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Platinum Black Electroplated Impedance Particle Sensor

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***Abstract*—We present a micro electrical impedance particle sensor. To solve the problem of large electrode electrolyte interface impedance, we electroplated the electrodes with platinum black. Devices are fabricated with integrated parylene technology. An electrical model for the system is proposed and analytic solutions are obtained. Impedance spectra measurement of the device filled with various media are in excellent agreement with model analysis. Signals from individual 10 μm polystyrene beads passing the sensing electrodes are successfully obtained.**

Keywords-impedance; particle; platinum black; electroplating; paryleen

I. INTRODUCTION

Electrical impedance sensing has been one of the preferred technologies for particle sensing and counting. Since the invention of Coulter counter by Wallace H. Coulter in 1940s and 1950s [1], it has been used in most of the macroscopic blood counters. The initial DC resistance sensing also extended to AC impedance sensing. AC impedance sensing is an important method for liquid-borne particle analysis used in flow cytometry and modern blood counter. AC signals can penetrate biological cells depending on frequency, so more information can be extracted [2].

Traditional macroscopic blood counters use macro-electrodes and aperture size in hundreds of micron diameter range. Recently, there's a push to shrink the sensor as miniaturization enables smaller sample volume, better sensitivity, lower coincidence error rate and faster detection. Unfortunately, simply downsizing the electrodes would not work. Shrinking the size of electrodes increases the impedance on electrode electrolyte interface thus reducing the signal level and deteriorating the signal to noise ratio. For micro DC Coulter sensors, very large thin film electrode [3], macroscopic gold pin electrodes [4] and salt bridge with Ag/AgCl electrodes [5] have been proposed to solve this problem. This problem is less prominent in AC impedance sensing, because operating in high frequency reduces the double layer impedance of electrode electrolyte interface. Generally speaking, high frequency AC impedance sensing requires more consideration in electrode design, chip isolation, signal conditioning and processing. Micro AC impedance sensors have been demonstrated to sense cell suspensions [6], as well as single cell [7, 8].

Here, we present a new solution by using platinum black to increase the double layer capacitance by two orders of magnitude and achieve a micro impedance sensor operated at 10 kHz. We also perform a complete theoretical analysis in excellent agreement with experimental data of sensing 10 μm polystyrene beads in saline.

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II. THEORY

A particle impedance sensor can be modeled as Figure. 1. R1 represents the electrolyte resistance between the electrodes, which changes as particle flows by the electrodes. Here we ignored the reactance part of the electrolyte impedance with the assumption that the resistance part dominates the effect in low frequency domain. The electrode-electrolyte interface is modeled as C2 and R2. C2 is the double layer capacitance between metal electrode surface and electrolyte interface. R2 is the leaking resistance of the interface. Also, a stray capacitance C1 is included (e.g., from coupling between the substrate and electrode).

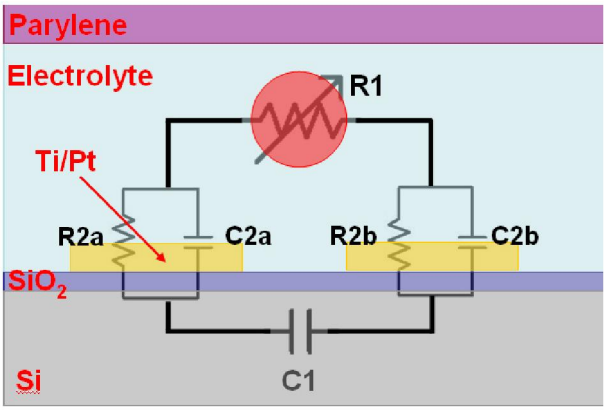

Figure 1. System Model

The transfer function of the model has two poles (P1 and P2) and one zero (Z1) and can be expressed as:

$$F(s) = \frac{R1+R2}{R2}\frac{1+s/Z1}{(1+s/P1)(1+s/P2)} \quad (1)$$

For microelectrodes, R2 (~GΩ) is much larger than R1 (<100kΩ). Therefore

$$Z1 = \frac{1}{R1C2} \quad (2)$$

$$P1 = \frac{C1+C2}{R1C1C2} \quad (3)$$

$$P2 = \frac{1}{R2(C1+C2)} \quad (4)$$

It can be proved that $P2 < Z1 < P1$ under the condition R2<<R1.

A general plot of the magnitude spectrum of the frequency response of equation (1) shows a plateau frequency region limited by Z1 and P1. It defines the possible frequency zone for sensing, in which the overall impedance is sensitive to solution resistance R1. We call this region in frequency as *sensing zone*. For frequency lower than Z1, the system behaves as double layer capacitor C2. For frequency higher than P1, stray capacitance C1 will dominate the frequency response.

Also shown in Figure 2 are two extreme cases where the sensing zone shrinks and the whole system behaves like a capacitor. The first case is when R1 is extremely large. This is the case where the channel is filled with air or DI water instead of electrolyte. The other case is when C2 decreases. In micro systems, downsizing causes C2 to decrease and R2 to increase. Thus the overall impedance is dominated by the double layer. The signal from resistance changes due to the passing particles is reduced. On impedance spectra, this means zero Z1 moves to higher frequency and approaches high frequency pole P1. The sensing zone diminishes (Figure 2). One possible solution is to reduce the stray capacitance C1. This will increase P1, which shifts the sensing zone to higher operating frequency. This is not always favorable. It can make the device structure and sensing circuitry more complicated. Also in some cases, there is a favorable range of sensing frequency determined by the nature of the particles. For example, human tissue and cell suspension are supposed to be sensed in frequency region of β dispersion, which is between 100 kHz and 10 MHz [2], for identification and differentiation purpose. Another solution is to increase double layer capacitance C2, which will decrease zero frequency Z1 and extend the sensing zone to lower operating frequency. To increase C2, our approach is to use platinum black electroplating, which can increase surface area of the electrodes and thus the capacitance by two or three orders of magnitude [9].

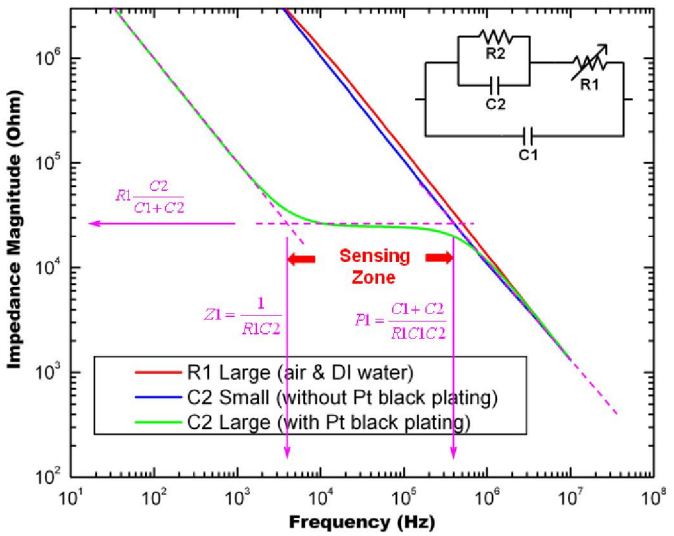

Figure 2. Numerical simulation of impedance magnitude spectra. Dash lines are asymptotes

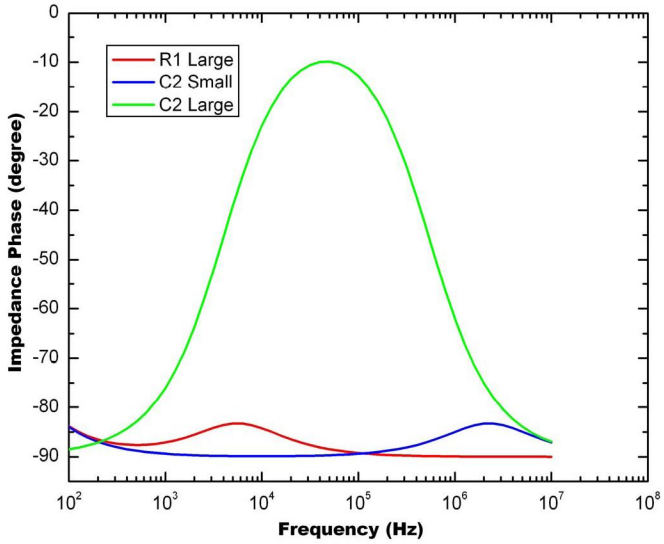

Figure 3. Numerical simulation of impedance phase spectra

Numerical simulation of impedance magnitude spectra. Dash lines are asymptotes.

III. Design and Fabrication

An impedance sensor was made (Figure 4) inside a parylene channel strengthened by SU8. First oxide is grown thermally to provide electrical isolation. Then Ti/Pt/Ti (200A/2000A/200A) electrodes are patterned by liftoff process. The portion of top Ti layer inside channels is etched away with buffered HF. Ti/Pt electrodes are used because they are chemically inert and can withstand high voltage without erosion. Other portions of the top Ti layer are kept to provide superior adhesion to parylene. To avoid parylene channel delamination during later processing stages, parylene anchors are made on silicon by DRIE. Then 10μm of parylene is deposited by CVD and patterned by RIE with Ti/Au mask. Before sacrificial photoresist releasing in acetone, a 50μm thick SU8 layer is coated and patterned to planarize the surface and strengthen the parylene channels.

Platinum black is electroplated selectively on the electrode surface exposed to the channel. Electroplating solution contains 1% chloroplatinic acid (Sigma C-3044), 0.0025% hydrochloric acid, and 0.01% lead acetate in water [9]. Electroplating solution is introduced into channel by either syringe injection or vacuum suction. Electrodes are plated with 5 V DC through a 10 MΩ resistor for up to four minutes. The resultant electrodes are shown as inset of Figure 8.

Figure 5 shows a fabricated device, with 20 μm channel height and 10 μm parylene thickness. There are three devices on each chip. The left one has two sensing channels in parallel with aperture size of 20 μm in length by 15 μm in width. Each aperture has one pair of sensing electrodes separated by 60 μm. The middle one has an aperture size of 20 μm in length by 15 μm in width. It has two pairs of electrodes separated by 50 μm. The right one has a 900 μm long and 30 μm wide sensing channel. Four 6 μm electrodes are equally separated by 3 μm. Later testing is all performed with the middle device.

A transparent acrylic packaging jig is designed and fabricated to provide convenient electrical and fluidic accessibility from the top and thus avoid backside wafer processing (Figure 6). The jig can provide eight fluidic accesses simultaneously. Commercial fittings (Upchurch Scientific, WA, U.S.A.) are used to connect between the jig and other fluidic components, such as syringe pump or outlet tubing. There is a recess at the bottom of the acrylic jig, which provides space for epoxy strengthened wire bond region of the chip.

After fabrication, the chip is glued and wire bonded onto a printed circuit board (PCB). There are a total of 44 pads on the PCB and we use 12 of them. A 200 μm thick PDMS layer with through holes is aligned and placed on top of the chip to provide sealing between the jig and the chip. Finally, the jig is tightened firmly onto the PCB with screws (Figure 7).

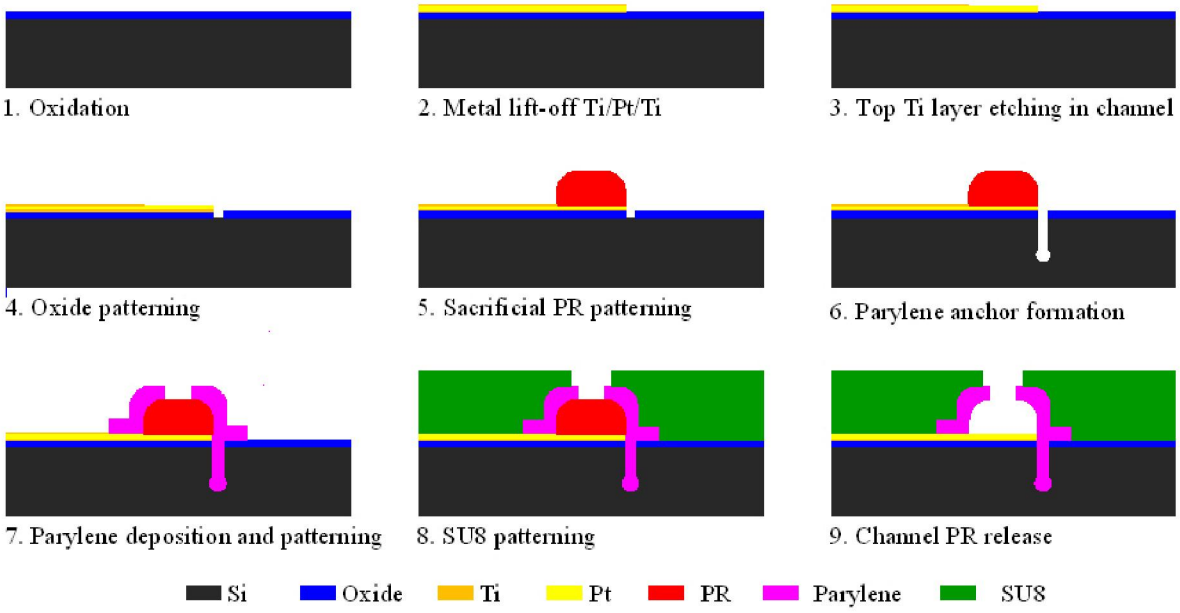

Figure 4. Fabrication process flow

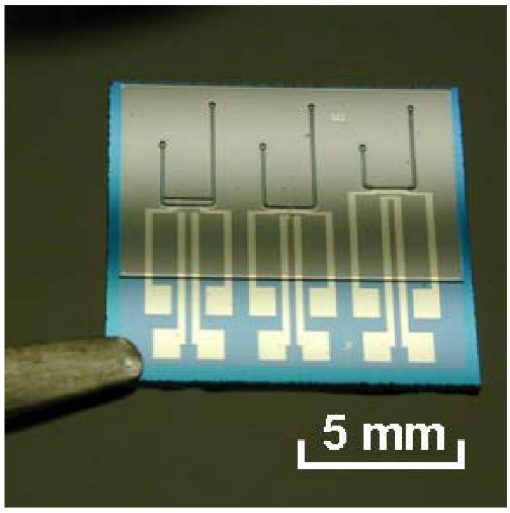

Figure 5. Fabricated device.

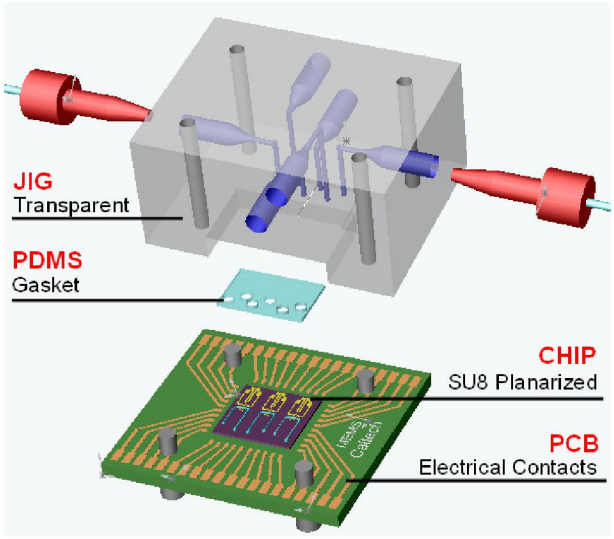

Figure 6. Chip assembly with jig and PCB

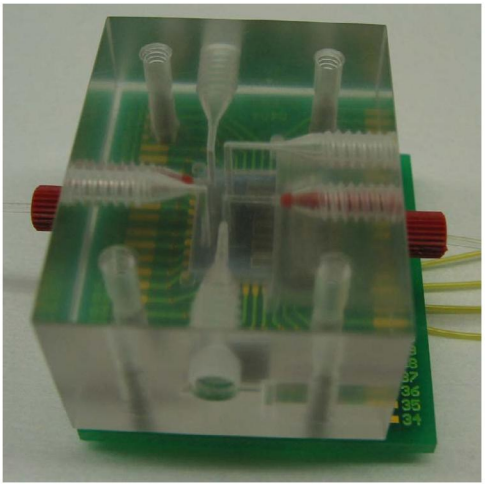

Figure 7. Assembled device with jig

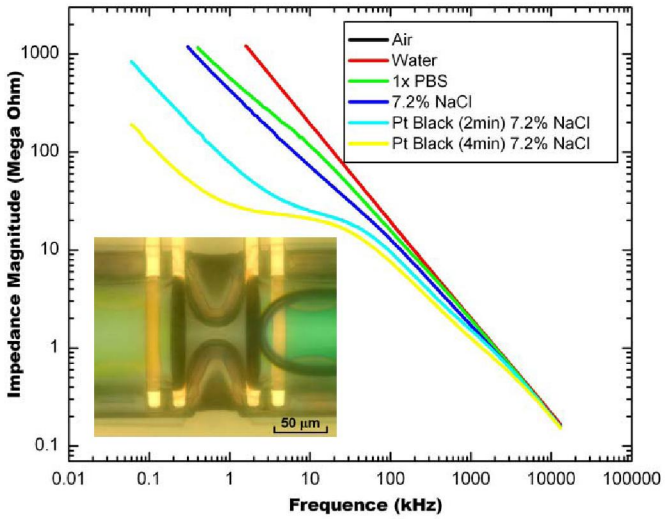

Figure 8. Measured impedance magnitude spectra in different environments. Electroplating with Platinum Black (the lower two curves) effectively expand the sensing zone.

IV. Results

The impedance spectra of the device filled with different medium is measured with LF impedance analyzer (HP 4192A). Measured impedance spectra in Figure 8 and 9 demonstrate the expansion of sensing frequency zone by platinum black electroplating. The frequency responses of air and water almost overlap. Their magnitude responses are straight lines and phase responses keep a constant -90 degree angle for the frequency range scanned. This is the case of large R1 in previous model analysis part and the system behaves as a capacitor of 80 pF.

Before platinum black electroplating, both phosphate buffered saline (PBS) and high salt solution (7.2% sodium chloride) have little impact on the frequency response. There is no sensing zone, which means the system is not sensitive to what is in the sensing region. Air, water and saline solution results in very similar impedance spectra. This is because at high frequency, the system is dominated by stray capacitance C1 and at low frequency by C2. Since Z1 and P1 are so close, the sensing zone in frequency domain shrinks to zero. After electroplating, C2 is measured to be 2.5 nF, an increase of two orders of magnitude. The magnitude response shows a plateau region between 100 Hz to 100 kHz. This region expands as the time of electroplating increases. The phase response has a large increase in the same frequency range and the maximal phase becomes -20 degree at 5 kHz for four minutes platinum black electroplating.

For particle sensing, the measurement system includes a high impedance buffer amplifier as input stage, a true RMS to DC converter and a high fidelity instrument amplifier (Figure 10). An oscilloscope in edge triggered mode is used to record the results. Testing with 10 µm polystyrene beads (Duke Scientific Corp, CA, U.S.A.) generates signals in agreement with expected pulse height and duration (Figure 11). On the other hand, sensor without platinum black can not sense any signal at all.

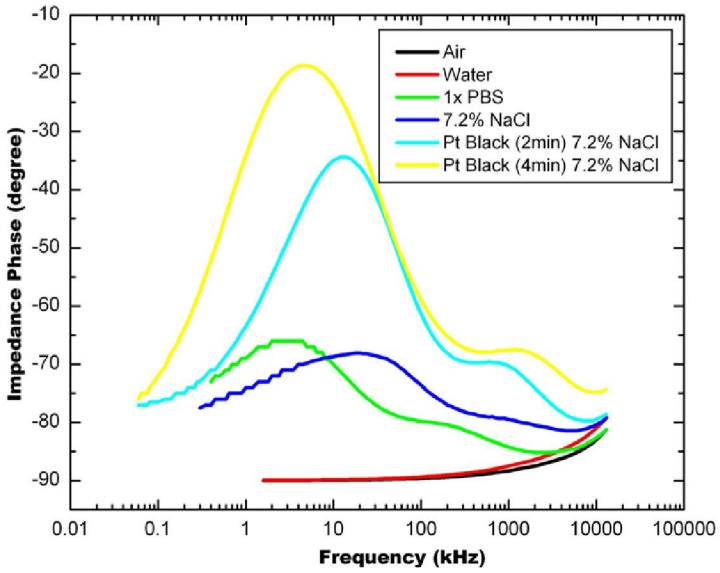

Figure 9. Measured impedance phase spectra

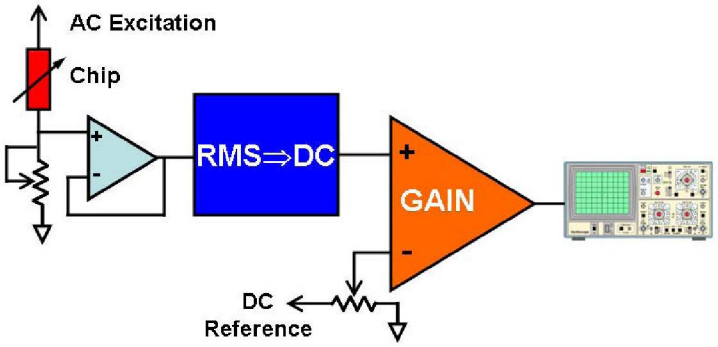

Figure 10. Measurement system

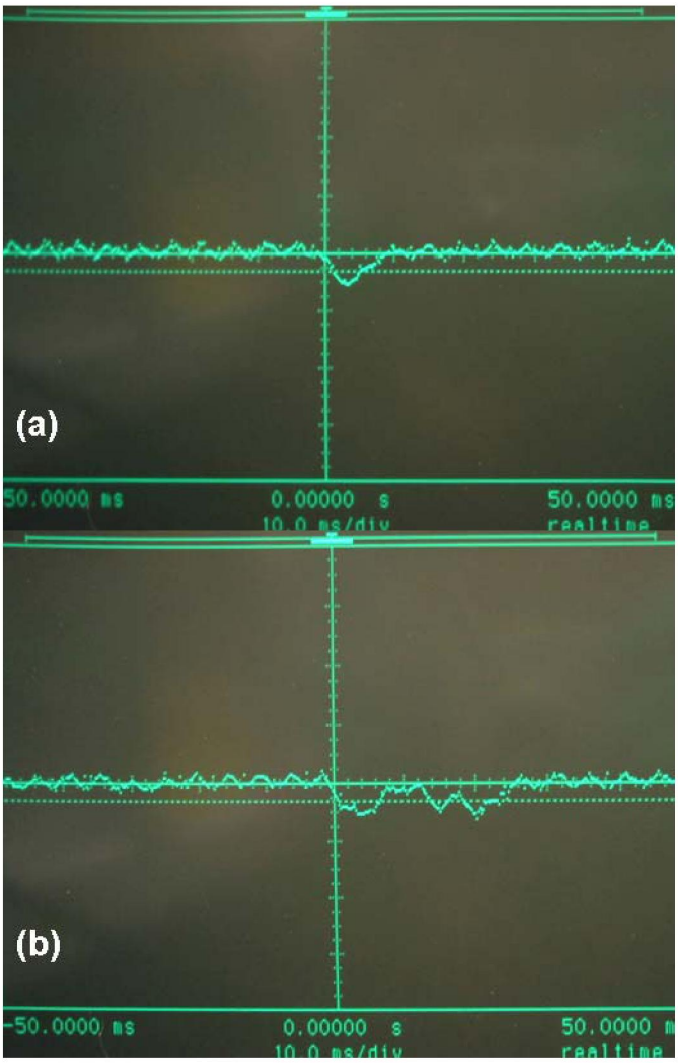

Figure 11. Two events of voltage signals of 10 μm beads

V. Conclusion and Discussion

We design and fabricate a micro Coulter sensor for particle sensing in fluidic flow. An analytic model is developed and used to explain testing results. From the analytic solution of the model, a sensing zone in frequency domain is identified. The lower bound of it is limited by the double layer effect of the electrode electrolyte interface. Miniaturization lowers the double layer capacitance and shifts this lower bound to higher frequency. Thus the sensing zone in frequency domain shrinks. We successfully increased the double layer capacitance of the micro Coulter sensor by platinum black electroplating, which increase the electrode surface area. We realize there are various protocols for platinum black electroplating and the properties of the resultant electrodes are different. There are work to be done to optimize this process to achieve maximal surface area increase and strong mechanic properties. The upper bound is set by the stay capacitance of the electrodes. Overall device structure needs to be carefully designed to shift the upper bound to higher frequency. We are also looking into other ways to reduce this effect.

Integrated parylene technology is used for device fabrication. The benefits of parylene include its processibility, stability, flexibility and biocompatibility. We also designed a jig to provide fluidic access and a PCB to provide electric connections both from top surface of the chip. This is preferred since it eliminates back wafer process. A thin PDMS piece is used as gasket between the chip and the jig.

Preliminary testing is performed with polystyrene beads. One problem happened during test is the blocking of aperture by beads of abnormal size or bead aggregate. Including an upstream filter may be necessary to block anything larger than the aperture size to ensure the chip can be operated long enough. Although we can get the signal from the sensing circuitry, improvements are necessary to increase signal to noise ratio.

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