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HYBRID TECHNOLOGY (3D ADDITIVE PRINTING – SILICON – GLASS) MULTILINE EVAPORATIVE CONCENTRATOR FOR WATER QUALITY MONITORING SYSTEM

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ABSTRACT

This paper reports the design, fabrication procedure and functional tests of a continuous-flow hybrid technology 16-line evaporative concentrator. The unique and complete integration of the microscale silicon-glass evaporator with a mesoscale 3D printed polymer manifold platform is successfully demonstrated. The hardware platform of concentrator control unit is described.

The continuous-flow concentrator chips were tested with respect to their evaporation capability. A very high water evaporation rates were achieved in evaporative concentrator. An evaporation rate of 10 µL/min, achieved in mild conditions, compatible with biological sample treatment (T≤37°C), allowed for 4-fold sample enrichment. High-throughput flow ability and 33-fold concentration factor achieved in temperature of 50°C facilitates the application of prototype for chemical sample enrichment.

KEYWORDS

Microevaporator, evaporative concentration, forced convection, sample concentration, sample enrichment, 3D printing.

INTRODUCTION

The monitoring and assessing water quality is of vital interest for every society.

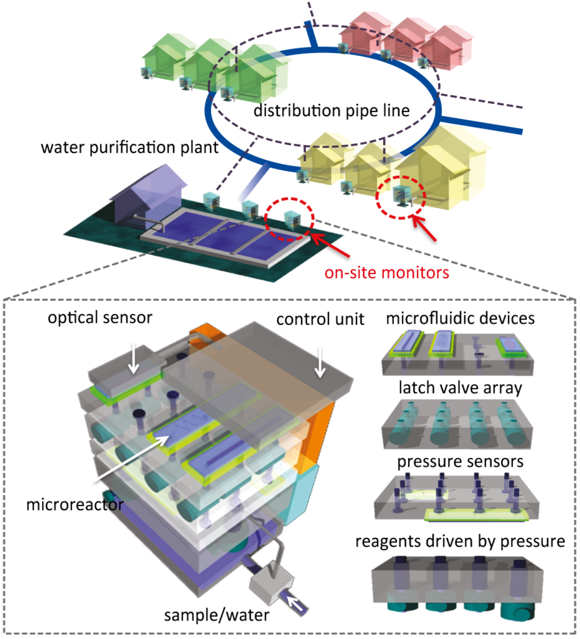

Figure 1: Schematic representation of a standalone water monitoring system accommodated within municipal water network grid.

Our current research focuses on development of a standalone, low-maintenance and sensitive detection system to monitor very small concentrations of chemical and biological contaminants in drinking water (Figure 1).

Our goal is to enhance the detection limit of such a system, therefore concentration/enrichment of the sample prior to the detection is required. We propose a novel approach to concentrate sample by evaporation of the solvent through micromachined perforated membrane in mild conditions compatible with the requirements of biological sample treatment. Another unique aspect of our method is related to the utilization of rapid prototyping technique (additive 3D printing) to encapsulate silicon-glass chip within polymer holder in semi-permanent manner.

The concept of evaporative concentration was proven by a single-line chip, as discussed in our previous report [1]. Briefly, the single-line device consists of a SiRN membrane (86.5 mm x 100 µm x 200 nm) perforated with ~86 500 micromachined via-holes (600 nm in radius), which forms the contact between a shallow liquid channel and an overlapping gas channel (Figure 2).

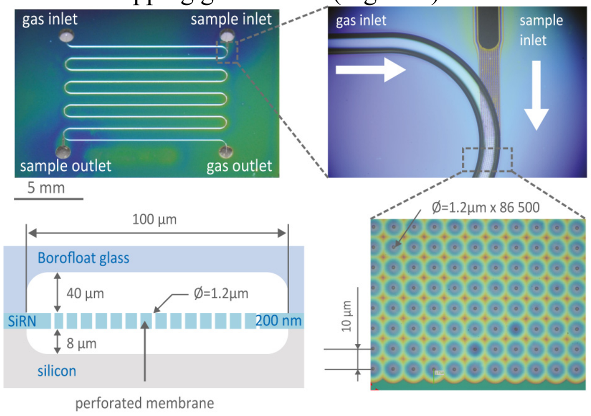

Figure 2: Photographs and schematic representation of cross-section of a single-line concentrator chip (not in scale).

The liquid evaporates through the grid of via-holes, and its evaporation rate is controlled by varying the gas flow over the membrane, thus by changing the efficiency of the gas layer renewal on the gas-liquid interface. The efficiency of the single line concentrator was evaluated by comparing the concentration of a marker solution prior and post to pumping through the chip via UV-Vis at 490 nm (Figure 3).

It was proven that it is possible to concentrate the sample using evaporation through perforated micromachined membrane and to have stable gas-liquid interface.

Nevertheless, the operational flow range of the single-line chip was too low to fulfill the requirements of 10-fold concentration of 1 mL of sample in less than 2 hours.

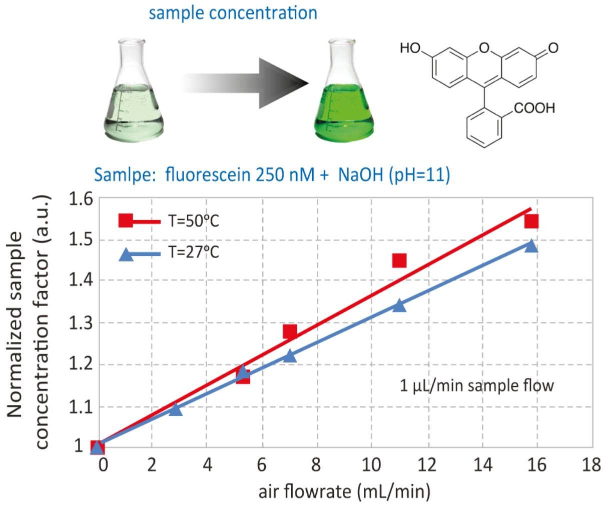

Figure 3: Normalized sample concentration factor as function of the gas volumetric flow rate. Plot based on [1].

In order to enhance the concentration capabilities of a single-line chip, the follow-up 16-line evaporator with dense via-hole grid (~1.85 million via-holes) was proposed.

FABRICATION

Silicon-glass chip

Two versions of the concentrator were designed. In the first one overlapping gas channels were etched in top glass layer encapsulating the perforated membrane. The second version has an open cavity in which the membrane is exposed to externally induced gas flow (Figure 4a).

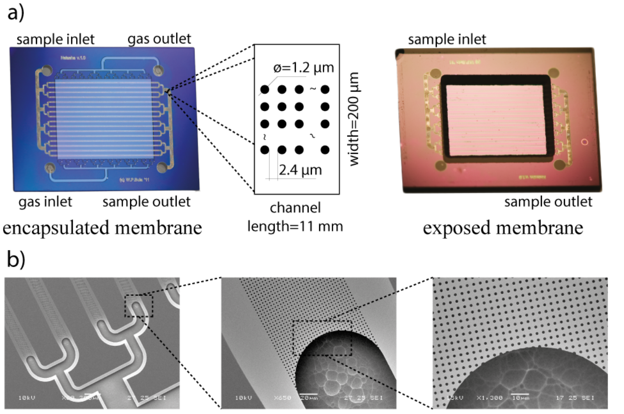

Figure 4: a) Photographs of a 16-line concentrator chip with encapsulated (left) and exposed (right) membrane. b) SEM image of perforated micromachined membrane.

Two etching processes were examined to form buried liquid channels in Si wafer through patterned SiRN membrane (300 nm in thickness): a gas phase isotropic etching in XeF_2, and a wet isotropic etching in HNO_3:HF:H_2O, T=20°C followed by wet anisotropic etching in KOH solution. While the second mentioned process facilitated formation of deep under-membrane channels (100-120 µm), the optimization of parameters of etching procedure was performed. The membrane was released by under-etching (Figure 4b).

The inlet and outlet via-holes were etched by deep reactive ion etching (BOSCH process) using a mask of SiRN. The silicon wafer was anodically bonded to Borofloat wafer with a powder-blasted cavity and diced into separate chips of 20 mm x 15 mm (Figure 5).

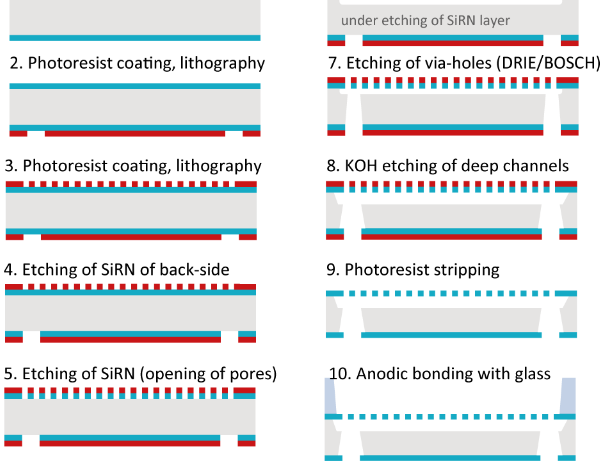

Figure 5: Fabrication process flow of silicon-glass 16-line evaporative concentrator (not in scale).

The top side of the membrane was then selectively hydrophobized by fluorocarbon-based passivation layer of DRIE. The contact angle was modified from 35° to 102°.

Silicon-glass-polymer hybrid module

3D additive printing technology was utilized to fabricate silicon-glass-polymer module.

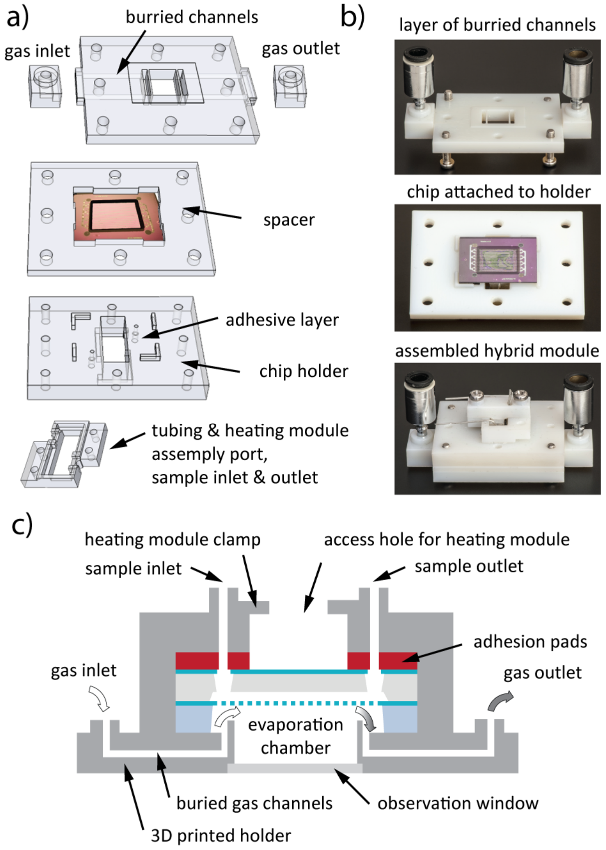

Figure 6: a) Schematic representation and b) photographs of the 16-line concentrator chip assembled with 3D printed elements. c) Cross-sectional view of hybrid module.

The additive 3D printer (Objet 24, Objet) was used for rapid prototyping of specialized chip holder, and gas/liquid ports. Object VeroWhitePlus^{TM} rapid prototyping polymer was used. The silicon-glass chips were semi-permanently assembled to gas channels buried inside 3D printed manifold by adhesive pads (Figure 6).

The adhesive pads were laser-cut (VLS3.50, Universal Laser Systems) to match the dimensions of microfluidic ports of silicon-glass evaporator.

Measurement setup

The evaporation rate was calculated by microcontroller based on the readouts of two liquid sensors (ASL1600-10, Sensirion) assembled to liquid inlet and outlet ports (Figure 7).

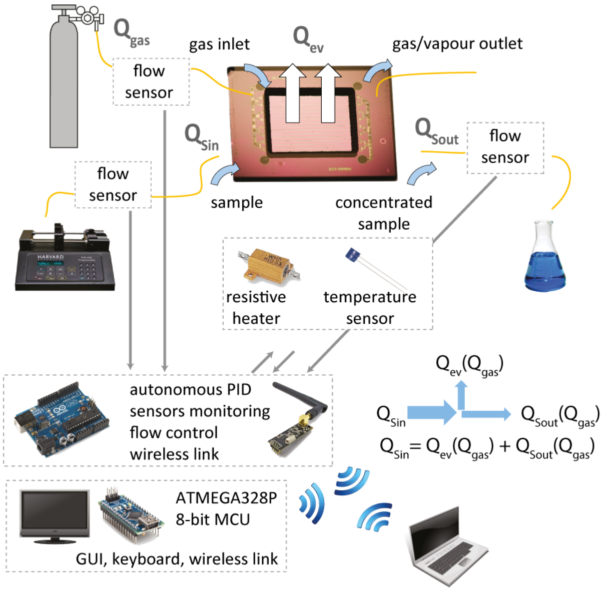

Figure 7: Schematic representation of measurement setup.

The temperature stabilization and monitoring of evaporation rate is provided by controller based on open-source Arduino electronics prototyping platform. The proportional-integral-derivative (PID) loop was software-implemented in 8-bit RISC microcontroller (AVR/ATMEGA328P, Atmel) to stabilize the temperature with variation of 0.1°C.

A resistive heater and a precision temperature sensor (LM35, National Instruments) were accommodated inside aluminum housing (12.5 mm x 10 mm x 5.3 mm) and brought into contact with the silicon surface of evaporator. In order to enhance the good thermal contact with the silicon-glass chip, thermally conductive glue (SCV-22, Sunhayato) was applied. Resistive heater is controlled by pulse-width modulation (PWM) and dissipates up to 3W of power elevating the temperature of the chip to 100°C.

The autonomous controller communicates with the external display unit via radio link established by single-chip 2.4-2.5 GHz ISM band transceivers (nRF24L01+, Nordic Semiconductor) and with any PC through Bluetooth module (WRL-10393). The bidirectional communication protocol with built-in error correction was designed and implemented.

RESULTS

Flow regimes and boundary conditions of gas-liquid flows were identified and determined. Preliminary experiments performed in 16-line concentrator with encapsulated membrane geometry, showed that the volumetric flow rate of the gas and its capacity for saturated vapour was not sufficient to establish satisfactory evaporation rates. Moreover, the high hydraulic resistance of gas lines led to unbalanced flow regimes resulting in unstable gas-liquid interface and flow through perforated membrane. In result, the chip family with closed gas chamber was discontinued and subsequent experiments were conducted by use of exposed membrane geometry chips.

Evaporation tests

Nitrogen was used as a drying gas. Stable gas-liquid interface was observed for various gas-to-liquid flow ratios. No flow of sample or gas through micromachined membrane was observed for the sample flow rate up to 150 µL/min. The membrane showed outstanding mechanical durability for stress induced by the pressure of gas and liquid sample and no fracture was observed during experiments.

The influence of the gas flow on the evaporation rate was investigated for various sample flow rates (Figure 8).

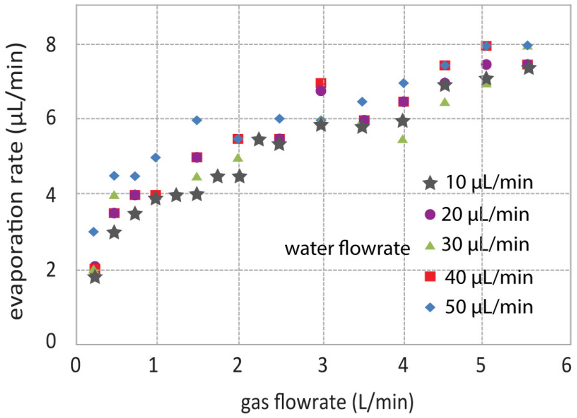

Figure 8: The influence of the gas volumetric flow rate on the evaporation rate of water for various sample flow rates (T_{gas}=25°C).

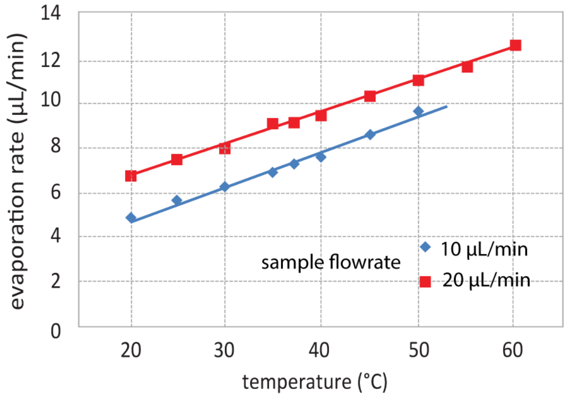

Figure 9: The influence of the temperature on the evaporation rate for various sample flow rates (Q_{gas}=2 L/min).

A linear relation between temperature and evaporation rate was observed for various sample flow rates (Figure 9).

High sample concentration factor (33-fold) was achieved for sample flow rate of 10 µL/min at 50°C, Q_{gas}=2 L/min (Figure 10).

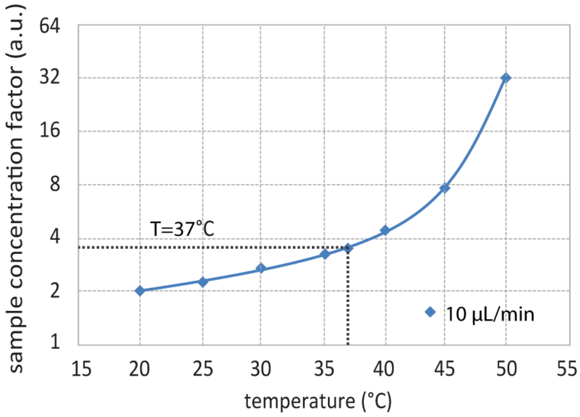

Figure 10: The influence of the temperature on the sample concentration factor for Q_{gas}=2 L/min and Q_{Sin}=10 µL/min.

By applying forced convection principle to chip with 16 parallel liquid channels, evaporation rates higher than 10 µL/min were achieved allowing for high sample enrichment in continuous-flow regime in temperature range compatible with biological samples.

To achieve even higher sample concentration and maintain high volumetric throughput of the sample, a new type of concentration setup with closed sample loop was proposed (Figure 11).

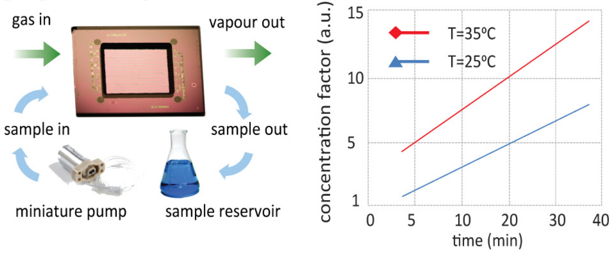

Figure 11: Schematic representation of closed sample loop setup (left) and estimated sample concentration factor as a function of concentration time (right).

In such system the temperature, gas and liquid flows are set constant, thus the concentration factor depends only on the time of pre-concentration.

The inert and robust design of the evaporative concentrator, together with the cleaning procedure ability, allowed the device to be operational for weeks of experiments.

CONCLUSIONS

In this paper we have presented a prototype of a micromachined evaporative sample concentrator based on forced convection. We have shown that this type of concentrator is capable to achieve high concentration factors in continuous-flow regime. Moreover, the efficient concentration process might be performed in the temperature range compatible with biological samples. We have briefly announced the hardware and software platform that was assembled to provide temperature stabilization and concentration monitoring. We have shown a successful integration of silicon-glass with rapid prototyping technology. The additive 3D printing was used to fabricate polymer module inside which the silicon-glass evaporator was semi-permanently encapsulated. The versatility of 3D printing technology allowed formation of buried gas channels inside polymer layer forming a mesoscale bridge between microfluidic domain of silicon-glass chip and macro scale fluidic equipment.

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