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TDLAS Detection of propane and butane gas over the near-infrared wavelength range from 1678nm to 1686nm

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Abstract. It is important in the petrochemical industry that there are high sensitivity, high accuracy, low-power consumption and intrinsically safe methods for the detection of propane, butane and their gas mixtures, to provide early warning of potential explosion hazards during both storage and transportation of oil and gas. This paper proposes a ‘proof of principle’ method for the detection of propane and butane using a Tunable Diode Laser Absorption Spectroscopy (TDLAS) technique over the near-infrared wavelength range from 1678nm to 1686nm. This method is relatively inexpensive to implement and is thus more practical, compared with detection methods using wavelengths further into the infra-red, near 3.3μm. The minimum detectable concentration was found to be low as 300ppm for propane or butane. Importantly, the relative measurement errors were all below 3% LEL, which meets the requirements from the petrochemical and oil-gas storage and transportation industries for a field-based system for monitoring of combustible gases.

1. Introduction and background
Both propane and butane are colorless and combustible and a major explosion hazard exists when propane/butane gas mixtures meet open flames or sparks[1]. Furthermore, propane and butane are volatile at normal temperatures and pressures and are heavier than air and thus tend to accumulate in low-lying areas, making them harder to disperse and increasing the hazard as gases can easily accumulate[2]. Thus having portable, inexpensive monitoring systems for propane and butane gas is important and Tunable Diode Laser Absorption Spectroscopy (TDLAS) is an infrared absorption spectroscopic technology which uses tunable diode lasers as sources. TDLAS has been widely used for the detection of many kinds of flammable gas such as methane, ethylene and the like because of its use of a stable laser source, the high spectral resolution available and the relatively simple construction parameters of a system designed for field use. Furthermore, such an instrument can readily be combined with fiber optic components, which can provide an ideal solution for gas sensing because of the simplicity of the system, which can be battery powered for on-site monitoring, creating an intrinsically safe instrument. TDLAS-based sensors are thus well suited to the monitoring of explosive gas mixtures in the petrochemical and oil-gas storage and transportation industries.
In recent years, there have been several reports of the detection of propane and butane by using TDLAS at longer wavelengths, near 3.3 μm\[^3\]^\[^4\]. However, both the current commercial tunable lasers emitting near 3.3 μm and the optical fiber needed are both expensive and not readily available, limiting the applicability of the technique in this mid-infrared wavelength range and thus the practicability of such a system for monitoring in these industries.

Switching to a near-infrared wavelength band would create a much more practical system due to the considerably lower cost of the lasers in the near infrared band and the use of ‘communications grade’ optical fiber. However, in this near-infrared region (and especially from 1678nm to 1686nm) the absorption spectra of propane and butane consist of spectral bands with overlapping absorption peaks, where in addition the spectra bands of propane and butane overlap with each other. It is therefore difficult to find obvious ‘standalone’ absorption peaks to use to allow the calculation of the component concentrations (using the usual TDLAS spectral method) because this is also a part of the overall absorption spectrum and what is worse, for the propane/butane gas mixture, the absorption spectra of propane and butane severely overlap with each other. All this makes it much more difficult to calculate the individual component concentrations using the absorption spectra monitored from the gas mixture (as they include components of both the propane and butane samples at every wavelength).

This work uses the spectral features of propane and butane over the range from 1678nm to 1686nm to create an instrument based on a Vertical Cavity Surface Emitting Laser (VCSEL) which covers a wider range than the usual DFB lasers, thereby allowing more information to be included – enabling both qualitative and quantitative analysis of the propane and butane gas components in the mixture. For propane or butane gas alone, the Principle Component Analysis (PCA) method was used for the quantitative analysis. For propane/butane gas mixtures, reference gas cells were used to allow in the analysis the removal of the severe spectral overlap from the propane and the butane, simplifying the corresponding quantitative analysis. The results obtained are of sufficient accuracy to satisfy the requirements for explosion early warning, emphasizing the applicability and value of the monitoring methods for use in industry.

2. Quantitative analysis of absorption spectra of propane and butane

The absorption spectra of propane and butane in the spectral region from 1678nm to 1688nm are shown as Figure 1.

As a result, a VCSEL laser source operating from 1678.7 nm to 1686.1nm was selected from a stock of commercial tunable lasers, to use with an analysis based on the PCA method. Experiments were carried out on sample mixtures and results of higher accuracy were obtained, compared traditional
TDLAS detection methods with the use of a DFB laser and without PCA analysis methods. These are shown in Figure 2 and Figure 3 where it can be seen that the minimum detectable concentration could reach as low as 300ppm (and thus relative errors of the gas concentrations measured in this way were all below 5%).

![Figure 2](image1.png)

**Figure 2.** Results for the calculation of different concentrations of propane when the known concentrations of propane were used (in steps, with 10 tests for each concentration) in 200ppm increments from a starting concentration of 300ppm, up to 2200 ppm.

![Figure 3](image2.png)

**Figure 3** Relative errors in the results shown in Figure 2

The propane/butane gas sample mixture used shows that the absorption spectra overlap with each other quite severely. To solve this, the approach taken used several reference gas cells to obtain the absorption spectra of pure propane and butane gases alone, each of known concentrations and using
these data, the problems of severe spectral overlapping could be effectively removed, as shown in Figure 4 where a propane/butane gas mixture sample was investigated using the method. The mixture consisted of propane, butane and nitrogen, where the concentration of propane was only 0.05 times that of the Lower Explosion Limit (LEL) of propane, which was calculated as 0.05 × 2.2% (the LEL percentage), that is 1100ppm; the concentration of butane was 0.35 time the LEL of butane, which was calculated as 0.35 × 1.8% (the LEL percentage), that is 6300ppm. The relative errors of the concentrations calculated for propane and butane in the mixture could reach as low as 0.16% LEL and 2.78% LEL for these gases respectively. This meets the requirements from the petrochemical and oil-gas storage and transportation industries for their monitoring needs.

![Figure 4 Illustration of the removal of the spectral overlapping for the sample shown](image)

3. Conclusion
To tackle the difficulty existing in the use of TDLAS-based detection techniques for combustible gases such as propane, butane and their mixtures, this work has proposed a method based on TDLAS detection of propane and butane, using near-infrared wavelengths between 1678nm to 1686nm. Such an approach is more practical and lower in cost than methods based on the detection of wavelengths near 3.3μm. Thus this provides an excellent ‘proof of principle’, based on experimental results, for the better design of TDLAS-based field sensors for field monitoring in the petrochemical storage and transportation industries.

4. References