AN ELECTROCHEMICAL-BASED THERMAL FLOW SENSOR

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ABSTRACT

The first thermal flow sensor to operate on the principle of measuring thermal changes in ionic mobility vis-à-vis electrochemical impedance resulting from flow was developed and demonstrated. Its biocompatible construction on a flexible polymer substrate enables unobtrusive integration with medical implants such as shunts and catheters. Flow is transduced by utilizing electrochemical impedance measurements between an electrode pair to detect temperature changes resulting from transport of liquid focally heated upstream. This approach offers increased sensitivity compared to solid sensing materials and compatibility in saline environments. Flow rates below 320 µL/min could be detected with a limit of detection of 10.7 µL/min. The simple design and performance range is suitable for *in vivo* applications.

INTRODUCTION

Thermal flow sensing is the most common method used in MEMS flow sensors, and involves measuring variations in heat transfer induced at different flow rates. Various thermal flow sensors have been developed in laboratory settings or implemented in commercial devices; these can be simply classified into three categories based on sensing modality. Hot-wire or hot-film sensors measure heat transfer from a heater to a fluid by indirectly monitoring the heater's temperature, calorimetric sensors measure the difference in temperature between an upstream and downstream sensing element which is imposed by a central heater, and time-of-flight sensors measure the transit time of a pulse of heat to a sensor located at a known distance downstream of a heater [1]. Each of these methods share the requirement of accurate measurement of the fluid's temperature.

The common strategy to measure temperature, both in MEMS devices and in large-scale electronic thermometers, is to fabricate sensors from materials whose electrical resistance varies in a predictable manner with temperature. All conductive materials exhibit such a temperature coefficient of resistance (TCR), and certain materials such as platinum have emerged as a popular choice due to their relatively large and predictable TCR over a large temperature range. Likewise, materials with a large TCR are necessary for thermal flow sensing, and the higher the TCR, the greater the sensitivity. Although TCRs up to 2% have been reported in the laboratory through the use of doped semiconductor junctions, most conductive materials exhibit a TCR of less than 0.5%/°C (Table 1). Despite platinum's wide use in commercial temperature sensors, its TCR is at best 0.392%/°C in bulk and between 0.15-0.25%/°C as a thin film [2].

Thermal flow sensors must be encapsulated to protect against corrosion during prolonged use in physiological conditions. The addition of a protective encapsulation layer limits sensor performance due to its thermally insulative properties, which result in increased power consumption and impose a detection delay associated with the transport of heat through the encapsulation to the sensor. MEMS thermal flow sensors are commonly fabricated on silicon, which is a good thermal conductor and provides alternative competing path for heat transfer from the heater to sensors. To overcome these limitations, free-standing sensors can be fabricated on thin film Parylene C substrates, which can act as both a biocompatible encapsulation layer and an electrically insulating substrate [3].

In order to overcome the inherent limitations of thermal flow sensors, other flow transduction methods have been developed. These include the electrochemical detection of the time-of-flight of a dissolved oxygen tracer [4], the change in ionic movement between electrodes parallel to flow [5], or detection of the time-of-flight of a microscopic bubble produced via electrolysis [6]. However, these approaches suffer from low sensitivity or surface tension issues which limit their performance and development.

Material	TCR (%/ºC)
Platinum	0.392
Copper	0.430
Gold	0.390
Polysilicon	-2.5 to 0.1
Germanium	2.0
5% NaOH Solution	-2.01
30% NaOH Soluion	-4.50
20% KCl Solution	-1.68
Human Cerebrospinal Fluid	-1.98

Table 1: Temperature coefficients of resistance (TCRs) for various solids and aqueous solutions. [1, 7-9]

CONCEPTS AND PRINCIPLES

Electrochemical Impedance

We introduce an alternate thermal flow sensing material and approach in which temperature-induced changes to the electrochemical impedance of a flowing electrolyte solution, instead of the electrical resistance of a solid sensor, are used to detect temperature changes and thereby deduce flow information. When an ionic solution is heated, the mobility of ions in solution increases, which results in a decrease in the solution's resistivity. The resulting change in resistivity per degree Celsius has been measured for several aqueous ionic solutions, including human cerebrospinal fluid, and the response is an order of magnitude higher than that of common temperature-sensitive solids [7-9].

The conductivity of an ionic solution can be measured via the solution impedance between a pair of electrodes. At

the appropriate measurement frequency, the impedance is dominated by the solution resistance, allowing direct measurement while bypassing changes associated with the electrode-electrolyte interface (*e.g.* capacitive components). While some impedance drift can occur over minutes or hours, it was observed that when the temperature of the fluid around the electrodes was altered, the resulting change in impedance, when taken as a percentage of the baseline impedance measured immediately before the temperature change, is consistent.

Flow Sensing Method

To measure flow rate, we utilized the time-of-flight principle which requires an upstream heater and downstream temperature sensor. Heat transfer between the heater and the temperature sensor is impacted by conduction and convection processes. Since the supporting substrate is Parylene, it is assumed that thermal conduction through the substrate is negligible. At low flow rates and heater temperatures, conduction via diffusion of the heat pulse in the fluid will dominate. Convection will alter the heating profile close to the heater.

The heater and temperature sensor are separated by 1 mm and exposed to flow. When the heater is activated, the temperature at the sensor rises to a constant level at a rate which varies predictably with the flow velocity; the maximum rate of change of temperature at the sensor is consistent for a given flow velocity and increases exponentially as the flow increases.

This technique is useful at low flow velocities at which thermal time-of-flight sensing using a solid detector would require prohibitively close placement to the heater, for limiting the fluid heating or the power used by the heater, or for situations in which using only one temperature sensor is desirable. Furthermore, bi-directional flow measurements are possible using only a single detector, but with a limited detection range in the backwards direction.

FABRICATION AND TESTING

Fabrication

Our sensor consists of a platinum resistive heater with an electrochemical impedance sensor placed 1 mm downstream (Fig. 1). The sensor is built on free-standing Parylene C, which is used as both a mechanical substrate and an encapsulation layer. Parylene C is a biocompatible polymer with a long track record of use as an encapsulant and substrate for implantable MEMS devices. The resistive heater is composed of a 25 μm wide serpentine platinum trace (resistance ~ 1400 Ω) and the impedance sensor is a pair of platinum electrodes measuring 200 μm by 100 μm, spaced 750 μm apart.

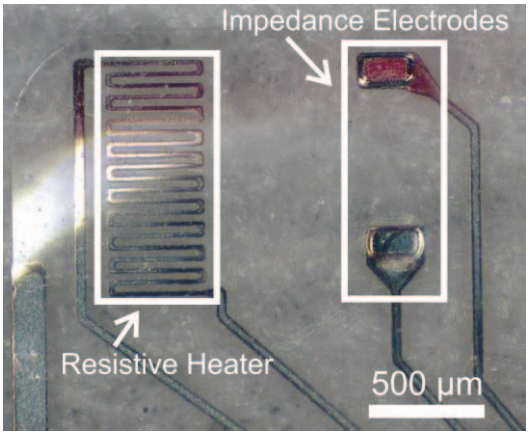

Figure 1: An electrochemical thermal flow sensor, composed of a resistive heater and a pair of impedance electrodes.

Fabrication (Fig. 2) starts with 12 μm thick Parylene layer deposited onto a silicon wafer (Fig. 2a), followed by standard UV lift-off lithography to pattern traces and contact pads in photoresist. A 2000Å layer of platinum was deposited on the wafer via electron-beam deposition (Fig. 2b) and lift-off was performed to define metal traces. A second 10 μm thick Parylene layer was deposited, and deep reactive ion etching was used to expose the electrode surfaces and separate individual devices (Fig. 2c). Finally, the sensors were released from the silicon wafer (Fig. 2d).

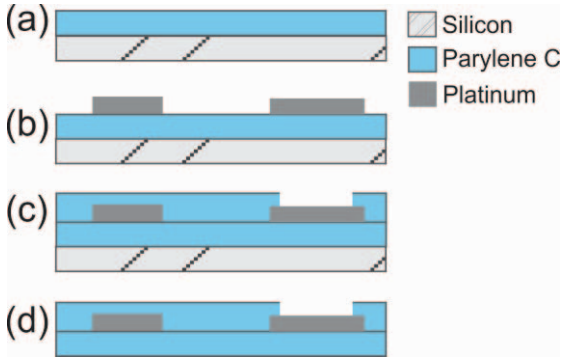

Figure 2: Flow sensor fabrication process

After fabrication, the sensors were affixed in a luer-lock adapter with an inner diameter of 3.25 mm (Fig.s 3,4). This luer-lock module was used to enable seamless connection to the shunt and catheter systems currently used in hospitals. External electrical connection was achieved via a Parylene ribbon cable with exposed platinum contact pads, which was then attached to a flat flexible cable using a zero insertion force (ZIF) connector and encapsulated in biocompatible epoxy.

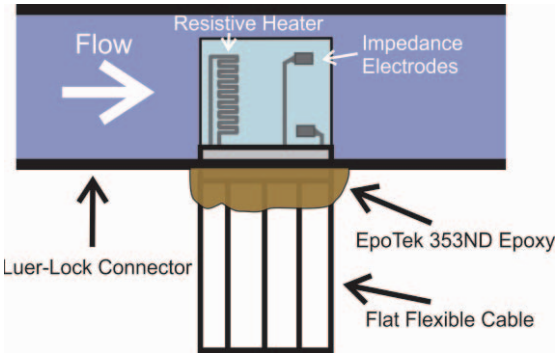

Figure 3: Diagram of luer-lock module packaging

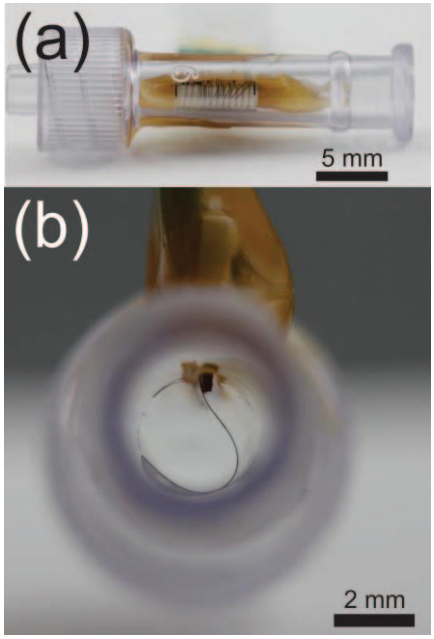

Figure 4: a) Side view and b) end view of sensors packaged in a luer-lock adapter

Experimental Setup

A peristaltic pump was used to drive liquid through the packaged sensors at rates from 0 to 400 µL/min, with minimum step sizes of 20 µL/min. Heat was generated using constant 2 mA current injections to the resistive heater for 10 seconds at a time and impedance was measured at 10 kHz with a precision LCR meter. 10 kHz was selected as the measurement frequency because at this frequency, the impedance consists almost entirely of the solution resistance between the two electrodes. Experiments were conducted at 21.0°C (room temperature) and 37.0°C (body temperature) with phosphate-buffered saline (PBS), a commonly used analog for physiological fluid. Deionized water at 21.0°C was used as a control.

RESULTS

The impedance between the electrodes was found to drop sharply when the heater was activated in PBS (Fig. 5). This is consistent with an increase in the mobility of ions in solution. When testing using deionized water, the impedance increased slightly, possibly due to a decrease in water's density or changes at the electrode-electrolyte interface. In PBS, the rate of change in impedance immediately upon heating increases in magnitude with an increase in flow rate. This rate was found to be an accurate measure of fluid flow over the majority of our tested range (Fig. 6).

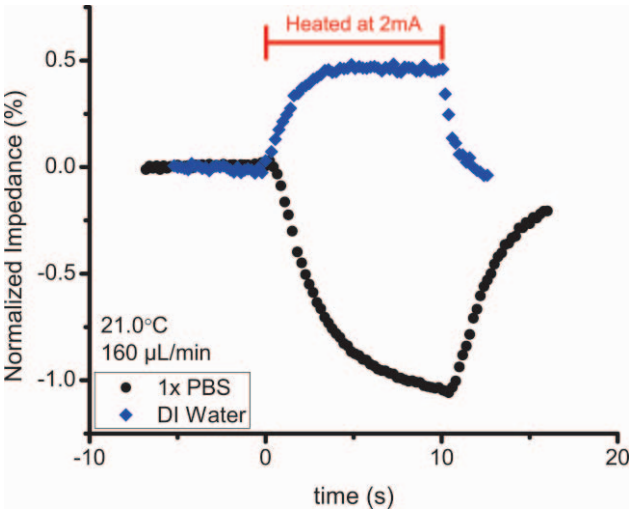

Figure 5: Normalized response of impedance electrodes to heating in 1x PBS and deionized (DI) water

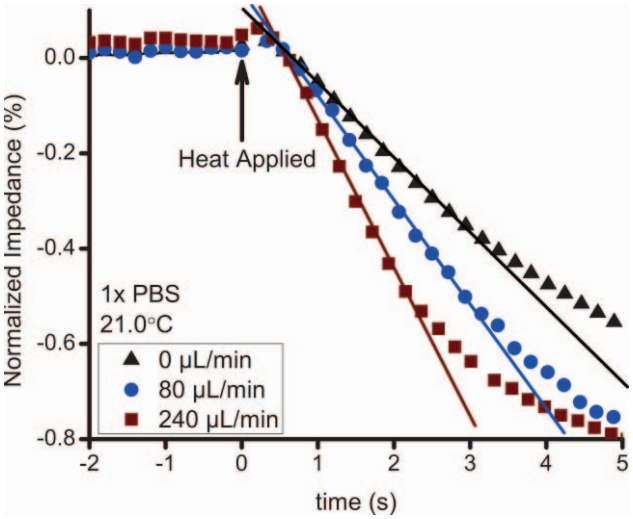

Figure 6: The rate of change of impedance upon heating increased in magnitude for increasing flow rates

The measured relationship between rate of change and flow is roughly linear at lower flow rates but tends to level off at higher rates. This may be due to the relatively low sampling rate of our precision LCR meter (~5 S/s), which would limit the maximum measureable slope. In the linear response range, which is from 0 to 320 µl/min, our sensor was able to clearly distinguish between flow differences of 20 µL/min, with a 3σ limit of detection of 10.7 µl/min at no flow (Fig. 7). Measurements at body temperature showed a small, consistent decrease in the magnitude of the sensor's response at each flow rate.

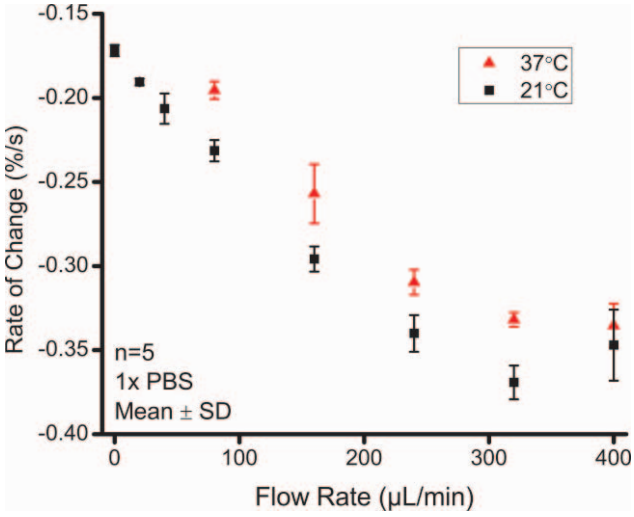

Figure 7: Rate of change of impedance versus flow rate at 21°C and 37°C

DISCUSSION

Our flow sensors were able to accurately detect flow rates within the physiologically relevant range for hydrocephalus shunts or external ventricular drains, and their flexible, fully biocompatible design makes them ideal for integration into shunts and catheters. When normalized to the pre-heating impedance, the response of the impedance electrodes was very consistent over multiple trials. The use of a solution's electrochemical impedance sensitivity to heat allows more sensitive flow measurements than when using the electrical resistance of solid temperature sensors. The required exposure of electrochemical impedance sensors directly to the flowing media sidesteps the problems associated with encapsulation of resistive solid sensors to provide protection from the physiological environment.

A small decrease in the magnitude of the sensor response was observed when measurements were made at body temperature. However, the response of the temperature followed the same linear trend, and the difference between room and body temperature measurements were relatively small compared to the 16°C temperature difference. The temperature in the body is relatively stable, so an implantable sensor may only experience a small amount of noise from localized or bulk body temperature changes. It may also be possible to calibrate the sensors to the surrounding temperature using the heater as a resistive temperature detector.

This flow sensor design is limited in its response to higher flow rates when the change in impedance becomes too fast for accurate measurement with our laboratory equipment, but the sensor could be tailored to a higher flow rate by increasing the distance between the heater and the impedance electrodes. However, this may come at the cost of a possible loss in accuracy at low and reversed flow rates. Alternatively, multiple pairs of impedance electrodes could be used to extend the range of discernable flows while maintaining low-flow accuracy. The use of multiple impedance electrodes could also allow more traditional thermal flow-sensing methods, such as calorimetric or true time of flight sensing. Either of these sensing methods implemented using impedance-based temperature sensing would be more sensitive and more robust in physiological environments than if they were implemented using resistive temperature sensors given in the improvement in the respective TCRs.

CONCLUSION

A thermal flow sensor has been developed which uses electrochemical impedance to sense the change in resistivity of the fluid upon being heated. The sensor is flexible and biocompatible, which allows easy integration into medical devices. The sensor was shown to be accurate in physiologically relevant flow ranges.

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