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High-resolution flexible temperature sensor based graphite-filled polyethylene oxide and polyvinylidene fluoride composites for body temperature monitoring

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ABSTRACT

Flexible and high accurate temperature sensors have great potential toward applications such as medical diagnosis and body temperature monitoring. Here, we demonstrated a flexible temperature sensor based on graphite-filled polyethylene oxide (PEO) and polyvinylidene fluoride (PVDF) composites exhibiting a high accuracy of 0.1 °C and high repeatability nearly 2000 times, in the sensing temperature range of 25–42 °C. Device performance is hardly affected in different curvature, which allows for conformal application to human skin. Especially, subtle temperature change on the skin surface was also measured to validate the accuracy and anti-interference ability. Under physiological conditions, the temperature sensor based on positive temperature coefficient showed a high-speed response time of 26 s with relatively simple fabrication technology. These results demonstrated the possibility and feasibility of fully using the sensors in body temperature sensing for medical use as well as sensing function of body temperature measuring.

1. Introduction

Body temperature is one of the most concerned physiological parameters in physiology, which is closely related to the chemical reaction in the physiological system [1–3]. Besides, body temperature plays a very important role in chronobiology study, predicting disease, and monitoring postoperative recovery [4,5]. Monitoring the temperature of human skin is required to be highly precise, easy-to-operate, soft, flexible and biocompatible, which draws a critical challenge. In traditional medical care, skin thermography was achieved by either sophisticated infrared digital cameras for spatial imaging or mercury thermometer for measuring body temperature. Infrared cameras can provide ultrahigh precision and fine resolution in imaging, but the drawbacks come with the high cost and immobilization of patients [6]. As for mercury thermometer, it is inconvenient for new-born babies or patients in anesthesia because they cannot stably keep the mercury thermometer in underarm as required [7,8]. What's more, mercury thermometer is prone to be shattered and the leaking mercury is poisonous and will pollute the environment.

Therefore, comfortable, biocompatible and flexible temperature sensors used as wearable electronic device [9–11] with satisfactory accuracy are gathering numerous interest in healthcare and medical applications. Over the past decades, many efforts have been made in soft and biocompatible temperature sensors, such as thermocouple sensors [12–14], thermo-resistance temperature sensors [15–17] and organic diodes temperature sensors [18–20]. Webb et al. introduced a temperature sensor with an ultrathin, compliant, skin-like array to provide continuous, accurate thermal characterizations of human skin [6]. Biodegradable and highly deformable temperature sensor based on thin Mg film and ecoflex could stay high mechanical stabilities when the devices were crumpled, folded, and stretched up to 10% [21]. In addition, Wu et al. developed a flexible and polysilicon-based temperature sensor with structural flexibility for brain temperature monitoring [22]. Kim et al. reported a single crystalline silicon nanoribbon doped twice to form p-n junctions as a temperature sensor [23]. More advanced is the composite materials made by doping functional materials into polymer matrix as temperature sensor to detect temperature through resistive changes. Bao et al. developed a flexible temperature sensor based on Ni microparticle-filled polymer com-

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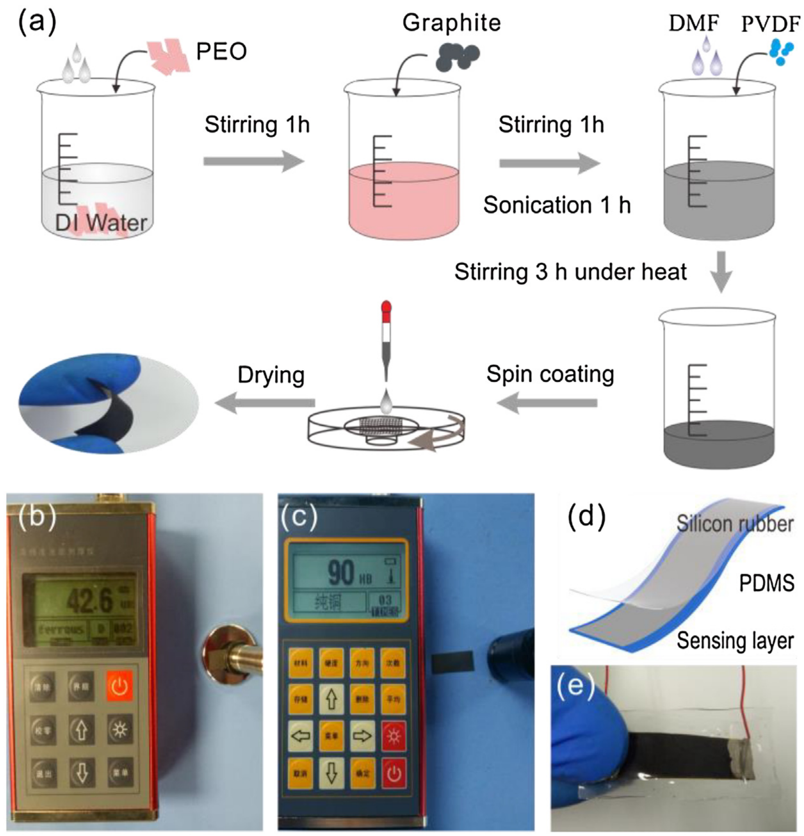

Fig. 1. (a) Schematic illustration of the fabrication process of the temperature sensor. (b) Thickness of the temperature sensing layer. (c) Hardness of the temperature sensing layer. (d) Schematic diagram of the sandwich-structured temperature sensor. (e) The photograph of the fabricated samples.

posite with high sensitivity to monitor body temperature, using an RFID antenna system [24]. Yokota et al. designed ultra-flexible temperature sensors based on copolymer with graphite filler, which can exhibit changes in resistivity by six orders of magnitude or more for changes in temperature of only 5.0 °C or less [25]. Although many eff ;orts have been made to fabricate various kinds of temperature sensors, none of them have excellent anti-interference ability and high resolution in the temperature range of 34.0–42.0 °C, which is the crucial range for the human body [26].

In this work, we presented a flexible and biocompatible temperature sensor with high resolution and high repeatability in the temperature range of 34.0 °C–42.0 °C fabricated by integrating temperature sensitive material with polydimethylsiloxane (PDMS) covering layer and silicon rubber substrate. Meanwhile, the device exhibits excellent anti-interference ability, high repeatability near body temperatures and prominent durability, which enable a wide range of medical and nonmedical devices. And subtle temperature change on the skin surface was also measured to validate the accuracy and anti-interference ability. In addition, the fabrication technology is relatively simple, low-cost, and processable, which make mass production possible.

2. Experimental details

2.1. Materials

Graphite powder (Gr) was provided by Sigma-Aldrich. *N,N*-Dimethylformamide (DMF) was obtained from Aladdin. Polyvinylidene fluoride (PVDF) was bought from Micxy Reagent. Polyethylene oxide 1500 (PEO1500), polyethylene oxide 6000 (PEO6000) and polyethylene oxide (PEO, Mv ~5,000,000) (PEO5000K) were purchased from Sinopharm Chemical Reagent Co., Ltd. All these reagents were used without further purification.

2.2. Fabrication of temperature sensor

The temperature sensor was fabricated by the following procedure (Fig. 1(a)): First, the PEO1500 was dissolved in the DI water using a magnetic stirrer for 1 h. Next, graphite powders were added to above solution followed by sonication for 1 h and magnetic stirring for 1 h, respectively. After that, PVDF and DMF were introduced and mixed for another 3 h under heat. The weight ratio of graphite, PVDF and PEO1500 is 4:3:3. The PEO1500/PVDF/Gr composite solution was dropped on the Polyimide flexible substrate and then coated uniformly using spin-coating for 15 s at a spinning speed of 50 rpm. For thermal annealing, the sample was annealed at 45 °C for 2 h and the DMF solution was removed. After the solution was dried, the dried composite was easy to remove from polyimide tape without any adhesion, so that the neat composite without polyimide tape could cut into specific dimensions for further fabricating. Then the PEO6000/PVDF/Gr composite and the PEO5000K/PVDF/Gr composite were fabricated by the same way.

As shown in Fig. 1(b) and (c), the thickness of the temperature sensing layer is 42.6 μm, and the hardness is 90 HB. The sandwich-like structure of our temperature sensor is shown in Fig. 1(d), which was composed of PDMS covering layer and temperature sensing layer on a silicon rubber substrate through layer by layer assembly. The sandwich-structured temperature sensors were fabricated by the following procedure: First, to obtain a silicon rubber substrate with the approximate thickness of 0.5 mm, the liquid silicon rubber was deposited on the silicon wafer by spin-coating for 2 min at a spinning speed of 500 rpm, and then cured in 70 °C for 2 h. Then copper wires were attached to the two ends of the temperature