Polymer Lab-on-Chip systems with integrated electrochemical pumps suitable for large scale fabrication

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Abstract

A low-cost, polymer based microfluidic platform is described which not only includes passive microfluidic parts, but also pumps based on an on-chip electrochemical gas generation by electrolysis. A hydrogel is used as electrolyte material, which allows a simple fabrication process by screen printing or stencil printing. Test structures were designed and fabricated to illustrate the feasibility of the approach for batch processing. Microfluidic chips including reservoirs and channel structures were fabricated by micro injection moulding and used to demonstrate the movement of liquids inside micro channels by the proposed micro pumps.

Keywords: micro pump, electrolysis, hydrogel, polymer, microfluidics, lab on chip, point of care

1. Introduction

A new polymer-based Lab-on-Chip (LoC) platform for protein detection in Point-of-Care (PoC) applications is currently being developed, aiming to integrate active optical components as well as microfluidic actuators inside the polymer chip [1]. While the integration of additional functionality can improve the performance and/or usability of such a device significantly, cost per chip/analysis remains a critical factor. Thus, not only passive components like channels and reservoirs should be "low-cost" components, but also new functions like pumps or valves should be selected and developed according to cost consideration.

As the controlled transport of liquids like a sample or buffer solutions is essential for an integrated LoC for PoC use, on-chip microfluidic actuators are desired, which are small in size, inexpensive, easy to integrate but on the other hand able to generate large displacements. Classical approaches for micropumps which consist of a pump chamber, an oscillating membrane and passive or active valves are usually very complicated and difficult to integrate in a polymer platform. Despite of these "conventional" micro pumps, certain non-conventional pumping principles have been investigated and developed during the past years.

One type of "non-conventional" pumps are surfaced based actuators, which employ e.g. electroosmotic effects, electro-/thermo capillary effects or surface acoustic wave (SAW) effects. While all of these techniques are very attractive for certain applications, it is usually very difficult to flexibly apply them to a low-cost, polymeric, channel-based lab-on-chip system. Furthermore, these principles also strongly depend on certain properties of the moved liquids and/or the used substrate/wall material.

In contrast, applying a direct pressure to a liquid inside a channel seems to be a more "universal" approach. Also for direct pressure actuation, different non-conventional approaches have been investigated. For example, paraffin actuators have been proposed by Carlen and Mastrangelo [2], which make use of the large thermal expansion of wax paraffin when melting (phase change). Another type of very promising actuators uses expandable microspheres incorporated in an elastic matrix, and was first proposed by a group around G. Stemme [3]. Both of these actuation principles use heating to generate a pressure, which leads to a high power consumption. Furthermore, these actuators may not be suitable for temperature sensitive applications, like e.g. protein assays.

Another interesting approach, first demonstrated for micro actuators by C. Neagu et al. [4], is the electrochemical generation of gas bubbles by the electrolysis of water. Actuators based on electrolysis have a very low power consumption as well as almost no heat generation, while still being able to generate large deflections or displacements and high pressures.

1.1. Theory

The electrolysis of water is the dissociation of water molecules into oxygen and hydrogen gas by means of an electric voltage. The reaction is given by:

\[ \text{Anode: } 2\text{H}_2\text{O} \rightarrow 4\text{H}^+ + 4e^- + \text{O}_2 \text{ (gas)} \]
\[ \text{Cathode: } 2\text{H}_2\text{O} + 2e^- \rightarrow 2\text{OH}^- + \text{H}_2 \text{ (gas)} \]

The amount of generated gas molecules is directly proportional to the current flowing through the electrolytic cell. Using pure (de-ionized) water, this reaction is very slow. Increasing the ion content in the water increases the conductivity and thus the speed of the reaction for a given voltage.

The generated gas can be either used to deflect a membrane or to drive a fluid by directly applying the gas pressure to the fluid without an intermediate membrane. Bubble actuators based on electrolysis have already been successfully demonstrated for microfluidics. [5-7]. Within these works, a liquid (usually water or a water based liquid) had to be integrated in the electrolytic reservoirs. However, the integration of a liquid can be critical for mass production and may also cause problems concerning long-term stability.

A hydrogel based on a poly (acrylic acid) sodium salt (PAAS) was therefore investigated in this work for its applicability as electrolyte, which combines the
electrolytic and gas generating properties of water-based electrolytes with a good processibility by screen printing or stencil printing. It is available at very low cost and non-toxic.

Crosslinked PAAS is a so-called super absorbing polymer which is able to dramatically swell in water. The swollen state is referred to as a hydrogel, which mainly consists of up to 99 wt% of water, but has a very high viscosity. When PAAS is brought in contact with water, the weakly bonded sodium is split off, leaving negatively charged polymer chains and movable sodium ions:

\[
\begin{align*}
\text{COONa} | C \equiv C | H \equiv H & \xrightarrow{H_2O} \text{COO}^{-} | C \equiv C | H \equiv H + n \text{Na}^+ \\
\end{align*}
\]

Using this effect, PAAS not only "thickens" the water, but also increases its ion content, thus leading to a much faster electrolytic reaction at moderate voltages.

2. Design of test structures

Two types of test structures were designed and fabricated. The first one aimed to allow the development of the gel integration process and the investigation of the electrolytic behavior, the second one was intended to demonstrate the feasibility of the gel actuators as pumps in a Lab-on-Chip system.

2.1. Electrode test structures

A set of test electrodes was designed and arranged in a 4” wafer layout together with cavities which should be placed around the electrodes. These test structures were also used to measure membrane deflection e.g. for use as a valve. The performance of these electrode designs with and without membrane was already presented [8]. Here, special attention is being put to the feasibility of the approach for large scale fabrication by stencil printing, which allows to fill a large number of flat electrolytic reservoirs at the same time.

2.2. Channel test structures

A microfluidic system was designed according to the requirements of an integrated lab-on-chip system for point-of-care diagnostics [9]. The microfluidic layout incorporates three inlet reservoirs, one waste reservoir and a channel system which connects all these reservoirs and delivers the liquids to a sensor area.

In addition, each inlet reservoir is connected to the "pump region" (Fig. 1) by a thin hydrophobic channel ("gas delivering channel") of a cross section of 100x100 µm². The pump region mainly consists of the gel reservoirs in which the gas will be generated by electrolysis. The gel reservoirs can be covered by a thin membrane which should open automatically when a certain gas pressure within the gel reservoir is reached.

The working principle of the pump is explained in Fig. 1. The bottom picture reflects the "real" setup and layout of electrodes and channels, which has been also used in section 4.

3. Fabrication aspects

3.1 Batch process for gel integration

Gold electrodes were fabricated by sputter coating, standard lithography and wet etching on both 4 inch silicon wafers (with thermal oxide for isolation) and 4 inch, injection moulded, thermoplastic wafers made of a Cycloolefin-Copolymer (COC). Cavities around the electrodes were subsequently formed by SU8 lithography and have heights between 80 and 300µm. Fig. 2 shows a set of electrode test structures (see 2.1) fabricated on a COC substrate.

Fig. 1: Schematic drawings of the pump region of a lab on chip system with a gel pump. a) principle of pumping, b) cross section through gel reservoirs

Fig. 2: Diced COC chips on blue dicing tape with gold electrodes and SU-8 chambers

The SU8-chambers were filled with the PAAS gel - either manually or by stencil printing on wafer scale. The process is illustrated in Fig. 3. As stencil, a commercially fabricated 50µm thick stainless steel stencil was used. The openings were chosen to be slightly smaller than the dimensions of the SU8 chambers.
Fig. 3: Filling of an electrolysis chamber with gel by using stencil printing.

Fig. 4 shows a successful stencil printing batch process on a silicon wafer with SU8 chambers. As sealing of the gel by a thin membrane directly after printing the gel is desired, current investigations focus on membrane material selection and process integration of these materials.

3.2 Micro injection moulding of polymer parts

The layout described in 2.2 was transferred to a two-sided mould (Fig. 5a) by micro milling. The mould was then used in a “Battenfeld Microsystem 50” micro injection moulding machine to fabricate the passive microfluidic polymer parts (Fig. 5b).

4. Results

Injection moulded COC and PMMA channel structures (“Channel substrate” in Fig. 2) were drilled at the end of the “gas delivering channels” to provide through-holes to the actuator chamber. The so prepared plastic parts were then sealed on the top with an adhesive tape. A structured membrane layer was fabricated by PDMS casting. This membrane layer incorporates reservoirs with a thin lid, which have been subsequently filled with PAAS gel and placed between an electrode substrate and the channel substrate. After the sample reservoir was filled with inked water, all pieces were then clamped together, resulting a tight sealing. (Fig. 6)

First investigations were carried out with a simple 9V battery, illustrating that the approach is well suited for portable applications. Fig. 7 shows the emptying of the first of three reservoirs at different times.

The electrode configuration used for this test was chosen to lead to a rather high cell resistance and thus to a slow gas generation at the used voltage. It has to be noted that the driving speed can be easily increased by increasing the current flowing through the cell.
Interestingly, almost no fluid entered the channels back to the other reservoirs, even if they were already emptied. This is caused by two effects: The first reason is that the flow resistance was selected to be higher from the junction of the channels towards the inlet reservoirs than to the outlet reservoir. The second (even more important) reason is, that only the waste reservoir (outlet) has an opening, while the air/gas in the channel towards the inlet reservoirs would have had to be pressurized to allow a flow in this direction.

Fig.6: Emptying of a reservoir by electrolytic gas generation. The electrode configuration used for this test allowed only a large cell resistance and thus only low driving currents at moderate voltages of about 9V.

5. Conclusion

On-chip gas generation by electrolysis was successfully used to empty reservoirs and drive fluids through channels inside a polymer based microfluidic system. The water used for electrolysis was “stored” inside a super absorbing polymer and could therefore be easily integrated e.g. by stencil printing. Next steps will focus on complete integration of all components and the development of an in-situ integration of the gel-covering membrane directly after the stencil printing process.

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