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Hydrogel-CNT biomimetic cilia for flow sensing

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Abstract— In this work, a biomimetic cilia flow sensor is reported which comprises of an electrospun PVDF nanofiber sensing membrane with vertically aligned carbon nanotube (VACNT) bundle at the center of the membrane. The VACNT bundle is infused with hydrogel to mimic the natural cupula found in fishes. Hydrodynamic oscillatory flow testing is conducted on the fabricated sensor using a vibrating sphere stimulus.

Keywords—Polyvinylidene Fluoride (PVDF); Electrospinning; Nanofiber; Piezoelectricity; Carbon Nanotube (CNT); Biomimetics; Flow Sensing

I. INTRODUCTION

Biological sensors found in nature are well optimized through the evolutionary processes. In nature we find examples of biological sensors that are compact, ultrasensitive and efficient. The excellent sensing capabilities of the biological sensors have attracted the attention of scientists and engineers to study their morphological and functional properties and mimic them for developing efficient artificial sensors. Hair cell mechanoreceptors are a class of highly efficient sensors found in a number of living organisms including fishes, insects, and mammals [1]. Due to the exceptional flow sensing capabilities of biological hair cell sensors, mimicking them to develop artificial sensors has been of particular interest. In the past, arrays of hair cell inspired MEMS flow sensors have been developed to mimic the functionality of lateral line sensors (LLS) found in fishes [2]–[8]. The individual flow sensors within lateral-line, known as neuromasts, feature elongated naturally secreted polymer-like structures called as cupula. In some of the MEMS sensors reported in the past, cupula and cupular fibrils inspired by the neuromasts have been mimicked to increase the sensitivity of such sensors [6,7]. In order to mimic the composition of cupula, materials like electrospun nanofibers and drop-casted hydrogel cappings have been used in the past [6], [7]. Cupula helps in enhancing the sensitivity of flow sensors by increasing the overall cross-sectional area exposed to fluid flow. In addition, the material properties of cupula were also reported to have a contribution towards enhancing the overall drag force and thereby the sensitivity [6], [7].

This work reports the development and characterization of a self-powered nanoelectromechanical systems (NEMS) flow sensor which uses a combination of soft-materials processing and state-of-the-art nano-fabrication techniques. The sensor reported in this work features an electrospun PVDF nanofiber sensing membrane with a hyaluronic acid (HA) infused high aspect ratio VACNT bundle which mimics the biological cupula.

Oscillatory flow tests are conducted on the sensor to determine the threshold detection limit and sensitivity. Sensors such as the one reported here are in demand in a wide range of future applications including microfluidics, biomedical devices, wearable sensors, and energy harvesters.

II. EXPERIMENTAL

A. Electrospun PVDF nanofiber membrane

PVDF powder (MW 534000) (1.5 g), is mixed with acetone (7 ml), and dimethylformamide (DMF) (3 ml) (all of which were acquired from Sigma Aldrich) to prepare a 15% (w/v) polymer solution. PVDF is dissolved at 70 ºC for 60 min using a hot plate stirrer. The resulting solution is incubated in a hot air oven at the same temperature for 30 min to obtain a homogeneous solution. The polymer solution is filled in a 10 ml syringe and fed to an 18 G needle using a syringe pump with a constant solution flow rate of 5 µl min⁻¹. A rotating mandrel collector (10 cm in diameter) is used for collecting the electrospun nanofibers. A voltage of 12 kV is applied between the rotating mandrel collector (with a rotation speed of 1500 r.p.m) covered with aluminium foil and the conductive needle tip while maintaining a constant 15 cm gap between them.

B. Hydrogel cupula

HA hydrogel modified with tyramine (Tyr) is used to form the hydrogel cupula. The morphology of the hydrogel infused high aspect ratio VACNT pillar is studied using a Jeol JSM6701F field emission scanning electron microscope (FESEM). The synthesis, material and mechanical characterization of HA-Tyr hydrogel is described in our earlier works in [9]. Rheological analysis is conducted on the hydrogel samples which are swollen in de-ionized water until an
equilibrium is reached. Young’s modulus of the HA-Tyr cupula as determined through rheology measurements is 10.4 kPa.

III. RESULTS AND DISCUSSION

A. Biomimetic cilia sensor fabrication

The sensor consists of a high aspect ratio VACNT bundle (CVD equipment Corp, New York, NY, USA) covered with HA-Tyr hydrogel cupula located at the center of electrospun PVDF nanofiber sensing membrane. Figure 1(a)-(c) shows the images of VACNT bundle. The VACNT bundle is fixed at the root but is free to move at the distal tip. The sensing membrane is a 20 µm thick PVDF nanofiber film formed through electrospinning (figure 1(d)). The electrospun nanofibers are transferred to an optically clear adhesive (OCA) film with a 2 mm cavity punched in the center of film.

Copper tapes connected to both the ends of PVDF nanofiber membrane are used to form electrical contacts to the sensor. An identical OCA film with a 2 mm cavity is aligned and fixed over the nanofibers in such a way that the PVDF membrane is suspended on the cavity formed between the two OCA films. Thereafter, HA-Tyr hydrogel is drop-casted on the VACNT bundle in order to encapsulate it. The VACNT bundle is exposed to oxygen plasma before the drop-casting process as the hydrophobic nature of CNTs did not allow conformal coverage of the hydrogel. The sensor is then immersed in de-ionized water to allow the cross-linked hydrogel to attain a swelling equilibrium. The fabricated sensor with and without the hydrogel-based biomimetic cupula is shown in figure 1(e) & (f).

B. Sensor testing and characterization

The fabricated sensor without the cupula was subjected to an oscillatory flow using a dipole stimulus to determine the flow sensing performance. A vibrating sphere (16 mm in diameter) connected to a permanent magnet mini-shaker (Bruel & Kjaer model 4810, Norcross, GA) by means of a 15 cm long rod (4 mm in diameter) is used to generate a dipole stimulus. The flow velocities generated by the dipole stimulus are pre-calibrated using a laser Doppler vibrometer (Polytec PSV-300) [8]. In the flow sensing experiments, the damping of oscillatory flow velocity due the fluid is not considered.

The sensor is immersed in a tank of water and is positioned 25 mm away from the vibrating dipole in water. The output from the sensor is connected to a National Instruments (NI) data acquisition card and is acquired in LABVIEW software. To generate varying oscillatory flow velocities, the dipole is vibrated at a constant frequency of 35 Hz in a plane perpendicular to the long axis of VACNT bundle. Figure 2 shows the schematic representation of the experimental setup used for flow characterization of the sensors.

A flow velocity sweep over the range of 0-100 mm/s is performed while the output of the sensor is continuously acquired. The drag force generated on the CNT bundle by oscillating water flow causes the bundle to oscillate at the same frequency as that of the flow. Since the CNT bundle is attached
to PVDF membrane at the base, the displacement of bundle causes a strain distribution in the nanofiber membrane. Due to the piezoelectric nature of membrane electric charges are generated, which are acquired as voltage output.

With increasing velocity of the oscillatory flow, the sensor output monotonically increased. Flow calibration experiments demonstrated the capability of sensor in detecting low flows with a threshold detection limit of 5 mm/s.

IV. Conclusion

In this work, a simple method of fabricating a biomimetic hair cell sensor is demonstrated. Electrospun PVDF nanofiber membrane is used as the sensing element while a VACNT bundle is used as the biomimetic cilia. The VACNT bundle not only transfers the flow-induced drag force to the PVDF membrane but also acts as a scaffold facilitating the hydrogel cupula formation. HA-based hydrogel with optimized cross-linking is used for mimicking the material and mechanical properties of the biological cupula. The fabricated sensor without the cupula is subjected to a dipole stimulus for studying its oscillatory flow response. Based on the flow characterization experiments, the sensor demonstrated a threshold detection limit of 5 mm/s. Flexible, self-powered and sensitive flow sensors such as the ones demonstrated in this work will find usage in a multitude of applications dealing with microfluidics, biomedical sensing, and environmental sensing. In addition, the novel materials and methods employed in the sensor could pave way for the design of future generations of biomimetic smart sensors.

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