

USING TUNABLE DIODE LASER ABSORPTION SPECTROSCOPY (TDLAS) FOR CARBON DIOXIDE MEASUREMENTS IN LIQUIFIED NATURAL GAS PROCESSING

The liquefied natural gas (LNG) plant is a large and complex industrial facility. The liquefaction process, which transforms natural gas to liquid, involves operations at very low temperature. At these conditions carbon dioxide (CO₂) can freeze on the exchanger surface, plugging lines and reducing plant efficiency. The natural gas entering the liquefaction plant will often contain several contaminants that must be reduced to ensure satisfactory LNG plant performance and to meet LNG sales specifications.

The liquefaction process involves the removal of components such as acid gases, helium, water, and heavy hydrocarbons. Acid gas removal is the next important step, reducing CO₂ levels to about 50 parts per million by volume (ppmv) to meet normal sales gas specification. There are many acid gas treating processes available for the removal of CO₂ from natural gas. The selection of these processes is based not only on economic feasibility but also the effective removal of CO₂.

These processes include chemical solvents, physical solvents, hybrid solvents, adsorption processes and physical separation based on membrane systems. Each of these technologies has advantages and limitations. The nature and amount of contaminants in the feed gas, as well as the targeted removal capacity, the amount of hydrocarbons in the gas, the amount of gas to be processed, and the desired selectivity, should all be considered. Despite the nature of the process used to purify the natural gas, optimization of the process requires measurement of the CO₂ before and after purification.

Amine gas treating refers to a group of processes that use an aqueous solution of various amines to remove hydrogen sulfide (H₂S) and CO₂. Today it is a common process used in refineries, petrochemical plants, and natural gas processing plants. The use of blended amines in gas treatment provides significant improvement in the absorption capacity, absorption rate and in meeting solvent regeneration energy requirements. In most cases the amine mixtures contain methyldiethanolamine (MDEA) as the base amine, with the addition of either one or two more reactive amines such as monoethanolamine (MEA) or diethanolamine (DEA).



The typical amine treatment unit involves two stages. The first stage is the amine contactor, where natural gas is brought into contact with amine. In the absorber, down-flowing amine solution absorbs H₂S and CO₂ from the up-flowing sour gas to produce a sweetened gas stream with significantly reduced acid components. Sweet gas leaves the top of the absorber and flows to a dehydration unit before liquefaction. The resultant rich amine, with absorbed CO₂ and H₂S, then flows to the regenerator, which is a stripper with a reboiler. In the second stage, H₂S and CO₂ are stripped from the liquid phase to regenerate the lean amine solution, which is returned to the amine contactor column. The general process flow diagram for amine CO₂ removal is shown in Figure 1.

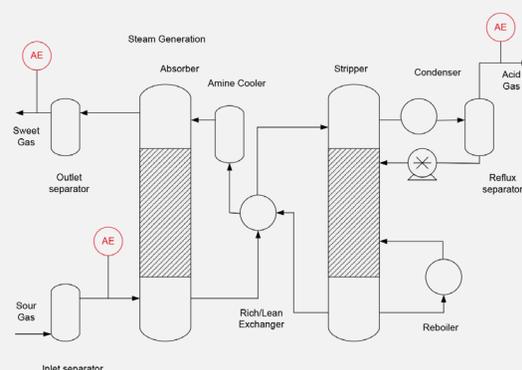


Figure 1. Amine CO₂ removal process

Acid gas and steam pass through a condenser, where the steam is condensed and cooled and returned to the top of the stripper as a reflux. Acid gas is separated and sent to the flare or compressed for the sequestration process. The acid gas leaves through the top of the stripper column. The lean amine from the bottom of the stripper is returned to the absorber through the heat exchanger and the cooler. There are several control points for the measurement of the CO₂ level for process optimization. These control points correspond to sour gas at the inlet of the absorber, sweet gas at the outlet of the absorber, and to the level of CO₂ in acid gas. Usually the concentration range of CO₂ in sweet gas varies in the range of 0 to 50 parts per million, the levels of CO₂ in sour gas correspond to the 0 to 5% range and the concentration of CO₂ in acid gas can vary in the range of 0 to 50%.

TUNABLE DIODE LASER ABSORPTION SPECTROSCOPY FOR MEASURING CARBON DIOXIDE

Tunable diode laser absorption spectroscopy (TDLAS) is a non-contact analysis technique with long-term stability, high specificity and selectivity. Laser-based CO₂ sensors offer the advantage of faster response time, large dynamic range and low drift in comparison with conventional techniques such as gas chromatography (GC). In applications like monitoring CO₂ in LNG processing plants, these attributes help meet requirements and optimize plant operations.

The AMETEK 5100HD is an extractive-type CO₂ analyzer designed for hot/wet sample analysis. There is no sample conditioning for the analyzer system, just fully integrated sample handling to transport the sample. The 5100HD uses a sealed reference cell for continuous on-line analyzer verification and offers high specificity and sensitivity. The analyzer uses a digital implementation of wavelength modulation spectroscopy (WMS), so changing the experimental protocol is simply a matter of uploading a file. Ethane (C₂H₆) has spectral interference with CO₂ near the CO₂ lines selected for measurements. Depending on the concentration of C₂H₆ in the natural gas measurements of CO₂, the concentration range 0 to 50 ppmv can be measured in near-infrared (IR) range (for concentration of C₂H₆ less than 7%) or in IR range (for concentration of C₂H₆ higher than 7%).

MEASUREMENT RESULTS

Measurement of CO₂ in the NIR range was performed with a distributed feedback laser (Figure 2). The output of the laser was coupled into single mode optical fiber which was connected to a fiberoptic beam splitter. The splitter was used to divide the optical power in a 50/50 ratio for use in the sample and reference measurements, respectively. Gradient refractive index (GRIN) lenses were used to collimate the output of the single-mode fibers and direct the resulting beams through the sample and reference cells. The sample and reference cells each contained in gas-photodiode detectors, which were connected to separate input channels of the electronics unit. With this configuration, it was possible to make simultaneous measurements of unknown samples and known references, which were used to lock the output wavelengths for the laser. It should be noted that configuration of the sample cell compartment, included two cells for the measurement of CO₂ in two different streams (measurement of the inlet and outlet of the acid removal unit is also available). The optical path of the sample cell could be adjusted, corresponding to concentration range of measured CO₂ including multi-pass Herriott cells used for low concentration range measurements.

Measurements in the IR range were organized in a similar optical set up (Figure 3). An interband cascade laser was

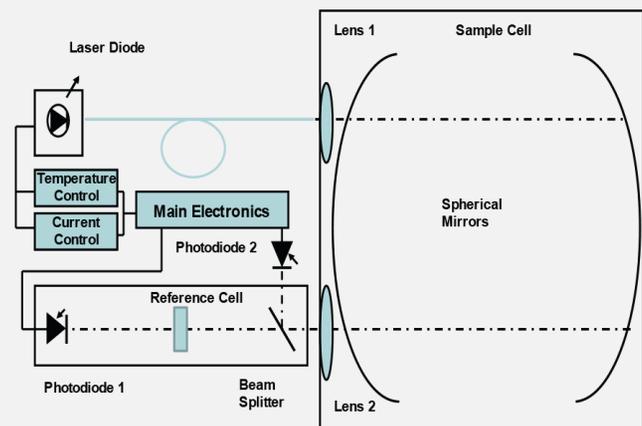


Figure 2. 5100HD analyzer optical setup for low CO₂ levels measurement in the NIR range

used as a radiation source. The beam was collimated and sent to the sample cell with an optical path of 10 cm. Indium arsenide antimonide photovoltaic detectors were used in sample and reference channels. As in the previous example, electronics provided control of the injection current and temperature of the laser.

The data shown in Figure 4 represents the response of the instrument to a series of CO₂ challenges in the concentration range of 0 to 50 ppmv. The duration of each of the challenges was from 10 to 20 minutes with return to the zero gas baseline, which was represented by CH₄ between challenges. The speed of the response T90 time was 100 seconds and was determined by the propagation of the gas in the sampling system with a flow rate of 1L/min. The data acquisition rate was 2 seconds/measurement. Repeatability of the CO₂ readings on each of the challenges was about 0.5 ppmv of the CO₂ concentration. The value of the accuracy evaluated at the levels of CO₂ from 0 to 50 ppmv was 1.5 ppmv.

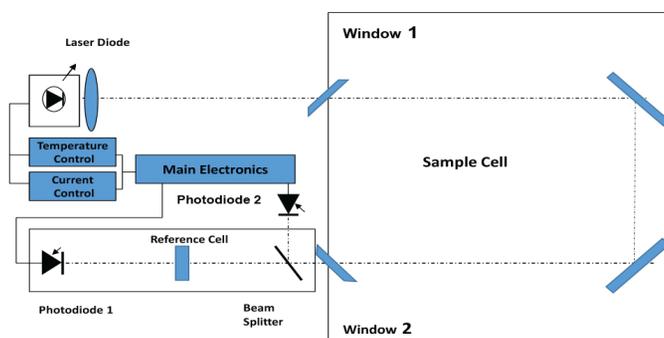


Figure 3. 5100HD analyzer optical setup for low CO₂ levels measurements in the IR range

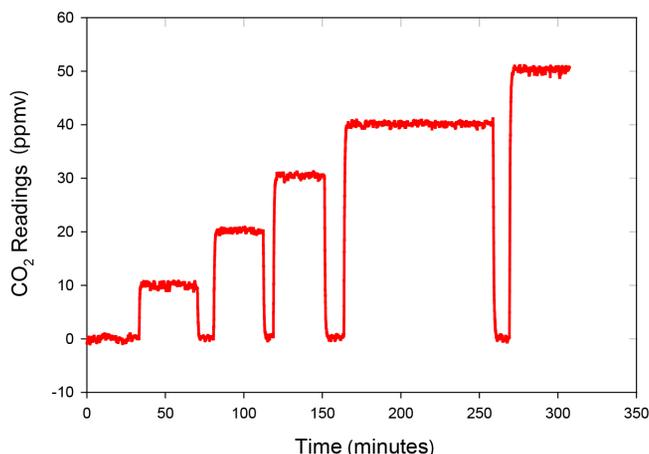


Figure 4. Response of the 5100HD analyzer to a series of CO₂ challenges

SUMMARY

To provide accurate measurements of CO₂ in natural gas it is important to account not only for CH₄, but also other components of the natural gas stream – especially C₂H₆ – and their effect on CO₂ readings. These components were accounted for by including them in the corresponding calibration model based on multivariate regression.

The advantage of using TDLAS-based technology for CO₂ measurements in the natural gas stream, compared with GC is speed of response and low maintenance value. Additionally, TDLAS provides higher accuracy and selectivity of measurements in comparison with NIR and IR photometry.

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