

Full paper

Flexible-detachable dual-output sensors of fluid temperature and dynamics based on structural design of thermoelectric materials

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ABSTRACT

Multifunctionalities with self-powered capability are crucial for miniaturized, scattered devices to sense temperature and dynamics of fluids, which are intrinsic parameters to monitor environmental or industrial features. Herein, we present flexible-detachable dual-output fluid sensors employing the structural design of thermoelectric materials (SDTMs) that are artificially patterned TMs. The SDTMs enable the successive thermoelectric waves as the raw voltage signals with two distinct peaks that can reflect fluid temperature and dynamics, in contact with working fluids. The 1st-peak voltage provides the precise sensing of fluid temperature, while the duration between 1st- and 2nd-peaks indicates the moving velocity. A flexible-detachable SDTM-based sensor comprising of pre-designed Bi_2Te_3 pattern between cellulose and PET substrates performs high-resolution sensing of temperature and velocity (< 0.19 K and < 0.03 cm/s) and facilitates the sticker-like functions through high-reproducibility ($> 93\%$) of sensing under transfers between flat and curved surfaces. Furthermore, a scalable sensor array (4-by-4 SDTMs array at 16 pixels) is developed as a large-area device for real-time detection of fluid temperature and dynamics at multiple positions, accompanying with self-power generation of $42 \mu\text{W}/\text{cm}^2$. The new methodology using SDTMs can contribute to developing next-generation sensors having advanced features, such as multi-detection and diversely integrated flexible-detachable functions.

1. Introduction

Sustainable monitoring of environmental or industrial elements through ubiquitous sensors is the most essential capability to advance the next-generation platform that enables self-adaptive function in response to the observation of minute changes, such as the Internet of Things (IoTs)-based applications [1,2]. To report real-time changes of monitoring elements everywhere, these platforms generally require a number of sensing devices, which are miniaturized and scattered in the system. Since IoT systems utilize various components, the multi-functional operation of the sensors is crucial to simplify the entire setup and to reduce the complexity [3,4]. Moreover, the functions of the individual sensing device are becoming more and more demanding due to the limitation of available spaces for installing sensors and the difficulty of power supply for widely distributed sensors in a large-area [5–7]. Meanwhile, the simultaneous monitoring of multiple elements allows the possibility to decrease the minimum number of sensing devices, while its flexible or detachable capability facilitates the installation or the integration of devices to complex structures in specific spots with restricted conditions [8,9]. For instance, dual-parameter sensors such as e-skin conduct multi-sensing of temperature and pressure [10], and

polymer based thermoelectric composites provide sustainable operation for temperature monitoring [11]. Moreover, the self-energy generation of sensing devices through the dynamics of the ambient conditions can provide sustainable operation for monitoring [12,13].

The sensing temperature and dynamics of working fluids, including water, is one of the most significant elements for environmental and industrial applications, such as detection of precise changes of fluid environment [14–16], monitoring effluence or cooling water in plant facilities [17,18], and flow analysis in micro/nanofluidic devices [19–21]. Furthermore, the recent researches have been advanced to have the self-power generation, as well as the sensing capability [22–25]. Hybrid-nanostructured piezoelectric composite can detect fluid velocity with self-powered operation [26], while electromagnetic-triboelectric hybridized nanogenerator are applied to fluid temperature sensor [27]. However, owing to the limitation of space or complex geometries in fluidic channels or interfaces, precise sensing of multiple elements with optimal integration to the operating platforms are still challenging issues. In addition, many fluid sensors inevitably disturb the natural flow in the platform, and may cause overall changes in the performance, despite the installation of the sensing devices should not affect the intrinsic characteristics of the targeted elements in fluidic

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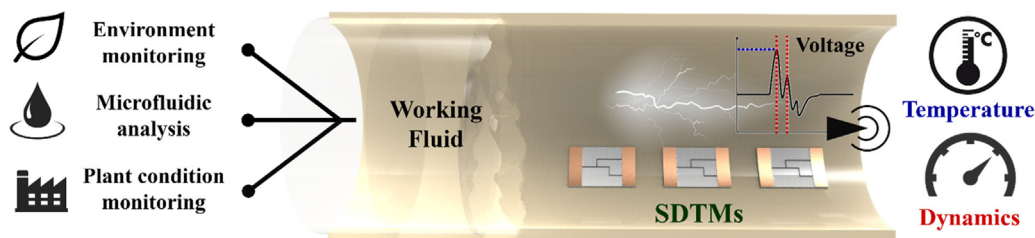


Fig. 1. Schematic of flexible-detachable dual-output sensors based on structural design of thermoelectric materials (SDTM) and applications to real-time monitoring of fluid temperature and dynamics in various systems.

applications [18]. These restrictions require a newly optimized design to set up the sensors at specific spots on the platform, as well as the multifunctionalities to monitor various environmental factors in a single device.

Herein, we propose a flexible-detachable dual-output sensor for fluid temperature and moving dynamics based on the structural design of thermoelectric materials (SDTMs), which are artificially patterned thermoelectric materials (TMs) arrays in flexible-detachable thin film layers (Fig. 1). When the working fluid contacts the layers, the structural pattern of TMs such as the ladder, produces successive thermoelectric waves with a slight time difference without any disturbance to the natural flow of the working fluid. The raw voltage signal induced by thermoelectric waves in SDTMs provides the magnitude of the first peak voltage and the duration between two peaks that can reflect the real-time temperature and the moving velocity of the working fluid, while the layered structures of SDTMs in the flexible-detachable substrates enable the development of sticker-type thin-film sensing devices. As a demonstration of the concept, thin film layers comprising pre-designed Bi_2Te_3 pattern between cellulose and polyethylene terephthalate (PET) substrates were prepared, and the dual-sensing capabilities were investigated in arbitrary surfaces of the fluid channels (flat and curved surfaces). The fabricated flexible-detachable sensors simultaneously detect changes in temperature and moving dynamics of water in high resolutions ($< 0.19\text{ K}$ and $< 0.03\text{ cm/s}$) through the analysis of the raw voltage signal. The working principles of SDTMs were verified in terms of the theoretical analysis and simulation, compared with experimental results. Furthermore, a scalable sensor array comprising multiple SDTMs (4 by 4 SDTM array) was fabricated and evaluated as a large-area fluid temperature-dynamics sensing device. It precisely produces different voltage signals reflecting the temperatures and dynamics of the working fluid on the 4 by 4 area (16 pixels) with high accuracy. The new methodology employing SDTMs can contribute to developing completely new techniques for next-generation sensors that require advanced features such as multi-elements detection and diversely integrated flexible-detachable functions.

2. Materials and methods

2.1. Fabrication of SDTM-based thin-film layers as dual-output sensors

A thin polyethylene terephthalate (PET) film (100 μm in thickness) was prepared as substrate for the SDTMs. The prepared PET film was cleaned using deionized (DI) water and a nitrogen blowing gun. Shadow masks (70 μm in thickness), which had various structurally-patterned designs, were made from thin film stainless steel (SUS 304). Target sources of bismuth telluride (Bi_2Te_3) (5 N purity) and copper (Cu) (4 N purity) were prepared for the SDTMs and electrodes, respectively. The active layer of the SDTMs comprising Bi_2Te_3 was deposited on the cleaned PET substrate through the shadow masks using RF magnetron sputtering technique at room temperature for 25 min with an input power of 100 W and pressure of 28 μtorr in 20 sccm argon flow. Copper electrodes were subsequently deposited using DC magnetron sputtering technique at room temperature for 1 h with an input power of 30 W (300 V, 100 mA) and pressure of 42 μtorr in 50 sccm

argon flow.

2.2. Characterization of fundamental properties of SDTM-based thin-film sensors

The electrical resistances of the SDTM-based sensors were measured using a multimeter (Fluke 1577). The characterization of the surface morphology of the deposited SDTMs in terms of the structural pattern, uniformity, and particle sizes was carried out using high-resolution scanning electron microscope (HR-SEM, FEI, Quanta 250 FEG). The elemental composition of the SDTMs was analyzed using X-ray diffraction (XRD) patterns (Rigaku, SmartLab). The atomic force microscope (AFM, NTEGRA Prima, NT-MDT) in the tapping mode was used to obtain the line profile and thicknesses of the individual layers of the SDTMs-based sensors.

2.3. Simulation of output voltage signals in response to fluid temperature and velocity

The output voltage signals from the SDTM-based devices were analyzed using the finite element method (FEM) through COMSOL[®] Multiphysics. To identify the correlation between the raw voltage signals and structural parameters of SDTMs, the real-time distribution of temperature, correlated with the specific transition of the signal was investigated in four subdivided steps depending on the location of the moving front of the fluid on the SDTMs sensor, which are the front-electrode, two junctions of patterned thermoelectric materials, and end-electrode. Experimentally-measured structural dimensions and parameters in thickness, length, and width were used to model SDTMs layers, while the basic properties, such as specific heat, thermal and electrical conductivity were obtained from other literatures. Meanwhile, Seebeck coefficient of Bi_2Te_3 in the SDTM-based device was experimentally measured by four-probe method. The three-dimensional, transient thermal conduction equation and Seebeck effect equation were applied to estimate voltage signals under a natural convection boundary condition.

2.4. Measurement of output voltage signals from SDTM-based sensors

A custom experimental set-up was designed to measure the sensing performances of the SDTM-based sensors. The fluid path and reservoir were made of acrylic-based plates, which were treated by pre-processes to remove static electricity. DI water was used as the working fluid to evaluate sensing performances. An oscilloscope (Tektronix, DPO2004B), which was attached to the electrodes of the SDTM-based sensors, measured real-time changes in the output voltage signals from the SDTMs in the denoise mode as the working fluid (DI water) passes through the devices. A high-speed charge-coupled device (CCD) camera (Phantom V7.3-8 GB) simultaneously observed the dynamic flow and velocity of the working fluid. For comparison, a thermocouple (Giltron, GTPK-01) fixed on the SDTM-based sensors recorded real-time changes in the temperature as the fluid passes through the surfaces. The overall temperature distribution on the SDTMs and the devices was obtained using a thermal infrared (IR) camera (FTIR, T-420).

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