inversion concentration and the actual concentration of the two gases were described.

In Fig. 13, the linearity (R²) of CS₂ concentration is up to 0.9997 mixed with different H₂S concentrations; this result indicates that the presence of mixed H₂S has little effect on CS₂ concentration inversion. In Table 2 and Fig. 14, when the CS₂...
concentration is small (10 and 20 ppb), little effect on the inversion of H$_2$S is observed. However, when the concentration of CS$_2$ is large (50, 100, and 200 ppb), the inversion of H$_2$S concentration is disturbed. Besides, the effect basically shows a linear increase with the increase of CS$_2$ concentration. The two fitting lines shown in Fig. 14 are almost parallel; this result indicates that the same concentration of CS$_2$ has a relatively close effect on different concentrations of H$_2$S. Thus, the lower concentration of H$_2$S is disturbed more by CS$_2$.

### 4.2.2 Correction expression of H$_2$S

In accordance to the previous section, the concentration of CS$_2$ in the mixed gases of H$_2$S and CS$_2$ can be calculated directly, and the H$_2$S concentration inversion expression must be corrected. Considering the H$_2$S detection limit, and the unavoidable errors, such as gas operation. Low concentration detection should be of priority. The concentration effects were calculated through the three concentrations. CS$_2$ in 50, 100, and 200 ppb were set to 0.6, 2.0, and 4.6 ppm H$_2$S increase. The correction expression was

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<th>$\Delta_h$ (ppm)</th>
<th>$\delta$ (%)</th>
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**Fig. 13** Inversion concentration of CS$_2$ with different concentrations of H$_2$S.

**Fig. 14** Inversion concentration of H$_2$S with different concentrations of CS$_2$. 

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RSC Advances

Direct inversion results of CS$_2$ and H$_2$S mixed gases

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50896 | RSC Adv., 2017, 7, 50889–50898

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Referring to the image, the text seems to be discussing the detection and quantification of hydrogen sulfide (H$_2$S) and carbon disulfide (CS$_2$) in mixed gases. The text highlights the use of UV absorption spectroscopy for detecting these gases, with emphasis on the correction expressions used to improve detection accuracy. The table provided at the bottom of the page shows revised inversion results of H$_2$S in mixed gases, with different concentrations of CS$_2$ and H$_2$S. The corrected values for H$_2$S concentration are shown for various concentrations of CS$_2$.

The corrected formulas are given as:

$$D_h = 0.0259 \times c_c - 0.45 \quad (R^2 = 0.9997) \quad (10)$$

Where $D_h$ represents the increase in H$_2$S caused by CS$_2$ (unit: ppm), $c_c$ represents the concentration of CS$_2$ (unit: ppb).

### Table 3 Revised inversion results of H$_2$S in mixed gases

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<tr>
<td>1</td>
<td>0.65</td>
<td>-0.33</td>
</tr>
<tr>
<td>2</td>
<td>1.82</td>
<td>0.18</td>
</tr>
<tr>
<td>5</td>
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<td>0.04</td>
</tr>
<tr>
<td>10</td>
<td>10.29</td>
<td>0.29</td>
</tr>
<tr>
<td>15</td>
<td>15.27</td>
<td>0.27</td>
</tr>
<tr>
<td>20</td>
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</table>

### 5 Conclusion

In this paper, the detection platform of H$_2$S and CS$_2$ based on UV absorption spectroscopy was established. First, two kinds of single gases were detected, and the concentration inversion expressions of the two gases were obtained. After that, the mixed gases at different ratios were detected. H$_2$S has little effect on the detection of CS$_2$, and the effect of CS$_2$ on H$_2$S concentration inversion is linear with CS$_2$ concentration. Basing on the data analysis, we obtained the correction expression of CS$_2$ gas to H$_2$S concentration. Expression validity was proved by the actual data verification. Hence, the H$_2$S and CS$_2$ gas can be detected by using the proposed modified detection expressions, which can realize the high-precision quantitative detection of the two gases and provide technical support for the online monitoring of SF$_6$ decomposition components.

### Conflicts of interest

There are no conflicts to declare.

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