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Micro-distillation system for formaldehyde concentration detection

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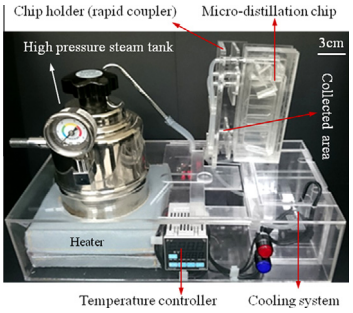
HIGHLIGHTS

- A micro-scale distillation system for distilled formaldehyde is proposed.
- Formaldehyde distillation efficiency as high as 98% at 4 bar steam pressure.
- Formaldehyde concentrations of 18 commercial Chinese herb samples are measured.
- System detection limit is around 0.5 ppm and deviation is less than 3.8%.

GRAPHICAL ABSTRACT



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ABSTRACT

A micro-distillation system is presented for formaldehyde concentration detection in food products. The proposed system consists of a micro-distillation device, a micro-condensed device, micro-distillation chip, a high-pressure steam tank, a temperature controller, and a cooling system. The distillation chip is fabricated on polymethylmethacrylate (PMMA) substrates using a CO_2 laser system and comprises three distilling chambers, a condenser micro-channel and a cooling micro-channel. In the proposed system, the formaldehyde sample is injected into the distillation chip and a high-pressure steam supply is used to vaporize the formaldehyde and carry it to the condenser region of the chip. The formaldehyde vapor is condensed under the effects of the low-temperature cooling channel and is transported to the collection region of the device. Finally, a colorimetric detection process is performed to measure the formaldehyde concentration of the collected sample using a commercial spectrophotometer. The validity of the proposed system is investigated using a control sample with a formaldehyde concentration of 6 ppm. It is shown that a distillation efficiency as high as 98% can be obtained given a steam pressure of 4 bar. The real-world applicability of the proposed micro-distillation system is demonstrated by measuring the formaldehyde concentrations of 18 commercial Chinese herb samples.

1. Introduction

Formaldehyde (CH_2O) is naturally produced in the human body during the metabolism process and can also be found in the air, natural food, some skin care products, and certain food preservatives. It is also widely used in construction, wood processing, furniture manufacture, textiles, carpeting, and the chemical industry. However, formaldehyde is associated with various medical prob-

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lems, including headaches, abdominal pain, breathing difficulties, vomiting, nasopharyngeal cancer and even leukemia. Moreover, formaldehyde has adverse effects on the urinary and central nervous systems of the human body, and can even cause death if ingested in excessive quantities [1]. Consequently, many countries prohibit the use of formaldehyde as an additive in food. To enforce this requirement, accurate methods for determining the formaldehyde concentration are required.

The concentration of formaldehyde in air is most commonly measured using spectrophotometry [2]. However, many other methods are also available, including colorimetry [3], fluorimetry [4], high-performance liquid chromatography (HPLC) [5], kinetic methods [6], gas chromatography (GC) [7], infrared detection [8,9] and gas detector tubes [10]. Many of these methods require the distillation and formation of a derivative for detection purposes. However, the distillation process requires the use of bulky, specialist equipment, and hence the formaldehyde detection process must usually be performed in a traditional laboratory environment. In practice, however, it is often desirable to perform formaldehyde detection in a more local, self-administered manner. Accordingly, this study proposes a miniaturized system for formaldehyde distillation designed to facilitate a cheaper and more convenient formaldehyde detection process.

Advances in microfluidics technology have revolutionized many biochemical and biomedical applications, including food safety inspection, quality control, environmental monitoring, and drug discovery [11–18]. Microfluidic structures typically incorporate micropneumatic systems, i.e., microsystems for the off-chip handling of the fluids (e.g., liquid pumps, gas valves, and so on), and microfluidic structures for the on-chip handling of micro and nanoliter volumes. Notably, compared to their macroscale counterparts, microfluidic systems can be easily produced in low-cost plastics and both increase the system throughput and sensitivity and reduce the sample/reagent consumption. Previous studies have demonstrated the use of microfluidic systems for the production of various biofunctionalized nanoparticles, including quantum dots, metallic nanoparticles, and other industrially-relevant materials (e.g., polymer particles) [19–27]. In addition, many researchers have presented integrated microfluidic platforms capable of performing such diverse functions as sample pre-treatment, sample preparation, polymerase chain reaction (PCR), species mixing, micro pumping, microfluidic rectification, fluid metering, droplet formation, filtering, separation and detection [28–37].

Distillation is an essential step in the downstream processing of many chemical processes. For micro-scale distillation systems, the smaller column dimensions cause the significance of gravitation to reduce in lieu of surface forces. Thus, novel column designs and packing materials are required to increase the surface forces so as to realize micron scale distillation. However, the high surface temperature of the column, combined with a high surface-to-volume ratio, leads to significant heat losses. Consequently, achieving a steady distillation performance requires a careful evaluation of the heat balance and the use of accurate heating control systems.

The literature contains many micro-scale distillation devices for liquid mixture separation and/or purification and solvent exchange. One of the first such systems was that proposed by Seok and Hwang [38], consisting of a circular pipe with porous glass sinter placed on the inside walls of the column to facilitate liquid flow. The column was operated in a horizontal orientation with the ends heated and cooled, respectively, via the column walls in a manner similar to that of a heat pipe. The experimental results obtained using ethanol/water and methanol/water mixtures, respectively, showed that the system achieved a similar separation performance to that of a conventional column with a length nine times longer.

Sundberg et al. [39–42] presented a planar micro-distillation column based on a zero-gravity principle. The column was designed to operate in the horizontal orientation and was packed with metal foam to serve as a wick material. The separation performance for a mixture of n-hexane and cyclohexane was found to be comparable with that of Sulzer DX commercial laboratory packing. In further studies, the operation of the micro-distillation column was optimized by adjusting the reflux and boiling flow rates. The feasibility of the optimize column was demonstrated using two binary systems (methyl formate + methanol, n-hexane + cyclohexane) and one ternary system (2-methylbut-2-ene + ethanol + 2-ethoxy-2-methylbutane).

Ziogas et al. [43] developed a flat micro-distillation system consisting of a stacked arrangement of micro-sieves designed to enhance the gas-liquid interaction. The separation performance of the proposed system was evaluated for various binary mixtures. It was shown that the optimal separation performance was obtained using a maximum of 12 separation stages. Hartman et al. [44,45] proposed a single-stage micro-distillation system based on a membrane micro-separator and a sweeping gas. In the proposed device, the sample mixture and sweeping gas were introduced into the micro-channel in the form of a slug flow and distillation occurred at a temperature lower than the boiling points of the individual mixture components as a result of the difference between their partial vapor pressures.

Zhang et al. [46,47] presented a microfluidic distillation chip consisting of two micro-channels (one for the liquid and one for the vapor phase) separated by a hydrophobic membrane. In performing the distillation process, condensation and evaporation were induced within the distillation unit by imposing a temperature gradient along the channels using cool water pumped through an additional, adjacent channel. The experimental results for a methanol–water solution showed that a theoretical plate number of up to 1.8 could be obtained under optimal operating conditions. Lam et al. [48–51] examined the separation performance of micro-distillation chips with various channel configurations using mixtures of acetone–water and methanol–toluene, respectively. For each device, the separation performance was evaluated for different temperatures of the heating and cooling regions and different compositions and flow rates of the liquid mixture. The optimal separation performance was found to occur under a higher heating temperature, a lower cooling temperature, and a zigzag-channel structure.

This study presents a PMMA-based micro-distillation chip for formaldehyde detection purposes. The chip is fabricated using a CO_2 laser system and comprises a micro-distillation device and a micro-condenser device. In the proposed chip, a mixed formaldehyde solution is injected into the first distilling region and is then driven through two further distilling regions by a high-pressure steam flow. The resulting formaldehyde vapor flows into the micro-condenser device and is cooled to a temperature lower than the vapor point under the effects of a cooling water flow. Finally, the condensed formaldehyde solution passes through a collection tube into a collection tube. Following a colorimetric reaction with a mixed indicator, the formaldehyde concentration is deduced from the absorbance measurement obtained using a commercial spectrophotometer.

2. Fabrication and experimental details

The proposed micro-distillation chip was designed using commercial AutoCAD (2012) software. The distilling chambers, condenser micro-channel and cooling micro-channel were patterned on PMMA substrates using a CO_2 laser ablation system [52–54]. As shown in Fig. 1(a), the micro-distillation device had overall