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Abstract

Isotope-Selective Gas Sensing Using Photoacoustic Non-Dispersive Spectroscopy †

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Abstract: The flow of carbons into the citric acid cycle can be readily traced by supplementation with ^{13}C stable isotope labelled nutrients. However, the quantification of the amount of fully oxidised nutrients to carbon dioxide is a challenging task. This contribution presents an isotope-selective, miniaturized gas detection scheme based on indirect photoacoustic spectroscopy. The results show that low-cost, continuous, in situ monitoring of the isotope ratio in gaseous samples is feasible.

Keywords: photoacoustic; isotope-selective gas sensing; breath analysis

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1. Introduction

The oxidation of high-energy carbohydrates is the dominant energy source of the vast majority of heterotrophic organisms, including all animals. The main compound classes donating their reduced carbon bonds are lipids, sugars, amino acids and, to a lesser extend, nucleotides, all of which converge at the so-called citric acid cycle (TCA, also known as the Krebs cycle), where the full oxidation to carbon dioxide (CO_2) takes place. Interestingly, the TCA cycle can also serve as a biosynthetic platform, where carbons from one class of metabolites are shunted into the biosynthesis of another. The utilisation in energy production or biosynthesis is a very dynamic process that integrates the metabolic needs of the cell with the availability of nutrients and oxygen [1,2]. While the flow of carbons into the TCA cycle can be readily traced by supplementation with ^{13}C stable isotope-labelled nutrients, and their detection via mass spectrometric analysis, the quantification of the extent to which these nutrients are fully oxidised to CO_2 poses a significant challenge due to its gaseous nature. Currently, this is mainly done by indirect methods such as the detection of radioactive ^{14}C from labelled compounds or use of a relatively expensive setup to detect $^{13}CO_2$ [3]. There are currently no established methods to accurately detect and quantify nutrient respiration via $^{13}CO_2$ detection in systems relevant for routine lab work such as tissue culture.

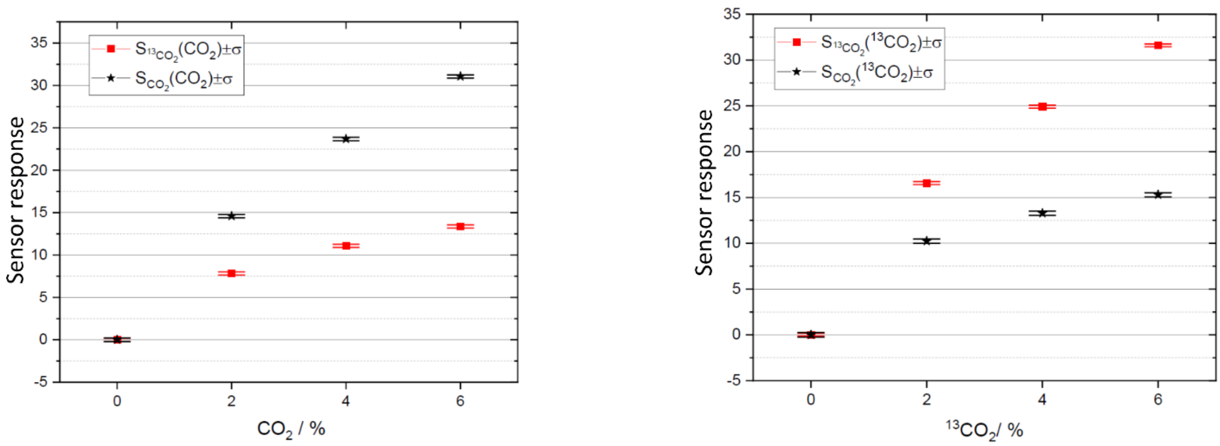
However, photoacoustic-based, non-dispersive infrared spectroscopy (NDIR) has been demonstrated to enable highly selective CO_2 sensors at a much-reduced system size due to its better sensitivity as compared to standard NDIR system [4]. In this contribution, the scheme is expanded to demonstrate isotope-selective detection of $^{12}CO_2$ and $^{13}CO_2$, thus paving the way for low-cost, in situ systems for the direct determination of the isotopic signature $\delta^{13}C = \left(\frac{\left(\frac{^{13}C}{^{12}C}\right)sample}{\left(\frac{^{13}C}{^{12}C}\right)standard}\right) \times 1000.$

2. Materials and Methods

The detection scheme relies on a single, mid-infrared light emitting diode (LED) from Hamamatsu (L15895LED) with a central wavelength of 4.2 µm illuminating a detection path length of 5 mm and two hermetically sealed, miniaturized photoacoustic detectors that each include a MEMS microphone (Invensense ICS-40720) and a 500 µm thick sapphire window for optical access, which are filled at 1 bar pressure with 100% CO_2 using standard isotope ratio of the carbon atom $\left(\frac{^{13}C}{^{12}C}\right) standard$ and 100% $^{13}CO_2$, respectively. To characterize the system, varying concentrations of standard CO_2 and pure $^{13}CO_2$ have been mixed with synthetic air and the photoacoustic signal has been recoded.

3. Discussion

The sensor response of both system channels is shown in Figure 1 for varying CO_2 concentrations using both standard carbon isotope ratio and pure $^{13}CO_2$ as test gases. Both channels show a response to both CO_2 isotope mixtures, but with a pronounced difference in sensitivity. Since only $^{12}CO_2$ and $^{13}CO_2$ cause signals, the setup enables a straightforward determination of $\delta^{13}C$, suitable for many applications that require isotope selective gas sensing applications.



(**a**) (**b**)

Figure 1. The sensor response to varying CO_2 concentrations in dry synthetic air: (**a**) Using the standard isotope ratio of CO_2, the sensitivity of the standard CO_2 channel is considerably higher than for the $^{13}CO_2$ channel. (**b**) For the gas sensitive characterization, using pure $^{13}CO_2$ gas, the situation is reversed.

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