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Dielectric barrier discharge micro-plasma emission spectrometry for the detection of acetone in exhaled breath

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ABSTRACT

Acetone is a predominant volatile organic compound (VOC) in the exhaled breath and a promising biomarker for diabetes and ketoacidosis. A non-thermal micro-plasma generated in a planar dielectric barrier discharge (DBD) is used as a radiation source for the excitation of gaseous acetone followed by its quantification with optical emission spectrometry (OES). Gaseous acetone can be directly sampled, while liquid acetone is evaporated by heated tungsten coil and then introduced into the DBD micro-plasma by a helium carrier flow for performing optical emission and detection at a 519 nm emission line. In the present study, the exhaled breath is collected and transferred into aqueous medium for sampling. With a sampling volume of 7 μL in a micro-drop, a linear range of 40–1600 mg L^{-1} is obtained along with a detection limit of 44 ng and a precision of 5.7% RSD. The present system is successfully applied to the determination of breath acetone for both diabetic patients and healthy volunteers.

1. Introduction

Acetone is the most abundant volatile organic compound (VOC) in exhaled breath and a known metabolic product of lipolysis [1,2]. Liver consumes fat deposits which are metabolized into acetone and other ketone bodies as an additional energy source in the absence of glycogen stores. The acetone generated travels through the blood and excretes from human body via lungs (as exhaled breath) or urine. Acetone level in the human body is generally low, while it might be high in the cases of starving, intense exercising, or glycometabolism disorders (including diabetes mellitus) which increases fat metabolism [3,4].

Diabetes is a prevalent chronic metabolic disease worldwide. Patients suffer from diabetes always need frequent blood glucose testing which is intrusive and painful. Breath analysis is a potential tool for the non-invasive monitoring of metabolic condition that might include diabetes mellitus [3]. Due to its association with diabetes and diet, acetone has gained considerable attention as a particularly attractive molecule to study in breath analysis [4]. Ever since the studies on breath acetone started in the 1950s, various methods have been developed for the accurate measurement of acetone. Among them, gas chromatography–mass spectrometry [5–8] and selected ion flow tube mass spectrometry [9,10] are most frequently employed for the accurate measurement of acetone in exhaled breath. Laser spectroscopy has recently been applied for the real time monitoring of breath acetone [2,11–14]. However, most of the currently available methods are expensive and bulky, restricting their uses in community hospitals or outpatient department as real time diagnostic tools. In this respect, it is required to develop portable and affordable acetone analyzers [15–18] for practical routine analysis.

Micro-plasma consumes very low power and generates limited heat; it can be operated in a very small space and is thus suitable for coupling with optical detectors to develop portable instrumentations for the detection of molecular fragments and elements. Among the various approaches for the generation of micro-plasma, dielectric barrier discharge (DBD), an old but recently renewed plasma technique, has earned extensive attentions, mainly due to its unique advantages of high dissociation/excitation/ionization capability, low temperature and atmospheric-pressure operation, ease of fabrication, and avoidance of electrodes contamination. Typical DBD plasma is formed between two parallel electrodes covered with dielectric barrier. With a high frequency AC power supply exceeding the breakdown voltage of the working gas between the two electrodes, a discharge is initiated to create high energy electron and various active radicals and ions. These active species in such an atmosphere facilitates fragmentation, excitation, and atomization of analytes or molecules. DBD has found applications as atomizer for atomic adsorption spectrometry and atomic fluorescence spectrometry [19–21], ionization source for ion mobility spectrometry and mass

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spectrometry [22–25], excitation source for optical emission spectrometry (OES) [26–32] and GC detectors for organic compound such as VOCs, halohydrocarbons, and amines [33–36]. The authors' group has long been devoting to the development of miniaturized instrumentations with DBD-OES systems for the determination of heavy metals [26,31], halogen compounds [29,30] and inorganic gases molecules [28]. In the present work, we exploit the optical emission characteristics of acetone in a DBD micro-plasma excitation source, and develop a portable DBD-OES device for the measurement of acetone in exhaled breath. Acetone in gaseous medium can be directly sampled, while that in liquid is evaporated by tungsten coil heating and then introduced into the DBD micro-plasma by a helium flow. The emission spectra of acetone with helium and argon as plasma working gas are carefully compared to eliminate potential spectral interferences as well as to reach highest sensitivity. In addition, the various experimental parameters have been scrutinized for enhancing the analytical performance of the DBD-OES system for acetone detection in exhaled breath.

2. Experimental

2.1. Apparatus and the experimental setup

Fig. 1A shows the schematic diagram of the DBD-OES system for acetone detection. The system is divided into three parts: sample introduction unit (a), DBD-OES emission unit (b) and detection unit (c).

2 Pieces of ceramic plates and 2 pieces of quartz plates are bond to form a DBD chamber with a dimension of (3 mm $H \times$ 4mm $W \times$ 30 mm L) as illustrated in Fig. 1B. The ceramic plates serve as a dielectric barrier to protect the electrodes from being contaminated and to eliminate the ambient spectral interferences, whereas the quartz plates are employed as observation window at which optical fiber of a CCD spectrometer is placed. Two pieces of copper electrode (4 mm × 30 mm) are attached onto the outer surface of the two ceramic plates by heat-resistant epoxy. With appropriate high-frequency and high-voltage electric field applied to the two copper electrodes by a neon powder supply (NG-B206JL, Jinshi Electronic Equipment Supply Company Ltd., Nanjing, China) the DBD micro-plasma is ignited and serves as an excitation/radiation source. The neon power supply is controlled by a touch regulator (TDGC2-0.5 kVA, Hongbao Electric Co., Ltd., Wenzhou, China) in order to regulate the output voltage for micro-plasma generation. The acetone vapor is introduced by a helium carrier flow into the DBD micro-plasma chamber, where acetone is excited and the emission spectra are recorded.

A QE65000 CCD spectrometer (Ocean Optics, USA) combined with a fiber-optic probe (20 cm length) with a core diameter of 1 mm is placed outside the quartz window for recording the optical emission spectra from 200 to 980 nm, which is furnished with a 50-μm slit and a 300 lines/mm grating. An integration time of 300 ms and an average of 3 scans for the CCD spectrometer are employed. The distance between the fiber-optic probe and the quartz window is set to be 5 mm without employing any optical

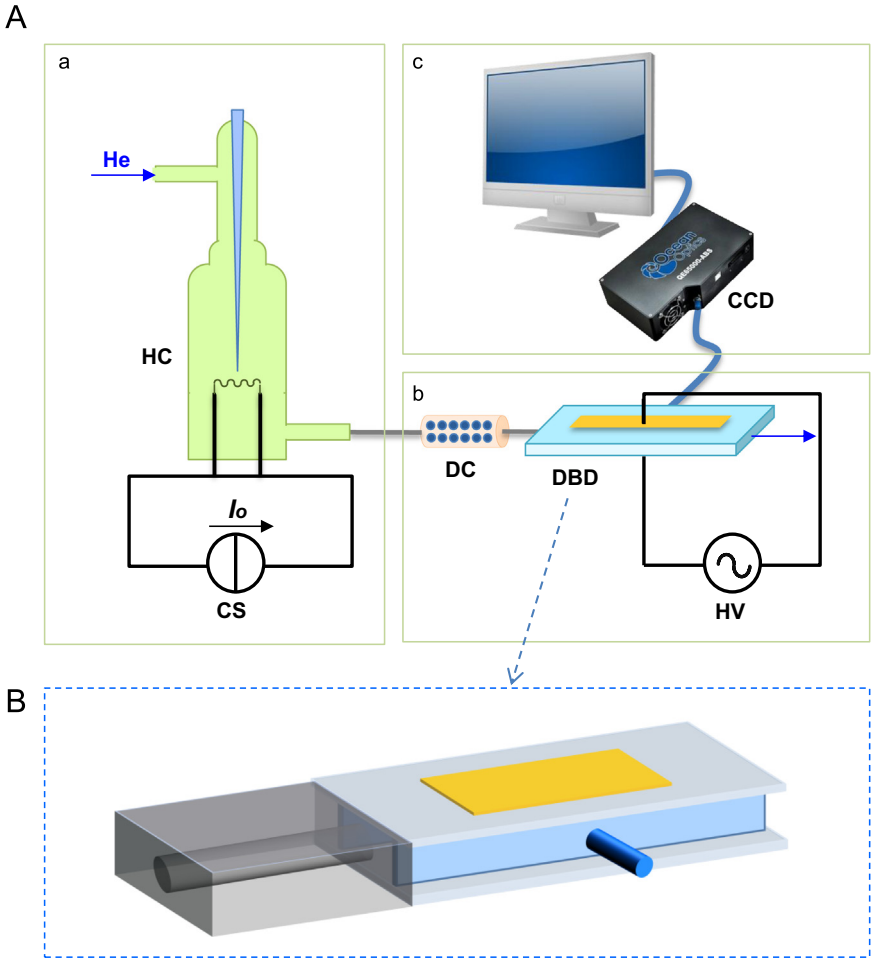

Fig. 1. (A) Schematic diagram of the DBD-OES system for acetone detection. (a) Sample introduction unit; (b) DBD-OES emission unit; and (c) Detection unit. HC: heating chamber; DC: drying column; CS: power supply; HV: high voltage. (B) Closeup of the gas inlet and planar DBD chamber.

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