A Novel Compact Lab-on-a-Chip Nanosensor for In-Channel Liquid Viscosity Detection

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ABSTRACT

Real-time monitoring of liquid's viscosity has significant meaning in many areas. To adjust the viscosity, glycerol solutions with various concentrations have been widely used. Different types of viscosity sensors have been developed using various principles. However, these sensors have complicated fabrication and testing procedures and are not truly compatible with fluidic systems for on-site viscosity detection. Sensors based on single-walled carbon nanotubes (SWNTs) have attracted significant attention in recent years due to their smaller size, more uniform geometry, and more consistent performance. In this paper, we report a novel lab-on-a-chip device with integrated SWNT nanosensors for glycerol concentration detection, which is an indirect indication of fluid's viscosity. The device enables real-time, in-channel measurements of the concentration of flowing aqueous glycerol solutions. The experimental results show that our device has a relatively high sensitivity for glycerol solutions. The sensor resistance increases when the glycerol weight ratio rises, indicating an increased viscosity. The sensor also responds to the flow velocity when the glycerol weight ratio is kept at a constant level. Our results demonstrate a simple and effective approach to create nanoscale in-channel glycerol sensors. The knowledge obtained from this research can enable the development of future novel glycerol-based viscosity sensors for biomedical applications.

Keywords: single-walled carbon nanotubes (SWNTs), nanosensor, viscosity, glycerol, lab-on-a-chip device

1 INTRODUCTION

Viscosity, defined as the resistance of a fluid to flow, is one of the most important parameters to characterize fluid properties in the field of rheology and tribology [1]. Real-time monitoring of liquid's viscosity has significant meaning in many areas such as automobile engineering, biomedical research, and petrochemistry [2]. For example, in the biomedical field, the changes in fluid viscosity usually lead to cell function disabilities and can cause blood-related diseases [3]. Aqueous glycerol solutions with various viscosities have been widely used in experimental studies of flow characteristics [4]. Glycerol can be obtained either by microbial fermentation or chemical synthesis from petrochemical feedstock. It has been widely used in a lot of areas such as food, pharmaceutical, pulp and paper, leather and textile industries. Because the viscosity change of glycerol solution largely influences the quality of the final products, real-time monitoring of the glycerol concentration in solutions is critical in these applications.

Recently, different types of viscosity sensors have been developed using various principles including the detection of resonant frequency change of vibrating micromachined cantilevers and the fluorescence intensity change of molecular rotor dyes [2]. However, for the micromachined viscosity sensors, the complicated fabrication processes and large sample scales make them ineffective in real-time, on site fluid monitoring. While for the fluorescence sensors, the need of specialized instruments and the limited spatial resolution limit their accessibility for routine laboratory experiments [3]. Similar to the micromachined devices, these fluorescence sensors with complicated testing procedures are not compatible with microfluidic systems for on-site viscosity detection either.

Single-walled carbon nanotubes (SWNTs) have attracted significant attention in many areas due to their extraordinary electrical and mechanical properties. SWNT-based device provide a label-free, real-time, and ultrasensitive approach for sensing applications [5]. They have been used in a wide range of micro/nanoscale devices as shear stress sensors [6] and chemical concentration sensors [7]. Since first introduced in the early 1990s, microfluidic devices, or lab-on-a-chip devices, have become increasingly prevalent in a wide range of areas; they have given sensor technology new meanings and new opportunities [8]. Microfluidic systems for biomedical or chemical studies have achieved significant success in recent years due to their advantages including low reagent and power consumption, short reaction time, low cost, and high compatibility to integrate with other miniaturized devices [9]. However, the real-time, in-channel detection of fluid's properties is still challenging.

In this paper, we report a novel lab-on-a-chip device with integrated SWNT nanosensors for indirect detection of viscosity, which is extracted from the measurement results of glycerol concentration in solutions. SWNTs are aligned across two electrodes with dielectrophoresis and then used as nanosensors for fluid detection. Glycerol solutions with different viscosities are obtained by mixing glycerol and deionized (DI) water with different weight ratios. The lab-on-a-chip device fabricated with photolithography and soft lithography methods enables real-time, in-channel monitoring of the flowing aqueous glycerol solutions. The resistance of the SWNTs in the sensor illustrates clear and repeatable dependence on the glycerol concentration. Overall the sensor resistance increases when the glycerol weight ratio rises, indicating an increased viscosity. The design, fabrication, and measurement of the device are described and discussed in this paper.

2 EXPERIMENTS

Previous studies show that both the aqueous ionic density and the velocity of the fluid flow have influences on the nanosensor's behavior [6, 10]. The SWNT-based sensors have a high sensitivity towards the electric potential caused by the specific binding and unbinding effects between ions and SWNTs in the microfluidic environment [10]. Therefore DI water is used in our experiments to minimize the electrical disturbance caused by the ions in the fluid flow. In our experiments, the testing solutions with different viscosities are obtained by mixing glycerol and DI water with different weight ratios [11]. Five solutions are made with different glycerol weight ratios of 10%, 20%, 30%, 40%, and 50%. Their corresponding viscosities are 1.31, 1.76, 2.50, 3.72, and 6.00 centiPoise, respectively [11] In the measurement process, these solutions are injected into the channel continuously with a high-precision syringe pump. A semiconductor device analyzer is connected to the sensor for the real-time detection of glycerol concentration.

Figure 1a shows a fabricated lab-on-a-chip device with integrated SWNT-based sensors used in our experiments. This device contains an array of sensors, enabling the possibility of performing parallel mesaurements. Figure 1b shows the schematic of the device, which has a silicon-SU-8-polydimethylsiloxane (PDMS) sandwiched structure. The SU-8 layer in the middle contains all the microfluidic components including an inlet, an outlet, and a microchannel. A thin PDMS piece is used as the top cover to seal the microfluidic components. SWNTs are deposited between the electrodes in the fully sealed microchannel and used as the sensing element.

The fabrication of the entire system can be divided into two parts: the fabrication of lab-on-a-chip device and the integration of SWNT nanosensors. In the first part, the device is fabricated using photolithography and soft lithography methods. Metal layers of Cr and Au are patterned on a silicon wafer surface as electrodes using metal sputtering, photolithography, and wet etching. The multi-teeth electrodes are designed to ensure the controlled dielectrophoresis deposition of SWNTs [12]. The gap between the opposite electrodes is 5 µm. Then a 40 µm thick SU-8 layer is patterned on top of the electrodes to form a microchannel with photolithography. Next, a piece of PDMS with an inlet and an outlet is bonded to the SU-8 layer to seal the channel. The bonding between the two polymer is a critical step. Surface treatment using chemicals and plasma are needed to ensure the bonding quality [13].

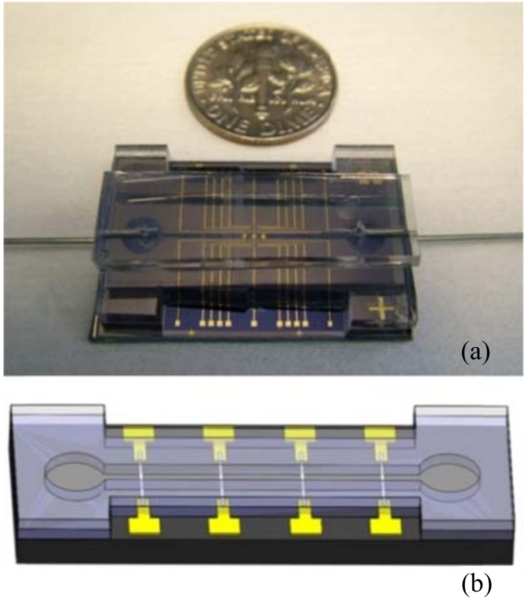

Figure 1: (a) A fabricated lab-on-a-chip device with integrated SWNT nanosensors. (b) Schematic of the device.

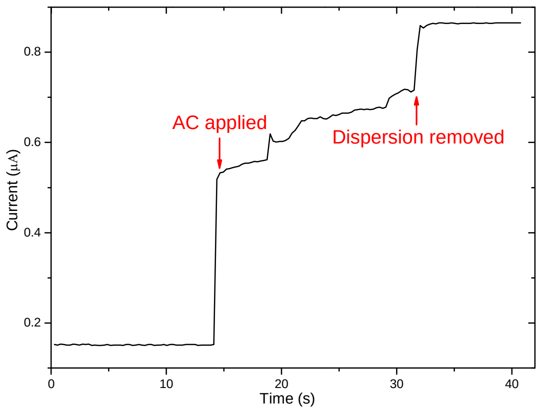

Figure 2: Real-time current change during the dielectrophoresis process.

In the second part of the process, dielectrophoresis is used to assemble the SWNTs between the multi-teeth electrodes. To ensure the motion and rotation ability of the SWNTs in DI water, a chemical method is used to treat the pristine SWNTs for enhanced water solubility [12]. The concentration of the SWNT solution is approximately 0.2 mg/ml. The SWNT dispersion is injected into the microchannel with a syringe. An alternating current (AC) signal is applied across the electrodes using a function generator to generate a non-uniform electric field in the solution to rearrange the dispersed SWNTs. A digital multimeter is used to monitor the real-time current change across the electrodes. A clear current change can be observed when the SWNTs are assembled across the electrodes, as shown in Figure 2. After the dielectrophoresis process, the SWNT dispersion is removed from the microchannel by pushing air into the device using a syringe.

Then DI water is injected into the microchannel with a low flow velocity for a few minutes to eliminate the unbonded SWNTs.

3 RESULTS AND DISCUSSION

The current-voltage (*I-V*) characteristics of the SWNT sensors are obtained with a semiconductor device analyzer. Pure water and three glycerol-water mixture solutions with glycerol weight ratios of 10%, 20%, and 30% are injected into the microchannel with different velocities which are controlled by the syringe pump. These four velocities are 0.1, 0.2, 0.3, and 0.8 ml/min. Resistances under these different conditions are collected and shown in Figure 3. Our SWNT sensor shows a high sensitivity for different glycerol solutions: under static (0 ml/min) conditions the resistance increases when the glycerol concentration increases. When a low flow rate of 0.1 ml/min is applied to the solution, a small increase in resistance can be observed for all the tested solutions. In the medium flow rate range of 0.2 ml/min and 0.3 ml/min, the resistance change is less obvious except for pure water. However, when a high flow rate of 0.8 ml/min is applied, the resistances of all the solutions present a decreasing trend.

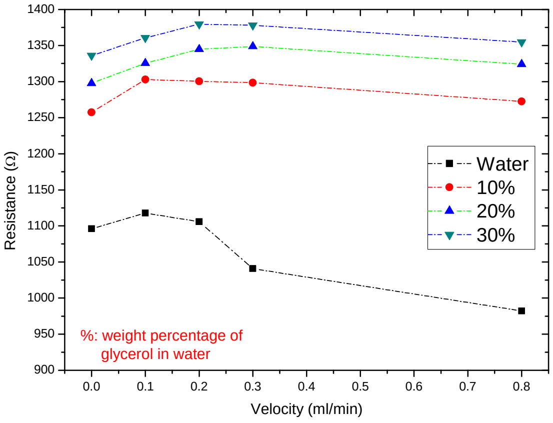

Figure 3: Sensor resistance in four different solutions with different flow velocities.

Previous studies found that a SWNT could adsorb chemical reagent molecules on its surface in a liquid. This adsorption could cause charge transfer between the SWNT and the molecules, leading to an electrical property change for the SWNT [7]. In our experiments, the nanosensor's resistance shows an obvious increase when the glycerol concentration rises. This trend may be explained by the interaction between the glycerol molecules and the SWNTs.

However, there are some other factors that can influence the sensing performance of SWNT sensors. Early studies reported that structural deformations of a SWNT could also change its electrical properties [14]. Bending or twisting a SWNT can decrease its transmission function and leads to an increase in resistance. However, there is a limit for this increase because once the bending exceeds a certain range, more kinks are generated in the structure. These kinks can lead to tubular structure collapse or damage [14]. Other recent studies demonstrated that when an electric current was applied to a SWNT, its temperature was increased due to generated heat [6]. Once a flow was introduced onto the SWNT, its temperature was decreased because of the heat convection between the flow and the SWNT. Consequently, a higher flow rate caused a lower temperature on the SWNT, creating a lower resistance reading.

The real-time sensing performance of our sensor in a wider velocity range is investigated. In a separate experiment, the syringe pump is used to change the flow rate in the microchannel and the real-time resistance change is recorded and shown in Figure 4. Three glycerol-water mixture solutions with glycerol weight ratios of 10%, 20%, and 30% are selected and the flow rates are set as 0.1, 0.5, and 0.9 ml/min. For each tested glycerol solution, the resistance shows a clear drop when the flow rate is increased and it reaches a steady state quickly after the flow rate change. Comparing the three tested solutions at any given flow rate, the resistance increases when the glycerol weight ratio is increased. This means that under the same flow rate, the nanosensor's resistance change reflects the glycerol concentration in the solution. This can be used to indicate the viscosity change of the flowing fluid in a microchannel.

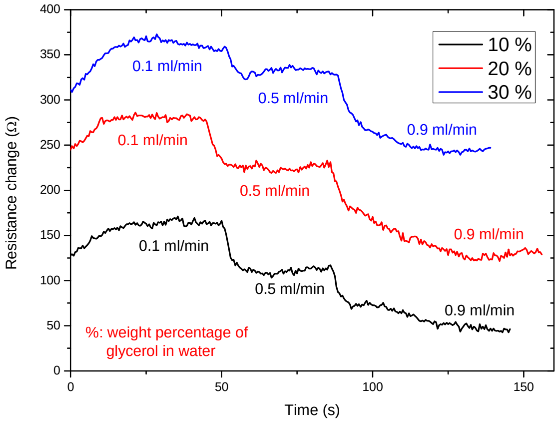

Figure 4: Real-time sensor resistance changes in continuous flows with different velocities.

Another experiment is conducted to evaluate the real-time response of the nanosensor for different glycerol solutions. In this experiment continuous fluids with a constant flow rate of 0.1 ml/min are injected into the microchannel. The real-time detection results are illustrated in Figure 5. The sensor's resistance increases when the glycerol concentration rises. The lowest resistance is measured as approximately 1100 Ω in water and the highest value is 1320 Ω in a 50% glycerol solution.

Our preliminary results demonstrate that the lab-on-a-chip device with intergrated SWNT nanosensors has a high sensistivity towards the glycerol concentration change. This behavior can be used to determine the fluid viscosity indirectly. Further experiments will be developed and performed to investigate the possibility of using this device to measure the liquid's viscosity directly. Both experimental and theoretical approaches will be used for us to achieve a better understanding of the sensing mechanisms. In addition, we will examine the applicability of the new device in practical applications.

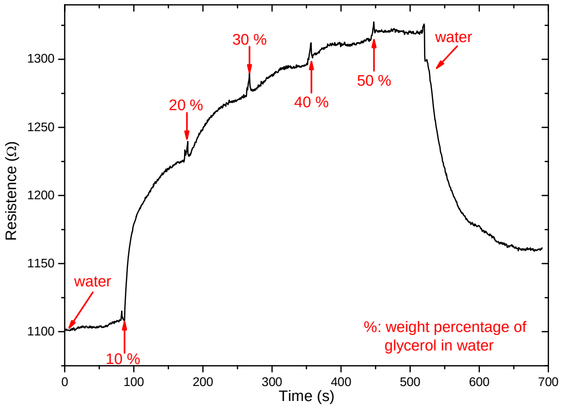

Figure 5: Real-time sensor resistance change in a continuous flow using fluids with different viscosities.

4 CONCLUSIONS

In summary, we have investigated a new method for the detection of glycerol concentration in solutions using an integrated microfluidic-nanosensor system. This method can be used for indirect measurements of liquid's viscosity. A SWNT-based glycerol sensor is fabricated with photolithography and soft lithography methods. The real-time resistance reading of the SWNT sensor reveals clear and repeatable dependence on the glycerol concentration. In general, the sensor resistance increases when the glycerol ratio rises, indicating an increased viscosity. Three potential sensing mechanisms are described and discussed in the paper. Even though the deformations of SWNTs and the heat loss caused by the flowing fluid may influence the SWNTs' electrical properties, these effects can be ignored when applied in a constant-flow environment. However, further studies are needed to investigate the sensing mechanisms. The knowledge obtained from this research can enable the development of novel lab-on-a-chip sensors for applications in medical areas such as blood testing and disease diagnostics.

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