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Micromechanical properties of biomedical hydrogel for application as microchannel elastomer

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ABSTRACT

Polymers are believed to be the building blocks for the creation of the next generation of materials and devices in practically all areas of biomedical research. There are a number of polymers that are being employed in varied applications in microfluidic platform due to the tremendous possibilities for soft matter based elastomers especially in biomedical applications. Polymeric hydrogels have been used as building block in micro-confinements and for specified function such as flow control. The need exists to suitably determine the mechanical characteristics of gel-based materials for possible use as a microchannel elastomer. In this investigation, we describe synthesis procedure, morphological, wettability characterization of hydrogel elastomer synthesized by free-radical polymerization crosslinked over varying acrylamide composition for 10% w/v: 25% w/w, 15% w/v: 25% w/w, 20% w/v: 25% w/w and 25% w/v: 25% w/w respectively. Micromechanical properties such as surface morphology, wettability, and micro-rheological behaviour of hydrogel elastomer using standard protocols was undertaken to determine roughness, contact angle, loss modulus and storage modulus over varied cross-linking of the constituent monomers. The impact of these parameters on flow transport and microchannel structural stability is well delineated in this report. We established that polymeric hydrogel could be a candidate for whole microchannel elastomer with suitable application in areas of tissues and biomedical engineering to mimic native biological transport conduits.

1. Introduction

Polymers have been extensively employed in microfluidic platforms leveraging on the advancement in micro-electromechanical systems (MEMS) (Liu, 2007). There are a variety of classes of polymers that have been employed as elastomers in several microfluidic-based investigations. Fakunle and Aguilar (2006) reported the use of organic and inorganic materials like ceramics in the forms of co-fired ceramics (CFC), vitro-ceramics and polymers in microfluidics. In the recent times, polymer-based materials have been well patronized for microfluidics applications. Yu and Shi (2015) fabricated 2D and 3D microfluidic paper-based analytical devices (μPADs) by photolithography-patterning microchannels on a parafilm and subsequently embossing them to paper.

Poly dimethyl siloxane (PDMS) is a household name amongst polymers employed in microfluidics owing to the ease of synthesis from the pre-polymer solution made by proportionate cross-linking of sylgard 184 and curing agent. The reason for increasing patronage and popularity of PDMS as the material of choice for microfluidic devices is due to its low cost, ease of fabrication, oxygen permeability and optical transparency (Markov et al., 2015). PDMS possesses inherent mechanical characteristics such as low mechanical properties with Young's modulus of 1.32–2.97 MPa, ultimate tensile strength of 3.51–7.65 MPa, compressive modulus of 117.8–186.9 MPa and ultimate compressive strength of 28.4–51.7 GPa. Johnson et al. (2014). PDMS has affinity for small hydrophobic molecules and thus could lead to biomolecule absorption/adsorption from the medium, thus biasing the experimental condition. The permeability of PDMS to water vapor can also lead to media drying and thus change its osmolarity. These issues have limited the application in a number of biomedical-based investigations. To this end, there have been reports on several alternate elastomers which could be near-native biological tissues to enhance reliability investigation in microfluidic-based platforms.

Hydrogels have been used in microfluidic platforms since its mention as flow control device in microfluidic channel. For instance, (Eddington and Beebe, 2004) as well as (Casson and Lutolf, 2014)

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employed hydrogel as a building material for precise biomolecule delivery in microfluidic set-up. Cheng et al. (2007) used hydrogel for biological experiments because of its inherent ability to respond to chemical stimuli. Hydrogels are a class of polymer that could be derived from synthetic and natural materials. Synthetic materials include poly (ethylene oxide) (PEO), poly (vinyl alcohol) (PVA), poly (acrylic acid) (PAA), poly (propylene furmarate-*co*-ethylene glycol) (P(PF-co-EG)), and polypeptides. Naturally derived gel-based polymers are agarose, alginate, chitosan, collagen, fibrin, gelatin, and hyaluronic acid (HA) (Drury and Mooney, 2003). Poly(acrylamide-*co*-acrylic acid) hydrogels are relatively easy to produce, more so, reports have mentioned that it could represent a useful matrix for analytical and synthetic surrogate for biological tissues (Faraji et al., 2011). There are a number of tissue engineering applications that have favoured the use of hydrogel because of its biocompatibility. Such applications include tissue engineering matrices, wound dressing, skin recovery and drug transport etc (Elabadawy and Xin, 2017; Caccavo et al., 2015). The intrinsic property of hydrogel such as its modulus of elasticity or stiffness which has been reported as a characteristic of its extracellular matrix alludes to its applications as cell-dependent anchorage (Pompe et al., 2009). Hydrogel has been extensively utilized as extracellular matrix fiber for micro-architecture adventitia of large blood vessels in a richly vacularized micro-environment (González-Díaz and Varghese, 2016). However, polymeric hydrogel is yet to be fully employed as whole elastomer for microchannel despite its excellent biocompatibility criteria, ease of synthesis and cost benefit among others owing to the challenge of fabricating microchannel-like hydrogel structures (Hammer et al., 2013). In the parlance of tissues engineering, vascular constructs are regarded as microchannels. Hence, techniques for vascularization of biocompatible structures such as hydrogel remain a subject of interest because of the prospects of fluid-carrying vessels in applications of biomedical engineering (Bae et al., 2012; Bertassoni et al., 2014). Several authors have employed strategies for fabricating gel-based conduits. You et al. (2016) fabricated cell-laden hydrogel based three dimensional constructs using 3D bio-printing technology. Yang et al. (2016) attempted to fabricate multi-layer vascular chip by utilizing 3d technology to cast hollow L-shaped microchannel constructs for use as vascular conduits which was incorporated with native umbilical vein endothetlial cells. Zhang et al. (2015) addressed the challenge of fabricating synthetic vascular fluidic conduits by employing projection-based stereolithography to construct 3D biocompatible hydrogel. Du et al. (2011) applied single and cost-effective layer-by-layer sequential technique to assembly hydrogel constructs with an embedded microchannel using photo-lithography processes. Lee et al. (2016) employed sacrificial template to fabricate 3D microvascular channel in a cell-populated environment using Poly(N-isopropylacrylamide) based hydrogel based on its credible thermoresponsive attribute.

Surface roughness as a determinant of flow in microchannels have been well investigated (Fanning, 1877; Fercana et al., 2017). Because of the practical importance of surface roughness in experimental procedure, several techniques have been proposed to measure this property. Kandlikar et al. (2003) employed optical method based on reflected beam intensity profile of the He-Ne laser beam and a fiber optic probe for detection. Rapid optical system based on eximer laser and beam profiler for nano-scale poly-Si thin film surface roughness measurement was introduced (Kandlikar et al., 2005a). The result was obtained with error to be less than 2.1% and the measurement time was shortened by up to 83%. The correlation between surface roughness and fluid flow was first illustrated by the submission of Darcy in the nineteen century that pressure drop and surface roughness are important factor in fluid flow (Ren et al., 2011). In a latter report, Fanning re-affirmed the same conclusion on surface roughness and pressure drop in pipe (Kandlikar and Schmitt, 2005). There are several reports on this subject such as (Jaeger et al., 2012; Nikuradse, 1933) that corroborated the significance of friction factor and relative roughness. The famous Moody's chart was based Colebrook equation and Nikurdase work with relative roughness in the range (Colebrook, 1939). Kandlikar (2005b) reassessed the experimental findings of Nikuradse's with special interest on roughness at micrometer scale. Hence, these studies underscored the importance of conduit roughness in flow transport. There are several techniques employed for measurement of surface roughness such as optical or scanning electron microscopy, profilometer, digital holographic microscopy (Moody, 1944) and Atomic Force Microscopy (AFM). In most reports, AFM is preferred over profilometer because the application of the latter results to localized damage to the surface being examined (Montifort et al., 2006). The impact of surface roughness on flow conduits becomes critical with reduction in channel dimension (Sundararajan et al., 2005; Young et al., 2009). At micron-seized length scale, surface roughness is characterized by short wavelengths presented in both amplitude parameters and spatial parameters which forms the basic parameter illustrated in AFM-based analysis. In addition to surface roughness, micro-rheological information of the flow confinement such as storage and loss modulus is being investigated in this work. This is to provide insight to the effect of monomer cross-linking on the compliant (flexible) nature of elastomers used as flow conduits. The wettability effect of polymerization processes on elastomer surfaces with variation in monomer crosslink is presented in this report.

In this study, attention is directed to acrylamide-based hydrogel elastomer and characterization procedures of micromechanical properties of polymeric hydrogel elastomer are delineated for the purpose of fluid transport applications in micro-confinements. This investigation contains presents detailed description of standard protocols for parameters like micro-rheology, surface roughness and wettability that could impact on microscale flow transport. The properties of flow conduit such as surface roughness, contact angle and micro-rheology is important for consideration of elastomer especially in a microfluidic platform. These properties provide the information that could predict possibility of resistance to flow transport on elastomer with specified acrylamide composition which is the proposed benefit of the present investigation.

2. Materials and method

2.1. Synthesis of polymeric hydrogel

There are three stages of the free-radical polymerization procedure employed in this study. The first stage is the synthesis of linear polymer with functional side group; the base monomer reacts with the carboxylic functional group in acrylic acid to form a linear polymer with the carboxylic group as side attachment. In the second stage, cross-linking reaction takes place where a crosslinker agent which is methylene-bis-acrylamide (BIS) reacts with the functional group attached to the side of the linear polymer in the previous stage. A well cross-linked polyacrylamide-based hydrogel polymer matrix is formed when initiators and accelerators are added. At the third stage un-reacted compound and solvent in the network are removed. The equation of chemical reaction of the monomers and the schematics of the polymerization process is highlighted in Fig. 1.

Using the above concept, hydrogels were produced using AAc and BIS as based monomers with APS and TEMED as initiators. Hydrogel compositions used were 10% w/v: 25% w/w, 15% w/v: 25% w/w, 20% w/v: 25% w/w and 25% w/v: 25% w/w. These cross-linker ratios when denoted as the percentage weight of monomers (AAm and BIS) to deionized water (DI water) is written as % w/v and when denoted as percentage of solute monomer to weight of AAc present in hydrogel matrix is written as % w/w. These cross-linker ratios are in reference to volume of deionized water and weight of AAc present in the hydrogel matrix. The initiators employed in acrylamide-*co*-acrylic acid hydrogel were ammonium persulfate (APS) and Tetraethyl dimethyldiamine (TEMED). The concentrations of these reagents used for the entire synthesis of hydrogel are 0.02 nM and 0.03 mM of APS and TEMED respectively. The concentration of APS used was prepared by diluting