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A new 3D printed radial flow-cell for chemiluminescence detection: Application in ion chromatographic determination of hydrogen peroxide in urine and coffee extracts

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HIGHLIGHTS

- Design and 3D fabrication of a new radial flow-cell.
- Computational fluid dynamic simulation of the flow behaviour in the developed flow-cell.
- Evaluation of the developed flow-cell for chemiluminescence detection in both FIA CLD and IC-CLD based assays.
- Development of a new IC-CLD assay for the urinary and coffee extract H_2O_2 determination.

GRAPHICAL ABSTRACT

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ABSTRACT

A new polymer flow-cell for chemiluminescence detection (CLD) has been designed and developed by diverging multiple linear channels from a common centre port in a radial arrangement. The fabrication of radial flow-cell by 3D PolyJet printing and fused deposition modeling (FDM) has been evaluated, and compared with a similarly prepared spiral flow-cell design commonly used in chemiluminescence detectors. The radial flow-cell required only 10 h of post-PolyJet print processing time as compared to ca. 360 h long post-PolyJet print processing time required for the spiral flow-cell. Using flow injection analysis, the PolyJet 3D printed radial flow-cell provided an increase in both the signal magnitude and duration, with an average increase in the peak height of 63% and 58%, peak area of 89% and 90%, and peak base width of 41% and 42%, as compared to a coiled-tubing spiral flow-cell and the PolyJet 3D printed spiral flow-cell, respectively. Computational fluid dynamic (CFD) simulations were applied to understand the origin of the higher CLD signal obtained with the radial flow-cell design, indicating higher spatial coverage near the inlet and lower linear velocities in the radial flow-cell. The developed PolyJet 3D printed radial flow-cell was applied in a new ion chromatography chemiluminescence based assay for the detection of H_2O_2 in urine and coffee extracts.

1. Introduction

Chemiluminescence detection (CLD) is a potential option for the sensitive determination of solutes which do not possess a strong

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Abbreviations

IC	Ion chromatography
CLD	Chemiluminescence detection
PMT	Photomultiplier tube
CFD	Computational fluid dynamics
IC-CLD	Ion chromatography coupled chemiluminescence detection
FDM	Fused deposition modeling
RANS	Reynolds-averaged Navier–Stokes (RANS)
SST	Shear stress transport
FOX	Ferrous oxidation-xylenol orange

chromophore or fluorophore, which has been used for various applications including clinical, agricultural, to industrial analysis [1–3]. CLD systems have the advantage of requiring relatively simple instrumentation and can offer extremely high sensitivity for certain solutes. A CLD system essentially consists of only two components, (1) a transparent reaction vessel or a flow-cell and (2) a photodetector. The design of CLD flow-cell defines the sensitivity and reproducibility of the detector, as it influences fluid mixing, band dispersion, the amount of emitted light transmitted to the detector, and consequentially the signal magnitude and duration [4]. A flow-cell design which provides these signal enhancements also enables detector miniaturisation by enabling the use of low-cost digital imaging detectors, as compared to expensive high sensitivity photomultiplier tubes.

Usually, CLD flow-cells are produced by simply coiling polymeric or glass tubing in a plane [4–6] or by milling/etching channels into polymeric materials [7–10]. Coiled-tubing based flow-cells have been widely used for CLD in flow injection analysis (FIA) manifolds [11–14]. However, these simple approaches have some disadvantages, including the rigid nature of most suitable tubing, making the formation of the flat spiral cell rather difficult and irreproducible [15]. Greater design flexibility and complexity can be achieved with the use of milling or etching techniques, with these techniques also providing greater fabrication reproducibility, and access to a wider range of materials. However, they have some notable limitations, including limited resolution of closely spaced channels, and inability to produce complex 3D channel geometries. Such techniques are also not able to produce sealed channels, and thus are rather laborious and time consuming, due to the multiple steps required for the production of the sealed device.

However, these limitations can potentially be overcome with the use of 3D printing techniques, which can provide rapid and simple production of complex CLD flow-cells in a variety of materials. With the continual development of higher resolution 3D printers allowing multi-material printing, these capabilities are expanding rapidly. In terms of the advantages over other fabrication methods, 3D printing offers (1) the ability to print complex three-dimensional architectures, (2) low cost and time efficient production, (3) minimum wastage of material, (4) a "fail fast and often" [16] approach to prototyping, customisation, and testing, and (5) fabrication of monolithically integrated systems. Accordingly, 3D printing is rapidly becoming a method of choice for both research and industrial fabrication of polymeric and metal based macro- and micro-fluidic devices [17–19]. Use of 3D printing in the production of CLD flow-cells has been recently investigated by Spilstead et al. [20]. However, in this preliminary work, due to the tortuous nature of the spiral flow-cell design investigated, the 3D printing process resulted in only partially cleared (of support material) internal channels [20]. This resulted in significant flow-cell staining, which was presumed to be due to the formation of Mn(IV) on the remaining wax support material in the channels. Accordingly, to obtain the support material free channels, they had to print incomplete channels, and later seal them with transparent films [20]. This obviously negated one of the core advantages of 3D printing and illustrated unsuitability of tortuous flow-cell designs in allowing 3D printing fabrication of analytical flow-cells.

Many varied CLD flow-cell designs have been reported to-date, and the following represents some of the key designs investigated/developed: (1) the most commonly used spirally coiled tubing based flow-cell by Rule et al. [6]; (2) the fountain flow-cell design by Scudder et al. [21], where fluid radially flows between two parallel plates without any channels; (3) the sandwich flow-cell by Pavón et al. [22], which is a membrane based flow-cell; (4) liquid core waveguide based luminescence detectors by Dasgupta et al. [23], which utilise fluoropolymer tubing; (5) the bundle flow-cell by Campíns-Falcó et al. [24], which is based on the random packing of a tube; (6) the vortex flow-cell by Ibañez-Garcia et al. [25], which consists of a micromixer based on a vortex structure; (7) the serpentine flow-cell by Terry et al. [10], which consists of reversing turns, and finally (8) the droplet flow-cell by Wen et al. [26], which is based on the formation of a small droplet in front of the photodetector.

Many of the above mentioned flow-cell designs, including the spiral, serpentine, and bundle flow-cells, exhibit complex and tortuous geometries, which would present similar difficulties in terms of 3D printing based fabrication as those discussed above [20]. Whereas, simpler flow-cell designs, such as the fountain flow-cell has resulted in inferior CLD performance with a lower signal intensity and a poor signal reproducibility [10]. These issues suggest the need for a new CLD flow-cell design, which is less tortuous than the conventional flow-cells, enabling 3D printing, while still providing a reproducibly response, ideally of higher signal magnitude and duration to the above alternative designs. Thus herein, a new flow-cell has been designed, developed, and evaluated in comparison with the most commonly used spiral flow-cell design for CLD. The new flow-cell has been designed by diverging multiple linear channels from a common centre port in a radial arrangement and hence named as a 'radial' flow-cell. This radial flow-cell has been produced using both 'PolyJet' and fused deposition modeling (FDM) 3D printing techniques. It has been evaluated and compared quantitatively to a similarly proportioned spiral flow-cell design on the basis of (1) simplicity of fabrication with the 3D PolyJet printing and the FDM printing techniques and (2) CLD performance using the cobalt catalysed reaction of H_2O_2 with luminol as the model system. The flow behaviour in the radial flow-cell and spiral flow-cell designs have been simulated through computational fluid dynamic (CFD) calculations to understand the underlying mechanism for the observed differences in the CLD signals obtained. Finally, to investigate the practical application of the developed radial flow-cell, it was evaluated within an ion chromatographic based assay for the analysis of H_2O_2 in urine and coffee extract.

2. Materials and methods

2.1. Materials

Luminol (Sigma-Aldrich, MO, USA), $CoCl_2$ (Univar, IL, USA), Na_3PO_4 $7H_2O$ (Mallinckrodt, Surrey, UK), NaOH (BDH, PA, USA), H_2O_2 (Chem-Supply Pty Ltd, South Australia, Australia), 5-sulphosalicylic acid (Sigma-Aldrich, MO, USA), ferrous ammonium sulphate ($FeSO_4(NH_4)_2SO_4 \cdot 6H_2O$) (England, UK), H_2SO_4 (Merck, VIC, Australia), xylenol orange (Sigma-Aldrich, MO, USA), Sorbitol (BDH, PA, USA), 0.45 μM PTFE captiva syringe filters (Agilent, CA,

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