



Design and fabrication of a new nonwoven-textile based platform for biosensor construction



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ABSTRACT

This study focuses on the fabrication of a novel, flexible and disposable textile based biosensing platform by the use of an absorbent microfibrinous nonwoven substrate as the base material. This platform was fabricated via photolithography technique. Physical barriers were designed using a hydrophobic photo-resist polymer which defined the liquid penetration pathways on the fabric surface. A good hydrophilic/hydrophobic contrast of the fabricated patterns on the fabric and a well-controlled liquid capillary penetration was achieved in the patterns. The potential of the system was tested by constructing an enzyme biosensor based on colorimetric detection of hydrogen peroxide. To obtain a more enhanced and reproducible signal, the reservoirs were modified with gelatin and a linear working range of 0.1–0.6 μM H_2O_2 was obtained. The system could work on temperatures as high as 50 °C without any loss in the signal and in a pH range of 3.0–7.0. This bio-sensing platform may later be combined by H_2O_2 producing oxidases such as glucose oxidase, lactate oxidase, etc. and used for the rapid detection of various kinds of important analytes.

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1. Introduction

Microfluidic analytical devices have been successfully used in the area of health related diagnosis especially in developing countries. They possess advantages of being inexpensive, easy to use, lightweight and low cost [1,2]. These devices are capable of detecting very small molecules such as glucose, lactose, alcohol, cholesterol, uric acid, antibody and antigen [3,4]. For their production, different fabrication techniques such as photolithography, inkjet printing, wax patterning and plasma treatment of hydrophobic paper have been used [5]. Etching or molding patterns into glass, silicon, PDMS (polydimethylsiloxane), PMMA (poly(methyl methacrylate)), SU-8 (an epoxy-based negative photoresist) or other polymers or plastics have been traditionally applied for the

fabrication of microfluidic devices. Alternatively their paper-based counterparts have been developed since they combine some of the capabilities of conventional microfluidic devices with the simplicity of diagnostic strip tests [6–10].

Whitesides and co-workers were the first to propose patterned paper as a paper-based microfluidic diagnostic platform [6]. Several methods related to fabrication of paper-based diagnostic devices for clinical assays have been reported. Martinez and co-workers [11,12] fabricated physical barriers by photolithography using a hydrophobic photo-resist polymer which defined the liquid penetration pathways on the paper surface. Transportation of liquid sample to multiple detection zones along the defined pathways could be achieved on the paper surface and multiple analytes could be detected simultaneously [11,12]. Photolithography was also used by Dungchai et al. in the design of a paper-based microfluidic device. In the detection zones of the device, multiple indicators were used for the analysis of different concentrations of an analyte [13]. The same group used photolithography technique to create microfluidic channels on the surface of filter paper and electrodes were fabricated using screen-printing technology. The utility of the microfluidic device was then demonstrated with the electrochemical detection of glucose, lactate and uric acid in biological samples

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[14]. Bruzewicz et al. fabricated barrier patterns via printing PDMS onto a paper surface using a modified plotter. Colorimetric assays were conducted on the paper based devices for glucose and protein analysis. The main disadvantage of system was the low resolution of channels [15]. Carrilho et al. reported a wax printing method using a printer and a hot plate for fabricating paper-based microfluidic devices. A printer was used to print patterns of solid wax on the surface of paper and wax was melted using a hot plate to provide penetration of wax through the full thickness of the paper. However, patterns made by wax printing technique showed low resolution compared to photolithography technique [16]. The same technique was used by Lu et al. to produce patterned paper to fabricate low-cost and flexible PDMS microdevices for the detection of analytes such as glucose and proteins [17]. Li et al. reported the use of paper sizing agents, i.e. alkyl ketene dimer and alkenyl ketene dimer, and ink jet printing to fabricate paper-based chemical and biological sensors at low cost [18]. Zhang et al. reported the fabrication of hydrophilic channels on a paper base material using printed circuit technology. Colorimetric assays including protein and glucose analyses were conducted using this paper based device [19].

The fabric-based fluid handling platforms, being more flexible and wearable alternatives to paper-based devices, were studied as potential biosensors to perform on-body bio-chemical analysis [20–25]. A body fluid such as sweat, blood etc., usually used as analysis sample, was delivered to the sensing device's active surface where a reaction took place resulting in a signal or color change [20,26–28].

Coyle et al. developed a fabric-based fluid handling system having advantages of no power requirement for the liquid transport, compact structure and easy fabrication. For that, a fabric channel system was made from a moisture wicking fabric and a super-absorbent material was embedded at the end of the channel to control the flow of fluid through the channel. In addition to this, a lateral flow valve was used to control the transmission of liquid sample to the channel system [25]. An analysis system was developed comprising different textile sensors distributed around the body to measure and perform real-time analysis of the pH, sodium concentration and conductivity of sweat, sweat rate, ECG and respiration during exercise. Miniature optical components could also be incorporated to the microfluidic devices as a detection system [20,21,29,30].

Unlike the sensors using glass and silicone as substrates, fabric based sensors do not require pumps or power sources to move fluids through the microfluidic device due to their ability to wick fluids by capillary action [31]. Fabric as a substrate offers a more efficient liquid sample delivery, i.e. wicking rate than paper by the help of the capillary action which is necessary for a rapid detection of analytes [32]. The wicking rate of material used for microfluidic diagnostic device is very critical since the sample fluid should wick along the channels on the material as quickly as possible to minimize the sample evaporation during transport which may lead to ineffective sample delivery within the microfluidic device [31,33]. If the distance from the introduction point of the analyte sample to the detection zone is considerably far, then during solution spreading, the local analyte concentration may decrease limiting the sensitivity of the device [34]. In addition, in the proposed nonwoven based sensor, during patterning the isotropic structure of the nonwoven substrate provides an even spreading of photoresist polymer in x,y -plane as well as z direction through the entire fabric thickness which is critical for creating a well-demarcated hydrophobic-hydrophilic contrast of the pattern. The two-dimensional orientation of fibers in x,y -plane of a sheet of paper has been reported as a constraint since the liquid polymer tends to spread in the x,y -plane of paper rather than in the z -direction leading to blurring of the patterns [12,31,33,34]. A good

hydrophobic-hydrophilic contrast of the pattern is also required to avoid the leakage of the fluid consisting of the analyte through the channels and spreading through the device.

In the current study, a novel, flexible, wearable, easy-to-use and disposable textile-based micro fluidic platform was designed and developed. For this purpose, a highly absorbant nonwoven textile fabric was used which allowed the collection and delivery of fluids through the micro channels to the reservoir zones without any need to an additional pumping system. Baysal and Önder et al. reported that the nonwoven fabric provided a wearable microfluidic platform and also it was superior to paper in collecting and delivering the sample fluid to the detection zone, which is necessary for the rapid detection of analytes [32]. A good pattern resolution which is necessary to prevent liquid leakage through the patterned channels and reservoirs could be achieved in the current study.

The traditionally used microfabrication materials like glass and silicon are rigid and expensive therefore plastics such as PDMS (polydimethylsiloxane) and PMMA (polymethylmethacrylate) have been utilized for the fabrication of microfluidic devices. However, their hydrophobic structure does not allow simple capillary flow, therefore sophisticated and expensive readers to give a read-out and separate pumping systems that can direct fluid flow are required [35,36]. Fabric based microfluidic devices works with the principle of using capillary force to wick and distribute fluid into hydrophilic channels without external pressure or control systems [36]. They require small amount of analyte sample solutions, a few microliters, to perform chemical and biochemical analyses [37].

The potential of the developed microfluidic system was tested by constructing a hydrogen peroxide (H_2O_2) biosensor based on colorimetric detection. H_2O_2 plays an important role in chemical, biological, environmental, food and industrial analysis and used to control pollution and to bleach papers, textiles and paper products, and is an essential mediator in food, medicine [38–41]. Horseradish peroxidase enzyme used in this study could also be coupled by oxidase enzymes to construct biosensors for various important analytes [42–44]. Detection of H_2O_2 amount with high selectivity and sensitivity is crucial for all these applications. To the best of our knowledge, this is the first study demonstrating the fabrication of a microfibrinous nonwoven-based microfluidic platform established by photolithography technique with potential use in biosensing applications. The platform could potentially be used as a sensing platform for the rapid detection of various kinds of analytes of medical and industrial importance.

2. Experimental

2.1. Materials

Freudenberg's Evolon[®] nonwoven fabric was used as the basis for the microfluidic platform. Evolon[®] is a spun bonded and hydroentangled bicomponent nonwoven fabric made from split polyester/nylon microfibers. For the production of the nonwoven fabric, polyester and polyamide polymers are spun into endless segmented filaments which are uniformly laid on a moving belt forming the nonwoven web. Subsequently, bonding and consolidation of the nonwoven web is achieved by high pressure and high speed water jets, the so called hydroentanglement process, during which the filaments are split into microfilaments. Due to its microfibrinous structure, the fabric becomes highly absorbent. The hydroentanglement process results in a three dimensional and dense fabric structure. In this study, Evolon[®] fabric was preferred for its high absorbance capacity to collect and deliver the fluids through the fabricated micro channels to the reservoir zones.

Evolon[®] fabric combines very good textile and mechanical properties with advantages of being soft, drapable and lightweight. The average breaking strength of a 130 g/m² Evolon[®] fabric was

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