2012

Bucky gel actuator for morphing applications

Ali Kadkhoda Ghamsari

Louisiana State University and Agricultural and Mechanical College, akadkh2@lsu.edu

Follow this and additional works at: https://digitalcommons.lsu.edu/gradschool_dissertations

Part of the Mechanical Engineering Commons

Recommended Citation

https://digitalcommons.lsu.edu/gradschool_dissertations/910

This Dissertation is brought to you for free and open access by the Graduate School at LSU Digital Commons. It has been accepted for inclusion in LSU Doctoral Dissertations by an authorized graduate school editor of LSU Digital Commons. For more information, please contact gradetd@lsu.edu.
BUCKY GEL ACTUATOR FOR MORPHING APPLICATIONS

A Dissertation

Submitted to the Graduate Faculty of the
Louisiana State University and
Agricultural and Mechanical College
in partial fulfillment of the
requirements for the degree of
Doctorate of Philosophy

in

The Department of Mechanical Engineering

By
Ali Kadkhoda Ghamsari
B.S., Amir Kabir University of Technology, 2004
M.S., K. N. Toosi University of Technology, 2007
August 2012
To my wonderful wife,

Amanda

For all her love and support
ACKNOWLEDGEMENT

First and foremost, I would like to express my deepest gratitude to my great parents; Hossein and Azam Ghamsari, for their unconditional and never-ending love and selflessness. Without their sacrifices, I would have not been able to reach this level of education. I am also very grateful for my two brothers, Navid and Kamran, and Sister, Nina, who are always loving and encouraging.

I would like to extend my gratitude to my advisor Dr. Eyassu Woldesenbet for his patience, guidance, and also believing in the scientist in me. Every single conversation of ours was inspiring for me to be a better researcher and above that a better person.

I am very grateful to my co-advisor Dr. Yoonyoung Jin for his expertise and support, for expanding my perspectives and patiently guiding me through my research. Also, for his great help in nano-indentation and gold-coating process.

I would like to thank my committee members Dr. Glenn Sinclair, Dr. Sunggook Park, and Dr. David Spivak. I appreciate the thoughtful suggestions I have received from them in conducting this study.

I would also like to thank my colleagues and friends. I am thankful to my research colleagues Ephraim Zegeye, Emmanuel Gikunoo, Fareed, Dawan, and Tamara Alexander for their great help throughout my research. I thank Dr. Manu John for his help in DMA testing. I specifically would like to thank my dear colleague Mesfin Terefework for his invaluable
assistance in fabrication process and displacement measurements. I would like to acknowledge Tige Brown for his help with providing equipment which were essential to this research. I deeply thank Breanna Mcquarter, Rafeal Amin, Roderick Keelen, Damien Lewis, for their valuable assistance in image processing, force measurements, CNC machining, and 3D printing, respectively. Special thanks to my dear friend; Mohammad Fesanghary, I appreciate his help with modeling and data analysis. I also have to acknowledge CAMD for providing me with the equipment necessary for flow rate measurements.

I would like to thank my dear friends, Pratap Bhat and Jennifer Robinson, for helping me with getting adapted to the new environment and culture here, and for making my first months of staying in Louisiana as pleasant as it could be. Also, I need to thank my dear friends Hamed Habibi, Ali Takbiri, and the Iranian students’ community for making Baton Rouge feels like my second home.

Above all, I am grateful for the love and support of my wife, Amanda. For standing beside me through good and bad times, and for believing in me, even when I didn’t. I always admire her passion for helping others and her dedication to perfection.

None of this was possible without you.
# TABLE OF CONTENTS

Dedication ....................................................................................................................................... ii

Acknowledgements ........................................................................................................................ iii

List of Tables ....................................................................................................................................... vii

List of Figures ...................................................................................................................................... viii

Abstract ........................................................................................................................................... xii

Chapter 1. Introduction ................................................................................................................... 1

Chapter 2. Materials and Fabrication .............................................................................................. 8

Chapter 3. Mechanical Characterization ....................................................................................... 14
  3.1 Characterization methods ................................................................................................... 14
    3.1.1 Dynamic Mechanical Analysis (DMA) ...................................................................... 14
    3.1.2 Nano-indentation ........................................................................................................ 16
  3.2 Characterization result and discussion ............................................................................... 19
    3.2.1 Storage modulus and viscosity ................................................................................... 20
    3.2.2 Hardness...................................................................................................................... 27
    3.2.3 Adhesion ..................................................................................................................... 29

Chapter 4. Displacement measurement ......................................................................................... 33
  4.1 Designed experiments ........................................................................................................ 33
    4.1.1 Displacement measurement ...................................................................................... 34
    4.1.2 Lifetime and BGA degradation................................................................................... 35
  4.2 Results and discussion........................................................................................................ 36
    4.2.1 Voltage and Frequency ............................................................................................... 36
    4.2.2 Weight fraction of constituents................................................................................... 40
4.2.3 Thickness, Thickness ratio ................................................................. 40
4.2.4 Lifetime ............................................................................................. 42
4.2.5 Degradation over time ................................................................. 44
4.3 Maximum displacement model .................................................... 45

Chapter 5. BGA Application in microfluidics ........................................... 53
  5.1 Different actuation mechanisms in microfluidics ............................. 53
  5.2 BGA morphing diaphragm for possible application in micropump .... 55
  5.3 BGA microvalve .................................................................................. 56
    5.3.1 Design and modeling ................................................................. 62
    5.3.2 Fabrication ................................................................................ 65
    5.3.3 Experimental setup ................................................................. 67
    5.3.4 Results ....................................................................................... 71

Chapter 6. Conclusions and Future Work ............................................... 74
  6.1 Conclusions ..................................................................................... 74
    6.1.1 Mechanical characterization ................................................... 74
    6.1.2 Displacement study ................................................................. 75
    6.1.3 BGA application in Microfluidics ........................................... 76
  6.2 Future work ..................................................................................... 76
    6.2.1 BGA coupled with photovoltaic cells ...................................... 76
    6.2.2 BGA as an active micromixer ............................................... 77
    6.2.3 Electroactive actuator employing Deep Eutectic Solvents ...... 77

References ............................................................................................... 78
Appendix A. Permission to reproduce published work .......................... 86
Vita ............................................................................................................ 87
LIST OF TABLES

Table 1 Weight fractions of constituents in different sets of samples. .......................... 12
Table 2 Setting used for Dynamic Mechanical Analysis tests. .................................. 16
Table 3 Nano-indentation data for each electrode set. ............................................. 27
Table 4 Work of adhesion (JKR, DMT and Johnson theories). ................................. 30
Table 5 Thickness, Thickness ratio, and layer combination of the tested samples......... 37
Table 6 Average displacement of the BGA samples days after first test. .................... 43
Table 7 Average, standard deviation, and coefficient of variation of BGA samples at different driving voltages over 71 days................................................................. 45
Table 8 Material constants obtained for different reported result of BGA displacement. .... 48
Table 9 The assumptions used for comparing the model prediction to other reported results. ... 50
Table 10 Parameters used in gold-coating process. .................................................... 55
Table 11 MP6 micropump technical information [92].............................................. 69
Table 12 Sampling time selected for different frequencies....................................... 71
Table 13 Output flow rate measured at different voltages and frequencies................. 72
LIST OF FIGURES

Figure 1 (a) Schematic of BGA bending motion (b) BGA strip bended toward one side as the result of applied voltage during experiment................................................................. 3

Figure 2 Ionic conductivity of electrolyte layers containing various ionic liquids (modified from [12]). .......................................................................................................................... 8

Figure 3 Fabrication procedure (a) Spex SamplePrep Mix/Mill 8000M used in ball-milling process (b) ball-mill vial and the two Zirconia Ceramic ball (12.7 mm) (c) Output mixture in the ultrasonic bath (d) Degassing chamber. ................................................................................................................ 9

Figure 4 (a) bucky gel mixture after ball-milling process (b) BGA mixture casted in silicon rubber molds.......................................................................................................................... 10

Figure 5 BGA sample after being hot-pressed, showing good flexibility........................................ 11

Figure 6 Different layer combinations used in this study (Black: electrode layers – White: electrolyte layers), 1-2-1 layer combination was only used for displacement study. ............... 13

Figure 7 Regular SWMT bucky gel electrode layer casted on glass substrate for nano-indentation tests................................................................................................................................. 13

Figure 8 (a) Rheometric Scientific RSA III DMA used in characterization study (b) an electrode layer failed during frequency sweep. ............................................................................... 15

Figure 9 (a) Hysitron TI-900 Triboindenter utilized in nano-indentation characterization (b) indentation stage and the Berkovich tip used in this study. ...................................................... 17

Figure 10 Load function used in nano-indentation tests. ............................................................... 18
Figure 11 (a) Frequency sweep of a regular SWNT electrode layer (b) Frequency sweep of an electrolyte layer. ............................................................................................................................ 21

Figure 12 (a) Storage modulus of different electrode layers via frequency sweep at 0.5% strain (b) Loss modulus of different electrode layers at 0.5% strain. .......................................................... 22

Figure 13 Frequency sweep of (a) Regular BGA and (b) COOH-MWNT BGA at different strain level. .............................................................................................................................................. 23

Figure 14 Frequency sweep of Regular SWNT and COOH-MWNT BGA at (a) two different thicknesses (b) unit thickness. ....................................................................................................... 25

Figure 15 Complex viscosity of (a) different electrodes and electrolyte layers at 0.5% strain (b) SWNT and MWNT BGAs at 1.0% strain. ........................................................................................................... 26

Figure 16 (a) Hardness vs. Contact depth for Reg SWNT (2000 µN), 12% SWNT (2000 µN) and the 55% IL (1000 µN) (b) Hardness vs. Contact depth for MWNT at two different maximum loads. ......................................................................................................................................................... 29

Figure 17 SEM image of the Berkovich tip used for nano-indentation. ............................................ 30

Figure 18 Displacement measurement setup (1) measurement displayed and recorded on computer (2) Agilent 33220A function generator (3) Agilent 54622A oscilloscope (4) light source (5) BGA strip (6) CCD camera. .................................................................................................................. 35

Figure 19 Average displacement of (a) SWNT and (b) MWNT BGA samples (with different thickness ratios and thicknesses from 256.54 to 546.10 µm) vs. voltage. ............................................. 38

Figure 20 Average displacement of SWNT and MWNT BGA samples (at several applied voltages with different thickness ratios and thicknesses from 256.54 to 546.10 µm) vs. frequency. ......................................................................................................................................................... 39
Figure 21 Average displacement per unit length of BGA samples with different weight fractions of constituents at different frequencies and 10 V ................................................................. 39

Figure 22 (a) Effect of BGA thickness on the displacement per unit length for SWNT and MWNT samples at 10 V and two different frequencies (b) displacement of Regular SWNT samples with various thickness ratios (10 V-0.1 Hz) ................................................................. 41

Figure 23 Change in maximum deflection of SWNT BGAs at different voltages by time .......... 42

Figure 24 Lifetime of SWNT and MWNT BGAs at 7.5 V ......................................................... 43

Figure 25 Change in the average maximum displacement of BGA samples by over weeks of being stored .................................................................................................................................. 44

Figure 26 The model fitted to the (a) regular SWNT and (b) MWNT BGA displacement tests conducted with different applied voltage, frequency, and thickness. ........................................... 49

Figure 27 Comparing the model prediction to the other reported results [16-18]. ................. 51

Figure 28 (a) Micropump assembly (b) Conjugated polymer membrane used as diaphragm pump (modified from [53]). .................................................................................................................... 54

Figure 29 (a) Schematic view of electrically-driven hydrogel sorter (b) examples of droplet sorting (water phase flow rate = 6µL/min) (modified from [47]). ................................................................. 54

Figure 30 Gold coated BGA diaphragms ..................................................................................... 56

Figure 31 Microvalve classification and some examples for each group (modified from [27]).. 57

Figure 32 Different types of actuation mechanisms of active microvalves with mechanical moving parts: (a) electromagnetic; (b) electrostatic; (c) piezoelectric; (d) bimetallic; (e) thermopneumatic; (f) shape memory alloy [27] ......................................................................................................................... 58
Figure 33 BGA sample being operated under water. ................................................................. 60

Figure 34 Plots of the measured blocking force of the bucky-gel actuator of 0.465 mm in
thickness reported by Mukai et al. (modified from [11]). ............................................................ 61

Figure 35 Observation of the BGA capability in lifting a 1.2 g nut (in addition to its own weight).
....................................................................................................................................................... 61

Figure 36 Base part of the designed microvalve device.............................................................. 63

Figure 37 Exploded view of the microvalve device assembly...................................................... 64

Figure 38 3D printer major units............................................................................................... 64

Figure 39 Arrangement of the parts on the build pad in 3D printing......................................... 65

Figure 40 BGA microvalve base part after being printed on the 3D printer build pad.............. 66

Figure 41 Washed parts in the UV oven for curing................................................................. 66

Figure 42 Final base part fabricated by 3D printing ................................................................. 67

Figure 43 MP6 mini-pump used in the flow rate measurement.................................................. 68

Figure 44 Flow rate measurement setup (1) Bartels controller MP-X (2) Agilent 33220A
function generator (3) fluid reservoir (4) Bartels MP6 mini-pump (5) Microvalve device (6)
Output container (7) Agilent 54622A Oscilloscope................................................................. 70

Figure 45 Fabricated microvalve device.................................................................................. 70

Figure 46 Measured flow rate at different frequencies and two different voltages. BGA was
driven under a rectangular shape signal with 50% duty cycle. .................................................. 73
ABSTRACT

Since the demonstration of Bucky Gel Actuator (BGA) in 2005, a great deal of effort has been exerted to develop novel applications for electro-active morphing materials. Three-layered bimorph nanocomposite has become an excellent candidate for morphing applications since it can be easily fabricated, operated in air, and driven with few volts.

There has been limited published study on the mechanical properties of BGA. In this study, the effect of three parameters: layer thickness, carbon nanotube type, and weight fraction of components, on the mechanical properties was investigated. Samples were characterized via nano-indentation and DMA. It was found that BGA composed of 22 wt% single-walled carbon nanotubes and 45 wt% ionic liquid exhibited the highest hardness, adhesion, elastic and storage moduli.

Most of BGA potential applications would require control over one BGA output: displacement. In this study, various sets of experiments were designed to investigate the effect of several parameters on the maximum lateral displacement of BGA. Two input parameters: voltage and frequency, and three material/design parameters: carbon nanotube type, thickness, and weight fraction of constituents, were selected. A new thickness ratio term was also introduced to study the role of individual layers on BGA displacement. In addition, an important factor in the design of BGA-based devices, lifetime, was investigated. Finally, possible degradation of BGA was studied by repeating displacement measurements after several weeks of being stored.

Based on displacement studies, a new model was established utilizing nonlinear regression to predict BGA maximum displacement based on the effect of these parameters. This
model was verified by comparing its predictions with other reported results in the literature. The model displayed a very good fit with various reported cases of BGA samples made with different types of CNT and ionic liquid.

Microfluidics is a promising field of application for BGA. A brief literature review on the electroactive mechanisms used in microfluidics is presented. Preliminary force studies proved that BGA has the capability to be employed as a microvalve. A flow regulator utilizing a BGA microvalve was designed and fabricated. Flow rate measurements showed the capability of BGA-valve in manipulating the flow rate in different ranges.
CHAPTER 1. INTRODUCTION

During the last two decades, considerable research has been conducted aiming at the development of electro-active polymers (EAP) capable of operating in air with minimum amount of power consumption and maximum performance. An EAP by definition is a polymer system that responds to electrical stimulation with significant change in size or shape [1]. Two major types of EAPs are Ionic EAPs such as Ionic Polymer Metal Composites (IPMC) or Ionic gels which operate via ion transfer, and Electronic EAPs such as Dielectric elastomers or Electrostrictive relaxor ferroelectric polymers which operate based on Coulomb force.

While most EAPs actuate in electrolyte solution, Wallace et al. used ionic liquid to make “built-in” solid electrolytes and successfully employed it in the first “dry” EAP actuator [2]. However, short lifetime and long response time were major drawbacks. These drawbacks came about because of the faradaically driven motion mechanism via oxidation/reduction of the conjugated polymers [2]. Baughman et al. resolved the response time issue by using carbon nanotubes. Baughman et al. was the first to report the deformation of carbon nanotube sheets submerged in an aqueous electrolyte solution as a result of electric potential application [3]. Since the actuation was the result of double-layer formation due to charging and discharging of the carbon nanotubes while non-faradaically driven, the response time issue was resolved. However, the actuation was only possible in the presence of an electrolyte solution. In 2005, Asaka et al. [4] introduced “Bucky Gel” and employed it in the first non-faradaically driven dry EAP.

Bucky gel is a room temperature gelatinous mixture of an ionic liquid and carbon nanotubes. Outstanding mechanical and electrical properties of CNT and high conductivity and
stability of ionic liquids made this mixture an attractive alternative in the field of smart materials and structures. This mixture has been utilized in fabrication of actuators, stretchable electronic devices and energy conversion materials [5]. Moreover, bucky gel has been used to develop electrochemical biosensors [6] and multifunctional materials with high activation for reduction of Oxygen (O$_2$) [7].

Bucky Gel Actuator (BGA) has been one of the most successful applications of bucky gel. BGA is a tri-layered bimorph nanocomposite system consisting of an electrolyte layer sandwiched between two identical electrode layers (Figure 1). The BGA electrode layer is composed of the bucky gel mixture reinforced with a polymer matrix. The electrolyte layer is made of the same polymer matrix and ionic liquid utilized in the fabrication of the bucky gel. When a potential difference is applied to the electrodes, BGA bends toward one side. The bending direction changes by changing the polarity of the applied current.

The reasons behind BGA bending motion are still not fully understood. The response is usually explained by two superimposing mechanisms; charge injection and ion transfer. Charge injection model states that when a voltage is applied to the electrodes, there will be a change in carbon-carbon bond length in carbon nanotubes that leads to the expansion and contraction of the opposite electrodes [8]. Therefore, change in electrodes size causes the actuator to bend toward one side, since the layers are bound together. The second mechanism states that the bending motion is the result of ion transfer between BGA layers. According to this hypothesis, bending motion occurs when positive and negative ions are separately accumulated on the opposite electrodes. Therefore, one electrode layer would swell while the other one shrinks as a result of the size difference of the positive and negative ions. This change in the electrode size will cause BGA to bend toward the anode.
Figure 1 (a) Schematic of BGA bending motion (b) BGA strip bended toward one side as the result of applied voltage during experiment.

BGA has characteristics that make it an excellent candidate for different morphing applications. One characteristic advantage is the fact that BGA is classified as a “dry” actuator which can be operated in air without the need for any electrolyte solutions. Another is that BGA
is driven by just a few volts (1-10v) and at a wide range of frequencies unlike typical Shape Memory Alloys (SMA) or conducting elastomers. Moreover, response time and lifetime is much improved compared to redox-based conducting polymer (polypyrrole) actuators, since BGA bending is carried out non-faradaically. One more major advantage of BGA, especially compared to alternatives such as ionic polymer-metal composites (IPMC), is the simple fabrication procedure [9]. BGA also has the mutual capability of being used as a sensor or an actuator since a low amount of current is induced when bent [10].

Different studies have been conducted on BGA to characterize its properties and improve the performance. Previous studies have proved that the applied voltage and frequency [11], thickness of layers [11], and ionic liquid type [12-15] have a direct effect on the morphing efficiency of BGA. However, these studies were conducted in a small range of investigated parameters and on a limited number of samples. Several attempts have also been made to achieve larger displacements in BGA specimens. Chemical modification of CNT; through adding aromatic diamine groups [16, 17] or crosslinking [18], showed an enhancement in BGA displacement. Employing conductive and non-conductive additives to boost BGA response has also been studied [15, 19]. It was observed that using conductive additives such as alkaline earth metal salts has increased the conductivity and capacitance of the electrodes and led to higher displacements. Furthermore, the possibility of improving the electrode capacity by using carbon nanofibers [20], activated carbon nanofibers [20], Au paste [21], or nanoporous carbide driven carbon [22] has been investigated. More recently, inclusion of millimeter long carbon nanotubes known as super growth single walled carbon nanotubes (SG-SWNT) was reported to improve BGA displacement significantly [2]. Fabrication procedure is another potentially effective parameter on BGA displacement. An automatic fabrication method using a printing system has
been demonstrated with the aim of minimizing the defects and improving sensing/actuating performance [23, 24].

BGA, with improved mechanical strength and load tolerance, has also the potential to be employed in smart structures with sensing capabilities. However, the main challenge is to maintain the morphing efficiency while improving the mechanical properties. Therefore, a better understanding of mechanical properties and geometrical and material parameters is essential in order to overcome this challenge. Limited investigation so far includes the elastic and fracture properties of BGA focusing on the influence of carbon nanotube cross-linking [25]. However, there is no comprehensive study on the mechanical properties of BGA in the literature.

In this study the effect of three geometrical and material parameters (layer thickness, carbon nanotube type, and weight fraction of constituents) on the mechanical properties were investigated by measuring the storage and loss moduli, viscosity, hardness and reduced elastic modulus with Dynamical Mechanical Analysis (DMA) and nano-indentation.

In addition, adhesion tests were carried out on all samples using nano-indentation. This characteristic of BGA plays an important role in microfluidic devices. In such devices, adhesion of moving parts (microvalves, micromixers, etc.) affects the flow and system efficiency [26, 27]. Therefore, measuring the work of adhesion and investigating the effect of different parameters on adhesion will help in selecting the proper material.

There is no published study on any practical applications or BGA-based devices. An important factor that affects the design and use of BGA in any device is the control over the output, specifically the displacement. Control over the displacement of BGA under different conditions requires an in-depth understanding of the effective parameters.
A successful advancement in BGA-based devices would be the establishment of a model to predict the displacement. Development of such model would facilitate the application of BGA in different fields such as MEMS applications, microfluidics, micro-robots, etc.

In this study, five parameters were selected to investigate their effect on BGA displacement. Two of those parameters (voltage, frequency) were input parameters and the other three (CNT type, thickness, weight fraction of constituents) were material and design related parameters. A new thickness ratio was also introduced to compare the role of electrolyte and electrode layer thickness on BGA response.

There is no published study on the lifetime of BGA. Lifetime of the BGA samples was also investigated to monitor the change in the maximum deflection of BGA. Lifetime tests were conducted at different voltages (inside and outside of the ionic liquid electrochemical stability window) to study its effect on BGA performance. Samples with two different carbon nanotube types were used in the lifetime tests to investigate the potential influence. Degradation of BGA after long shelf-life was the next topic of research. There was a concern that BGA performance may deteriorate after being off-line or in storage. Therefore, some samples were tested again after being stored for weeks and months.

Microfluidic is a very attractive field of application for smart materials. The concept of disposable, easy-to-use, and “at the point of care” lab-on-a-chip has led many researchers toward applying electroactive materials in fabrication of micro-scale devices. In the last chapter, a brief literature review on application of similar electroactive materials is presented. The primary studies and advantages of employing BGA in such devices are discussed.
Preliminary force studies on BGA revealed that this actuator can provide enough force to be utilized as a microvalve. A BGA-based device was designed and fabricated to test this hypothesis. The parameters considered in the design are highlighted in the last chapter. The designed device was fabricated using a 3D printer. Afterwards, the device was tested to check its capability in manipulating the flow rate of a passing fluid.
CHAPTER 2. MATERIALS AND FABRICATION

Bucky Gel Actuator consists of three major constituents: carbon nanotubes, ionic liquid and the polymer matrix. An appropriate solvent is also required to be used as the binder for the polymer matrix and also to serve as the medium for the ball-milling and ultra-sonication of carbon nanotubes.

A major component in Bucky gel is the ionic liquid. Ionic liquids (IL) are liquid salts at room temperature and have superior properties of high conductivity, non-volatility and wide potential window. EMIBF$_4$ (1-ethyl-3-methylimidazolium tetrafluoroborate) was selected as the ionic liquid in this research. EMIBF$_4$ had shown the best performance compared to other available ionic liquids [12] (Figure 2). KYNAR FLEX 2851 PVDF (Archema Inc., Philadelphia, USA) was selected as the polymer support. This fluorinated thermoplastic copolymer was chosen for its outstanding mechanical properties and chemical resistance [12]. Dimethylacetamide (DMAC) was employed as the solvent and the binder for the polymer.

![Figure 2 Ionic conductivity of electrolyte layers containing various ionic liquids](modified from [12]).
A regular BGA sample was fabricated containing 22 wt% SWNT, 45 wt% IL and 33 wt% PVDF. The fabrication procedure for the electrode layers began with ball-milling the mixture of 0.2 g pristine SWNT, 0.4 g IL, and 4 mL DMAC for 30 minutes (Figure 3). Ball-milling of the mixture was carried out to break the carbon nanotubes bundles, since they have a tendency to form agglomerates (Figure 4).

Figure 3 Fabrication procedure (a) Spex SamplePrep Mix/Mill 8000M used in ball-milling process (b) ball-mill vial and the two Zirconia Ceramic ball (12.7 mm) (c) Output mixture in the ultrasonic bath (d) Degassing chamber.
In order to create the layered structure of BGA, two outside electrode layers and one middle electrolyte layer (1:1 combination), are hot pressed together at 140°C for 10 minutes, while under uniform pressure. The outcome is a thin and flexible strip of BGA, which can be cut to any desired size (Figure 5).

The outcome of the ball-milling process was a gelatinous black mixture (Figure 4a) to which 0.32 g PVDF and 8 mL DMAC were added. This mixture was ball-milled again for another 30 minutes, sonicated for 24 hours, casted in silicon rubber molds and cured in an oven for 12 hours at 50°C (Figure 4b). Finally, samples were put in a vacuum chamber at reduced pressure for another 100 hours in order to let the DMAC evaporate gradually. Negligible difference between the theoretical and measured density of the BGA samples showed that DMAC has evaporated completely. For the fabrication of electrolyte layers, IL and PVDF are mixed together (1:1 ratio), dissolved in DMAC, and then casted similar to the electrode layers.

Figure 4 (a) bucky gel mixture after ball-milling process (b) BGA mixture casted in silicon rubber molds.
Different sets of samples had to be prepared for mechanical characterization and displacement study. Three geometrical/material parameters (weight fraction of components, carbon nanotube type, and thickness) were selected to study their influence on mechanical properties and BGA displacement.

The weight fraction of constituents (CNT, IL and PVDF) was considered as a potential effective parameter on BGA properties and performance. Therefore, in addition to regular SWNT BGA, three other sets of samples were fabricated with different weight fraction of constituents. These sets were labeled as “12% SWNT”, “27% SWNT”, and “55% IL”. The weight fractions of constituents in each fabricated set of samples are presented in Table 1. The “27% SWNT” batch was the only one used for the displacement measurement studies.

Carbon nanotube type was also selected as another potential effective parameter on the mechanical behavior and displacement of BGA. Functionalized multi-walled carbon nanotubes (COOH-MWNT) were also used to fabricate a set of BGA samples. This set was labeled as

Figure 5 BGA sample after being hot-pressed, showing good flexibility.
“MWNT”. The weight fraction of constituents in the MWNT samples was the same as in the regular BGA with pristine SWNTs (Table 1).

For the purpose of investigating the thickness effect, electrode and electrolyte layers were fabricated at various thicknesses. However, there was no precise control over the final thickness of individual layers. In addition, thicker BGA samples were fabricated via hot pressing six layers (four layers of electrodes and two layers of electrolyte) instead of three. These samples were referred to as “2-2-2 combination” (Figure 6).

A thickness ratio was introduced in this study to examine the potential effect of each individual layer thickness on BGA displacement. Therefore, another layer combination consisting of thicker electrolyte layer (“1-2-1 combination”) was also prepared for displacement studies (Figure 6).

A different set of samples was also prepared for nano-indentation tests (Figure 7). These samples were made by casting the sonicated mixture of nanotubes, IL and PVDF on a glass substrate and putting them in degassing chamber at 70°C and atmospheric pressure for six to eight hours.

Table 1 Weight fractions of constituents in different sets of samples.

<table>
<thead>
<tr>
<th>Name</th>
<th>Carbon nanotube (wt%)</th>
<th>Ionic liquid (wt%)</th>
<th>PVDF (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Regular SWNT</td>
<td>22</td>
<td>45</td>
<td>33</td>
</tr>
<tr>
<td>12% SWNT</td>
<td>12</td>
<td>55</td>
<td>33</td>
</tr>
<tr>
<td>55% IL</td>
<td>22</td>
<td>55</td>
<td>23</td>
</tr>
<tr>
<td>27% SWNT</td>
<td>27</td>
<td>40</td>
<td>33</td>
</tr>
<tr>
<td>MWNT</td>
<td>22</td>
<td>45</td>
<td>33</td>
</tr>
</tbody>
</table>
Figure 6 Different layer combinations used in this study (Black: electrode layers – White: electrolyte layers), 1-2-1 layer combination was only used for displacement study.

Figure 7 Regular SWMT bucky gel electrode layer casted on glass substrate for nano-indentation tests.
CHAPTER 3. MECHANICAL CHARACTERIZATION

3.1 Characterization methods

In this study, Dynamic Mechanical Analysis and nano-indentation were used to characterize BGA. Dynamic Mechanical Analysis was employed to study the viscoelastic behavior of BGA under different strain rates. Storage modulus, Loss modulus, and viscosity of different samples were measured. Moreover, hardness, Reduced Elastic modulus and adhesion of electrode layers were also measured via nano-indentation. The method used to calculate work of adhesion in this study has not been reported before.

3.1.1 Dynamic Mechanical Analysis (DMA)

DMA is the most common and useful characterization tool for investigating the viscoelastic behavior of thin films. In this study, Frequency sweep test was conducted on individual electrode and electrolyte layers, and the BGA samples using a *Rheometric Scientific RSA III Dynamic Mechanical Analyzer* (Piscataway, USA) (Figure 8). The sample dimensions and test parameters were selected following ASTM D4065, D5026 and E1640–04 standards. These tests were designed to measure the viscosity and also the storage and loss moduli of each sample.

Since storage and loss moduli measurements are only valid in linear elastic zone, strain sweep tests had to be conducted on samples from each batch. Based on strain sweep results, different initial strains in the linear elastic region were selected for each set of samples in order to observe the trend at different levels. Tests on individual layers were conducted on 40 x 9 mm rectangular films with thickness of 59 ± 6 µm. General settings used in DMA tests are presented in Table 2.

---

1 Ali Kadkhoda Ghamsari *et al* 2012 *Smart Mater. Struct.* 21 045007
Frequency sweeps of BGA samples were carried out at two different thicknesses (1-1-1 and 2-2-2 combinations). The thickness of 1-1-1 and 2-2-2 combination BGAs was $189 \pm 15$ and $332 \pm 46$ microns, respectively. The effective sample length (fixture gap) was 20 mm in all tests and the frequency was varied from 0.01-70 Hz in logarithmic increments. An initial preload and a constant dynamic to static force ratio were applied to each sample to avoid buckling during DMA tests. All tests were conducted at room temperature, except tests which were carried out at an elevated temperature ($80^\circ$C) to investigate temperature effects. $80^\circ$C is in the vicinity of the maximum temperature that BGA will experience in most potential applications. As shown in Table 2, a maximum amount of force was set to avoid rupture of the samples while under frequency sweeps.
Table 2 Setting used for Dynamic Mechanical Analysis tests.

<table>
<thead>
<tr>
<th>Test type</th>
<th>Dynamic strain frequency sweep</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial and final frequency</td>
<td>0.01-70 Hz</td>
</tr>
<tr>
<td>Auto tension adjustment</td>
<td>On</td>
</tr>
<tr>
<td>Max auto tension displacement</td>
<td>2.0 mm</td>
</tr>
<tr>
<td>Max auto tension rate</td>
<td>0.01 mm/sec</td>
</tr>
<tr>
<td>Max allowed force</td>
<td>40.0 g</td>
</tr>
<tr>
<td>Strain adjustment</td>
<td>0.05%</td>
</tr>
</tbody>
</table>

3.1.2 Nano-indentation

Determining the mechanical properties of thin films based on their load-displacement curve during indentation has become a common characterization tool. Nano-indentation has been utilized to investigate different mechanical properties of composites reinforced by nanofillers; such as hardness, elastic modulus, adhesion, and fracture toughness [28-33]. In this method, hardness is measured based on the amount of force experienced by the probe during loading and unloading. Moreover, since indentation can be conducted at different depths, evaluation of both bulk and surface properties is possible with a single indentation test.

The data obtained from nano-indentation were interpreted by the improved technique introduced by Oliver et al. [34, 35]. This technique is an extension to the flat punch approximation method proposed by Doerner and Nix [36]. It’s worth mentioning that in nano-indentation, the quantity which is being measured is “Reduced” elastic modulus ($E_r$) which takes the non-rigidity of the indenter tip into account:

$$\frac{1}{E_r} = \frac{1 - \nu^2}{E} + \frac{1 - \nu_i^2}{E_i}$$  \hspace{1cm} \text{Equation 1}

where $E$, $\nu$, $E_i$ and $\nu_i$ are Young’s modulus and Poisson’s ratio of specimen and indenter, respectively.
Nano-Indentation tests were conducted using the \textit{Hysitron TI-900 Triboindenter} (Minneapolis, USA) and a Berkovich tip (Figure 9). Since the distribution of nanofillers in each specimen was random, a significant number of indentations at different points were performed on each specimen.

The load function used for these indentations is similar to the load curve recommended by Oliver \textit{et al.} [34]. As shown in Figure 10, the sample was loaded and unloaded three times (A on the figure) to check the reversibility of the deformation and ensure that unloading data used for calculating the hardness were elastic. Effect of small changes in temperature that may cause expansion/shrinkage in machine components causing faulty tip displacement had to be taken into account [34]. Therefore, the third unloading was followed by a hold at 10\% of the maximum load (B) to investigate the thermal drift. This hold was followed by one last loading and a hold (C) at maximum load. This hold was applied to allow any final time dependent plastic effects to diminish before unloading the specimen to eliminate the influence of temperature on data analysis. Holding load at maximum would also prevent the overestimation of hardness and elastic modulus. The slope of the last unloading curve (D) was used for hardness calculation.
Figure 10 Load function used in nano-indentation tests.

The maximum load used for indentation was 2000µN, except for “55% IL” samples. 1000µN was selected as the maximum force for testing the “55% IL” samples in order to avoid large displacements. In all cases, a pre-load of 2µN was used to define the initial point of contact. In the case of MWNT samples, the indentation tests were conducted at two different maximum loads to study the load effect on the results. Moreover, in order to investigate the effect of time dependent deformation, hardness and reduced elastic modulus of samples was calculated after the elastic loadings (the third unloading - E on Figure 10).

Adhesion was one of the other properties investigated in this study. Numerous methods of measuring adhesion in different structures have been reported based on their geometry, application and materials [32]. Nano-indentation is one of the methods that have been used to evaluate adhesion especially in the case of thin films [29, 33]. In this study, Johnson-Kendall-Roberts (JKR), Derjaguin-Muller-Toporotov (DMT) and Johnson contact mechanics theories were employed to characterize adhesion [37].

In JKR and DMT theories, adhesion is assumed to be a function of two factors: the force required to separate the two surfaces in contact (separation force), and radius of two contact surfaces. Another assumption is that adhesion is independent of surface material. Using this
approach, Han et al. [38] have employed Atomic Force Microscopy (AFM) pull-off test to measure the separation force and subsequently the work of adhesion on different surfaces. In JKR and DMT methods, two surfaces in contact are assumed to be elastic. However, during indentation the sample surface go through permanent deformation, before the indentation tip is pulled apart. Johnson has suggested an approximate method to analyze the work of adhesion assuming that one of the surfaces goes under fully plastic deformation [39, 40]. In addition to separation force and contact radii, the model suggested by Johnson also takes material’s effects into account.

In this study separation force was measured with nano-indentation method. Separation force was defined as the maximum force experienced by the indentation tip while being withdrawn from the specimen. As for the second required factor for calculations, the radii of the sample surface and indentation tip should be determined. The radius of the indentation tip was measured based on Scanning Electron Microscope (SEM) images, and sample surface was assumed to be perfectly flat. Only samples tested at 2000 µN maximum load were considered for adhesion study.

### 3.2 Characterization result and discussion

Three major mechanical properties (reduced elastic modulus, hardness, adhesion) were evaluated in this study. Reduced elastic modulus and hardness were studied via nano-indentation, whereas Storage and loss moduli were measured using Dynamic Mechanical Analysis. Evaluation of adhesion via nano-indentation was a unique procedure that there was no published study on, before this research. Observations made for each individual case is presented in its own section in the following.
3.2.1 Storage modulus and viscosity

The first mechanical property determined in this study was the storage modulus. Storage and loss moduli were studied by frequency sweep tests. In each frequency sweep, the frequency was varied from minimum to maximum (0.01-70 Hz) at fixed strain and temperature. Based on the tests carried out at room temperature, increase in frequency resulted in an increase of storage modulus in both electrode and electrolyte layers (Figure 11). Since loading in this test was uniaxial, increase in frequency was equivalent to increase in strain rate. It was also observed that the slope of the storage modulus curve was similar for different initial strains. Therefore, initial strain did not affect the rate of the increase in storage modulus. In order to investigate the sensitivity of storage modulus to temperature, a frequency sweep was conducted on the electrolyte layer at 80˚C. It was observed that storage modulus decreased significantly at elevated temperature. However, the storage modulus increased with increase in frequency as in the case at room temperature.

Figure 12 presents the frequency sweep of different types of electrode layers at the same strain level (0.5%). Regular SWNT electrode layer (22 wt% SWNT, 45 wt% IL and 33wt% PVDF) displayed the highest storage and loss moduli. Considerable decrease in the storage and loss moduli for the MWNT electrode occurred compared to regular SWNT samples with the same weight fractions of materials. The 55% IL electrode (22wt% SWNT, 55wt% IL and 23wt% PVDF) had a higher storage modulus than the 12% SWNT electrode (12wt% SWNT, 55wt% IL and 33wt% PVDF) which confirmed the assumption that weight fractions of constituents; specifically of carbon nanotubes, is an effective parameter on mechanical response of BGA.
Figure 11 (a) Frequency sweep of a regular SWNT electrode layer (b) Frequency sweep of an electrolyte layer.
Figure 12 (a) Storage modulus of different electrode layers via frequency sweep at 0.5% strain
(b) Loss modulus of different electrode layers at 0.5% strain.
Figure 13 Frequency sweep of (a) Regular BGA and (b) COOH-MWNT BGA at different strain level.
Similar to individual electrode layers, the storage modulus of three-layer regular SWNT BGA also increased with increase in frequency. However, unlike the individual electrode case, the rate of this increase was not the same at different strain levels (Figure 13). At lower strains a steeper increase was observed compared to higher strains. A second set of three-layer BGA samples was prepared using MWNT electrodes, in order to observe the effect of carbon nanotube type on the visco-elastic behavior of BGA. MWNT three-layer BGA showed lower storage modulus than Regular SWNT BGA. However, similar to regular SWNT samples, MWNT samples storage modulus increased with increase in frequency.

In order to investigate the effect of layer thickness, regular SWNT and MWNT BGAs were fabricated with (2-2-2) and (1-1-1) combination. As shown in Figure 14, higher thickness resulted in higher storage modulus. Moreover, (1-1-1) regular SWNT BGA had the highest storage modulus per unit thickness. Increase in thickness of regular SWNT BGAs was also more influential on mechanical properties than in case of MWNT BGAs. Thin regular SWNT BGA (1-1-1) displayed better mechanical properties than thicker MWNT BGA (2-2-2). Therefore, regular SWNT BGA will result in structures with higher strength to weight ratio. This higher strength to weight ratio of regular SWNT BGA makes it a better candidate for being used in multifunctional structures such as sensors and micro-robotics.

Viscosity of BGA was also investigated in this study. As shown in Figure 15, all samples had similar viscosity at higher frequencies (>0.1 Hz). However, viscosity exhibited a dependence on weight fraction of constituents and nanotube type at lower frequencies (<0.1 Hz). In this range of frequency, regular SWNT samples displayed higher viscosity in comparison with MWNT samples with the same weight fractions. But, comparing MWNT and 55% IL samples with the same weight fraction but different types of nanotubes, revealed that weight fraction of
all constituents influenced viscosity. Viscosity of SWNT and MWNT BGAs with (1-1-1) and (2-2-2) layer combinations was also studied. As shown in Figure 15, thickness does not affect viscosity of BGA.

Figure 14 Frequency sweep of Regular SWNT and COOH-MWNT BGA at (a) two different thicknesses (b) unit thickness.
Figure 15 Complex viscosity of (a) different electrodes and electrolyte layers at 0.5% strain (b) SWNT and MWNT BGAs at 1.0% strain.
3.2.2 Hardness

Normal distribution of the hardness data was obtained from nano-indentation. The measurement was checked using MINITAB® Release 14 (Minitab Inc., 2004) software package. The normality test was used to make sure that data points that were in 95% confidence interval.

A common source of error in measuring the hardness of samples using nano-indentation is neglecting the pile-up of the material around the periphery of the indentation impression. The “Pile-up effect” occurs in case of elastic-plastic materials and leads to the overestimation of hardness. A factor that prevents the pile-up effect is the ability of the material to work harden.

According to finite element studies by Knapp et al. [41], pile-up is large only when the ratio of “depth of final impression left on the surface” to “depth of contact at maximum load” \( \frac{h_f}{h_{max}} \) is close to one and the work hardening is small. Based on their observation, the pile-up is negligible when \( \frac{h_f}{h_{max}} < 0.7 \). This ratio was checked in all nano-indentation tests to confirm that the pile-up effect could be neglected. Since the ratio of \( \frac{h_f}{h_{max}} \) was less than 0.7 in all cases, the pile-up effect was assumed to be negligible in this study.

Table 3 Nano-indentation data for each electrode set.

<table>
<thead>
<tr>
<th>Type</th>
<th>Hardness (MPa)</th>
<th>Reduced Elastic Modulus (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Mean Standard Deviation COV*</td>
<td>Mean Standard Deviation COV*</td>
</tr>
<tr>
<td>Regular SWNT</td>
<td>10.520 1.382 0.131</td>
<td>327.0 49.1 0.150</td>
</tr>
<tr>
<td>MWNT (2000µN)</td>
<td>5.911 0.837 0.142</td>
<td>176.9 23.8 0.135</td>
</tr>
<tr>
<td>MWNT (1000µN)</td>
<td>5.994 0.872 0.145</td>
<td>178.4 27.2 0.152</td>
</tr>
<tr>
<td>12% SWNT</td>
<td>5.413 0.653 0.121</td>
<td>137.0 20.9 0.153</td>
</tr>
<tr>
<td>55% IL</td>
<td>3.260 0.465 0.143</td>
<td>114.1 14.8 0.130</td>
</tr>
<tr>
<td>SWNT (after the elastic loading)</td>
<td>10.560 1.427 0.135</td>
<td>376.6 54.7 0.145</td>
</tr>
</tbody>
</table>

* Coefficient of Variation
The average calculated thermal drift rate for tested samples was < 0.3 nm/min. Since the hold at maximum load is 20 sec, the effect of thermal drift was negligible. The hardness and reduced Elastic Modulus of different sets of samples are presented in Table 3.

The results determined that regular SWNT electrode type had the highest hardness. The second highest hardness belonged to MWNT samples. Therefore, it was concluded that the type of carbon nanotube plays an important role in controlling the hardness of BGA and using SWNT in BGA would lead to higher hardness. However, the carbon nanotube type was not the only influence. The lowest hardness measured belonged to 55% IL samples which had the same SWNT content (22 wt%) as the regular SWNT layer, but with less PVDF content. These observations dictated that the weight fraction of constituents was a major factor on hardness.

As mentioned earlier, nano-indentation of MWNT samples was conducted at two different maximum loads, 1000 and 2000 µN, to investigate the load effect on measured hardness. According to the results presented in Table 2, the mean hardness value of the two sets of samples showed only a slight difference of 1.4%. Similarity of these results allowed the comparison of 55% IL samples (conducted at maximum load of 1000 µN) to the other sets of samples (conducted at 2000 µN).

Figure 16 shows hardness versus contact depth of the indentation of all samples. Based on the indentations conducted at different contact depths, it was observed that there is a power-law relationship between hardness and contact depth. The hardness of samples decreased with increasing depth. This decrease could be the result of poor carbon nanotubes distribution in the bulk of the composite and segregation of nanofillers near the surface [31]. Further investigation is required to fully understand the reason behind this phenomenon.
Figure 16 (a) Hardness vs. Contact depth for Reg SWNT (2000 µN), 12% SWNT (2000 µN) and the 55% IL (1000 µN) (b) Hardness vs. Contact depth for MWNT at two different maximum loads.

3.2.3 Adhesion

Johnson, Kendall and Roberts were pioneers in incorporating the adhesion effect into the classic Hertzian contact model. In the case of low loading between two elastic spheres with the radii of $R_1$ and $R_2$, JKR theory predicts the force required to overcome the adhesion work between two surfaces as [37]:

$$P = \frac{3}{2} \pi \gamma R$$

Equation 2

where $\gamma$ is the work of adhesion and $\frac{1}{R} = \frac{1}{R_1} + \frac{1}{R_2}$. In the case of nano-indentation on flat surfaces
Figure 17 SEM image of the Berkovich tip used for nano-indentation.

$R$ is equal to the radius of the indentation tip. Based on the SEM images (Figure 17) of the Berkovich tip used for all the measurements in this study, the radius $R$ was determined to be 2.2 µm. Adhesion force $P$ was determined according to the maximum amount of negative force that was sensed by the indentation tip while being withdrawn from the surface of the sample.

JKR model, however, only considers the contact pressure and adhesion forces inside the contact area. Derjaguin, Muller and Toporotov introduced a new model (DMT) taking into account the effect of interactions existing outside the contact area.

Table 4 Work of adhesion (JKR, DMT and Johnson theories).

<table>
<thead>
<tr>
<th>Type</th>
<th>Average pull-off force (µN)</th>
<th>Work of adhesion (J/m$^2$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>JKR</td>
</tr>
<tr>
<td>Regular SWNT</td>
<td>23.886</td>
<td>2.304±0.386</td>
</tr>
<tr>
<td>12% SWNT</td>
<td>17.842</td>
<td>1.721±0.312</td>
</tr>
<tr>
<td>55% IL</td>
<td>15.914</td>
<td>1.535±0.227</td>
</tr>
<tr>
<td>MWNT</td>
<td>6.492</td>
<td>0.626±0.234</td>
</tr>
</tbody>
</table>
According to DMT theory, the separation force is larger than the JKR model prediction by 50% and is equal to [38]:

\[ P = 2\pi \gamma R \]  
Equation 3

As mentioned before, Johnson [40] has suggested an approximate method for analysis of work of adhesion in elastic-plastic contacts. In this method, it is assumed that one of the surfaces in contact will go through fully plastic deformation under a compressive load \( P_0 \). According to Johnson model the separation force is equal to [39]:

\[ P = \frac{2\gamma E^* P_0^{1/2}}{(\pi H^3)^{1/2}} \]  
Equation 4

where \( H \) is hardness and:

\[ \frac{1}{E^*} = \frac{1 - \nu_1^2}{E_1} + \frac{1 - \nu_2^2}{E_2} \]  
Equation 5

Work of adhesion was calculated using the maximum loading force as \( P_0 \).

As shown in Table 4, the regular SWNT samples showed the highest adhesion. Comparing the adhesion work of Regular SWNT samples to the 12% SWNT and IL 55% samples, it was confirmed that similar to the case of hardness the weight fraction of all three constituents are influential on the work of adhesion.

Having the same weight fractions as the regular SWNT samples, MWNT samples displayed remarkably lower (~ four times) work of adhesion. This observation revealed the significant effect of carbon nanotube type on the adhesion of BGA samples. As mentioned earlier, adhesion plays an important role in microfluidic devices with microvalves that require rapid response and low operating force. Therefore, this minimal work of adhesion makes MWNT BGA a potential candidate for such devices.
Here is the highlight of the mechanical characterization results:

- All selected parameters (type of carbon nanotube, weight fraction of constituents, and thickness) were effective parameters and influence the mechanical properties.
- Based on DMA results, this composite showed viscoelastic behavior (higher storage modulus at higher strain rates).
- The BGA samples employing 22% weight fraction of Regular Single-Walled carbon nanotube displayed the highest hardness, Reduced elastic modulus, Storage modulus and adhesion.
- Adhesion of BGA was greatly influenced by the type of carbon nanotube (MWNT showed almost four times less adhesion)
CHAPTER 4. DISPLACEMENT MEASUREMENT

Most of the research conducted on BGA so far has been focused on characterization and performance enhancement. However, there is no published study on any practical applications or BGA-based devices. An important factor that affects the design and use of BGA in any device is the control over the main output, that is, displacement. Control over the displacement of BGA under different conditions requires an in-depth understanding of the effective parameters. A successful advancement in BGA-based devices would be the establishment of a model to predict the displacement. Development of such model would facilitate the application of BGA in different fields such as MEMS applications, microfluidics, micro-robots, etc.

The first step in establishing a model for BGA displacement was identifying the effective parameters. In this study, five parameters were selected to investigate their effect on BGA displacement.

4.1 Designed experiments

A set of experiments was designed to investigate the effect of five selected parameters on BGA maximum lateral displacement. Two of those parameters (voltage, frequency) were input parameters and the other three (CNT type, thickness, weight fraction of constituents) were material and design related parameters. A new thickness ratio was also introduced to compare the role of electrolyte and electrode layer thickness on BGA displacement.

BGA lifetime; repeatability of the oscillation and change in the displacement over time was also studied. There is no published study on the BGA lifetime, to the best of our knowledge. Samples were set to oscillate under a constant voltage and frequency in order to monitor if there
is any change in maximum lateral displacement over time. In addition to the lifetime of BGA, the change in performance of the BGA after storing for months was also studied to investigate if there is any degradation affecting its shelf-life. The size of samples used for displacement tests were the same as sample used in mechanical characterization.

4.1.1 Displacement measurement

The maximum displacement of the BGA was measured using an optical CCD camera. The oscillation of the BGA under four different peak-to-peak voltage (2, 5, 8, and 10 V) and five frequencies (0.1, 0.25, 0.5, 1, and 2 Hz) was recorded for the duration of 45 seconds. The frames showing the maximum deflection was identified using ImageJ software package. From each recorded video of oscillation, 3-5 frames displaying the maximum lateral displacement were identified using open source ImageJ software. Each frame was evaluated separately to measure the displacement, and the average of displacement values was reported as the maximum displacement for that experiment. Based on the focal distance of the lens, the resolution of the optical system was between 0.020 – 0.025 mm.

Similar to mechanical characterization study, weight fraction of constituents was one of the selected parameters to study. Four sets of samples were made and labeled as “Regular SWNT”, “12% SWNT”, “27% SWNT”, and “55% IL’. The weight fraction of constituents in each set is as presented in Table 1.

Functionalized multi-walled CNT (COOH-MWNT) was also used in fabrication of a set to investigate the effect of CNT type on the performance of BGA. This set is referred to as “MWNT” batch. The weight fraction of constituents in this batch was the same as regular SWNT batch.
Figure 18 Displacement measurement setup (1) measurement displayed and recorded on computer (2) Agilent 33220A function generator (3) Agilent 54622A oscilloscope (4) light source (5) BGA strip (6) CCD camera.

Thickness was the third parameter. BGA samples with various thicknesses were fabricated with hot-pressing different combinations of the electrolyte and electrode layers. Final thickness of tested samples varied from 185-546 µm, based on the layer combination. It was expected that in addition to the final thickness, thickness of the electrolyte layer (as the main source and medium for ions) should play an important role in BGA displacement. A new thickness ratio was introduced to investigate this hypothesis. The thickness ratio was defined as the sum of the electrolyte layers to the sum of electrode layers. Prepared samples had a wide range of thickness ratios (0.5-3.0) with different layer combinations and thicknesses (Table 5).

\[
\text{Thickness ratio} = \frac{\text{Sum of electrolyte layers}}{\text{Sum of electrode layers}}
\]

Equation 6

4.1.2 Lifetime and BGA degradation

Regular BGA samples were tested at three different peak-to-peak voltages (5, 7.5, and 10) at 0.1 Hz to study the change in maximum deflection by time. Each test was running for several days and each sample went through 30,000-60,000 cycles of deflection. Lifetime test was
repeated for a MWNT sample at 7.5 V and 0.1 Hz to investigate the effect of carbon nanotube type on BGA lifetime.

4.2 Results and discussion

4.2.1 Voltage and Frequency

Voltage and frequency are two major factors controlled by the operator that can influence BGA displacement. Increased applied voltage resulted in more ion transfer between BGA layers as voltage is related to the transferred charge through capacitance ($\Delta Q = C\Delta V$). Subsequently, higher BGA displacement occurred when more ions were transferred between BGA layers. This effect was more noticeable at lower frequencies, at which ions had more time to be transferred between BGA layers (Figure 19). However at higher frequencies, the response time was too short for ions to move between layers. Therefore, the displacement of BGA at any applied voltage was significantly reduced at higher frequencies.

Frequency influenced the effect of voltage on the displacement (Figure 19a). At lower frequencies (<0.5Hz), displacement increased exponentially with increasing applied voltage. However, at higher frequencies (>0.5Hz), displacement was limited attaining only a linear increase with the applied voltage.

Displacement tests were conducted on SWNT and MWNT BGA samples to study the effect of carbon nanotube type. Average displacement of MWNT samples was lower than SWNT at all voltage levels (Figure 19c). Similar to SWNT samples, the rate of change of displacement for MWNT samples was dependent on frequency. However, displacement was a linear function of voltage in MWNT BGA samples at the range of frequencies used in the study (Figure 19b). This consistent linear response would create an advantage for use of MWNT BGA in sensing
applications. This advantage would be realized since obtaining the constants in calibration would be simple and straightforward in a linear relationship. Whereas for SWNT BGA sensors, the frequency at which the linear response changes to exponential should be identified exactly.

Frequency on its own also showed remarkable influence on BGA displacement. In both SWNT and MWNT BGA samples, displacement displayed a power-law dependence on frequency (Figure 20). It was observed that at higher frequencies (>1Hz), CNT type and applied voltage had no effect on BGA displacement. However, as the frequency decreased, selected parameters (voltage, thickness, thickness ratio, and carbon nanotube) proved to be influential.

Table 5 Thickness, Thickness ratio, and layer combination of the tested samples.

<table>
<thead>
<tr>
<th>No.</th>
<th>Set</th>
<th>Layer Combination</th>
<th>Total Thickness (µm)</th>
<th>Thickness ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Regular SWNT</td>
<td>1-1-1</td>
<td>256.54</td>
<td>0.65</td>
</tr>
<tr>
<td>2</td>
<td>Regular SWNT</td>
<td>1-1-1</td>
<td>294.64</td>
<td>-</td>
</tr>
<tr>
<td>3</td>
<td>Regular SWNT</td>
<td>2-2-2</td>
<td>546.10</td>
<td>0.83</td>
</tr>
<tr>
<td>4</td>
<td>Regular SWNT</td>
<td>2-2-2</td>
<td>513.08</td>
<td>0.97</td>
</tr>
<tr>
<td>5</td>
<td>Regular SWNT</td>
<td>1-2-1</td>
<td>388.62</td>
<td>1.33</td>
</tr>
<tr>
<td>6</td>
<td>Regular SWNT</td>
<td>1-1-1</td>
<td>185.42</td>
<td>0.96</td>
</tr>
<tr>
<td>7</td>
<td>Regular SWNT</td>
<td>2-2-2</td>
<td>541.02</td>
<td>0.87</td>
</tr>
<tr>
<td>8</td>
<td>Regular SWNT</td>
<td>1-2-1</td>
<td>294.64</td>
<td>2.33</td>
</tr>
<tr>
<td>9</td>
<td>Regular SWNT</td>
<td>1-2-1</td>
<td>289.56</td>
<td>3.09</td>
</tr>
<tr>
<td>10</td>
<td>MWNT</td>
<td>1-1-1</td>
<td>294.64</td>
<td>1.12</td>
</tr>
<tr>
<td>11</td>
<td>MWNT</td>
<td>1-1-1</td>
<td>276.86</td>
<td>0.86</td>
</tr>
<tr>
<td>12</td>
<td>MWNT</td>
<td>2-2-2</td>
<td>482.60</td>
<td>0.67</td>
</tr>
<tr>
<td>13</td>
<td>MWNT</td>
<td>2-2-2</td>
<td>500.38</td>
<td>0.80</td>
</tr>
<tr>
<td>14</td>
<td>IL 55%</td>
<td>1-1-1</td>
<td>302.26</td>
<td>0.83</td>
</tr>
<tr>
<td>15</td>
<td>IL 55%</td>
<td>1-1-1</td>
<td>203.20</td>
<td>0.72</td>
</tr>
<tr>
<td>16</td>
<td>IL 55%</td>
<td>1-1-1</td>
<td>269.24</td>
<td>0.90</td>
</tr>
<tr>
<td>17</td>
<td>SWNT 12%</td>
<td>1-1-1</td>
<td>274.32</td>
<td>0.82</td>
</tr>
<tr>
<td>18</td>
<td>SWNT 12%</td>
<td>1-1-1</td>
<td>231.14</td>
<td>0.78</td>
</tr>
<tr>
<td>19</td>
<td>SWNT 12%</td>
<td>1-1-1</td>
<td>342.90</td>
<td>1.03</td>
</tr>
<tr>
<td>20</td>
<td>SWNT 27%</td>
<td>1-1-1</td>
<td>360.68</td>
<td>0.86</td>
</tr>
<tr>
<td>21</td>
<td>SWNT 27%</td>
<td>1-2-1</td>
<td>355.60</td>
<td>1.56</td>
</tr>
<tr>
<td>22</td>
<td>SWNT 27%</td>
<td>1-2-1</td>
<td>292.10</td>
<td>1.48</td>
</tr>
</tbody>
</table>
Figure 19 Average displacement of (a) SWNT and (b) MWNT BGA samples (with different thickness ratios and thicknesses from 256.54 to 546.10 µm) vs. voltage.
Figure 20 Average displacement of SWNT and MWNT BGA samples (at several applied voltages with different thickness ratios and thicknesses from 256.54 to 546.10 µm) vs. frequency.

Figure 21 Average displacement per unit length of BGA samples with different weight fractions of constituents at different frequencies and 10 V.
4.2.2 Weight fraction of constituents

The effect of weight fraction of constituents on the performance of BGA was studied by fabricating four different sets of samples with various proportions. Displacement of all sets of samples was measured at different voltages and frequencies. The average displacement of all tested specimens showed no statistically significant difference when weight fractions varied as shown in Figure 21. This study also demonstrated that the difference got even narrower at lower applied voltages. Based on these observations, the effect of weight fraction of constituents on the displacement of the BGA was assumed to be negligible.

4.2.3 Thickness, Thickness ratio

BGA thickness was one of the first parameters ever studied to see its effect on performance. Preliminary studies by Mukai et al. [11] showed that BGA displacement is reduced by increasing specimen thickness. However, this result was based on tests conducted on only two different thicknesses. Optimizing the performance and establishing a theoretical model for BGA displacement required a more comprehensive study on the effect of thickness on BGA displacement. There is currently no detailed investigation found on the performance of the BGA at different thicknesses, to the best of our knowledge.

Thickness and thickness ratio were found to be key factors in achieving maximum displacement. Thicker BGA samples generated smaller displacements under the same applied voltage. As shown in Figure 22a, the displacement of BGA samples decreased linearly with total thickness. This smaller displacement could be justified by increase in moment of inertia and bending stiffness of BGA with increase in specimen total thickness. Total thickness was not the only investigated parameter in this study. Thickness ratio was introduced as a new parameter to investigate the effect of each layer thickness on BGA displacement. It was observed that for
samples with a thickness ratio in a certain range (0.9-2.5 for SWNT samples) the displacement is improved significantly (Figure 22b). For example, the maximum displacement of a sample with thickness of 513 µm and thickness ratio of 1 was larger than a sample with half the thickness; 261 µm but thickness ratio of 0.6. The larger displacement of the thicker sample could only be explained by the thickness ratio, since all the other parameters were the same for both samples. This revealed that the thickness of electrolyte layer; as the medium for free ions, plays an important role in BGA displacement. Therefore, considering both the total thickness and thickness ratio in BGA fabrication is recommended to achieve maximum displacement.

Figure 22 (a) Effect of BGA thickness on the displacement per unit length for SWNT and MWNT samples at 10 V and two different frequencies (b) displacement of Regular SWNT samples with various thickness ratios (10 V-0.1 Hz).
4.2.4 Lifetime

BGA lifetime was monitored by oscillating regular SWNT samples continuously under constant voltage and frequency for several days. It was observed that the displacement of all tested BGAs decreased over time displaying a logarithmic trend. The amplitude of displacement dropped rapidly and then gradually reached a plateau (for example: 37% in the first 10,000 cycles and 13% reduction from 10,000 to 50,000 cycles for sample tested at 10V) (Figure 23). This reduction in displacement over time was more severe at higher voltages, and this could be explained by the electrochemical window of the ionic liquid. Electrochemical window of a substance is the range of the voltage in which the substance does not get oxidized or reduced. If the value of the applied voltage is higher than the electrochemical window, the ionic liquid starts to decompose. Electrochemically stable window of EMIBF4 is reported to be 4V [42], and therefore driving BGA under voltages higher than that for a long time should cause significant reduction in BGA morphing efficiency.

![Figure 23 Change in maximum deflection of SWNT BGAs at different voltages by time.](image-url)
Lifetime tests were conducted on MWNT BGA samples as well. Lifetime of MWNT samples was found to be remarkably lower than the SWNT samples (Figure 24). For example, the displacement of a MWNT sample under 7.5V and 0.1 Hz went down linearly to almost zero after only 7,000 cycles (~ 20 hours). Whereas a SWNT sample under same applied voltage and frequency showed a %35 reduction in displacement amplitude after 58,000 cycles (~165 hours). Based on these observations, BGA lifetime should definitely be considered in design and development of novel applications for BGA. Reduction in BGA displacement over time can be a major challenge in applications where continuous oscillation is required. However, BGA lifetime can be improved by using single-walled CNT and lower driving voltages.

**Table 6**: Average displacement of the BGA samples days after first test.

<table>
<thead>
<tr>
<th>DAY</th>
<th>2V AVE (mm)</th>
<th>2V STD</th>
<th>5V AVE (mm)</th>
<th>5V STD</th>
<th>10V AVE (mm)</th>
<th>10V STD</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.316</td>
<td>0.130</td>
<td>0.993</td>
<td>0.363</td>
<td>2.319</td>
<td>0.723</td>
</tr>
<tr>
<td>8</td>
<td>0.186</td>
<td>0.043</td>
<td>0.558</td>
<td>0.127</td>
<td>1.377</td>
<td>0.294</td>
</tr>
<tr>
<td>13</td>
<td>0.227</td>
<td>0.077</td>
<td>0.762</td>
<td>0.226</td>
<td>1.969</td>
<td>0.490</td>
</tr>
<tr>
<td>20</td>
<td>0.483</td>
<td>0.090</td>
<td>1.296</td>
<td>0.184</td>
<td>2.842</td>
<td>0.411</td>
</tr>
<tr>
<td>34</td>
<td>0.355</td>
<td>0.110</td>
<td>0.952</td>
<td>0.304</td>
<td>2.613</td>
<td>0.599</td>
</tr>
<tr>
<td>71</td>
<td>0.350</td>
<td>0.077</td>
<td>0.943</td>
<td>0.227</td>
<td>2.119</td>
<td>0.364</td>
</tr>
</tbody>
</table>

Figure 24 Lifetime of SWNT and MWNT BGAs at 7.5 V.
4.2.5 Degradation over time

Maximum displacement of several samples was measured under different applied voltages and frequencies. Samples were stored at room temperature and atmospheric pressure. The same tests were repeated after several weeks to study the possible degradation in the maximum displacement of BGA.

Different 1-1-1 regular SWNT BGA samples were prepared for this study. Displacement tests were carried out under different driving voltages (2, 5, and 10V) and a constant frequency (0.1 Hz). Same tests of the sample were repeated after 13, 20, 34, and 71 days to check if there was any changes in displacement. Room temperature was recorded on each test to assure that the effect of temperature on BGA displacement was eliminated.

![Figure 25 Change in the average maximum displacement of BGA samples by over weeks of being stored.](image-url)
As shown in Figure 25, the maximum displacement of the BGA samples displayed some variations over weeks of being stored. BGA samples displacement values were within the range of the average at each interval over seventy one days (Table 7). Despite this variation in BGA displacement over weeks, no distinct decay or improvement in BGA performance was observed. The general trend of BGA displacement over weeks proved to be constant. Therefore, based on our observations there was no remarkable degradation in BGA maximum displacement over time after being stored (Figure 25).

4.3 Maximum displacement model

Different studies have been conducted to characterize BGA displacement based on geometrical and mechanical properties. Takeuchi [12] calculated the induced strain in a BGA strip based on expansion and contraction of the electrode layers and curvature of the actuator. This induced strain was calculated based on geometrical relation between the maximum displacement (δ), length (L), and thickness of each layer of the BGA (d).

\[
\varepsilon = \frac{2d\delta}{L^2 + \delta^2} \tag{Equation 7}
\]

However, this strain was defined based on geometrical relations only. Therefore, another parameter needed to be found in order to incorporate the effect of the material.

Table 7 Average, standard deviation, and coefficient of variation of BGA samples at different driving voltages over 71 days.

<table>
<thead>
<tr>
<th>Driving voltage (V)</th>
<th>Average displacement (mm)</th>
<th>Standard Deviation</th>
<th>Coefficient of Variation</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>0.320</td>
<td>0.088</td>
<td>0.275</td>
</tr>
<tr>
<td>5</td>
<td>0.917</td>
<td>0.238</td>
<td>0.260</td>
</tr>
<tr>
<td>10</td>
<td>2.206</td>
<td>0.480</td>
<td>0.218</td>
</tr>
</tbody>
</table>
Alici et al. introduced a new parameter, generated strain ($\alpha$), to model the bending motion of tri-layered redox-driven polypyrrole polymer (PPy) actuators [43]. This parameter was calculated based on momentum equilibrium of a beam subjected to bending caused by electrode deformations. Major assumptions in this model were isotropic elastic behavior of all constituents, constant and uniform thickness, linear strain distribution, and no change in elastic modulus under different voltages and frequencies [8, 44]. Generated strain ($\alpha$) was assumed to be constant and represented by:

$$\alpha = \frac{EI}{RE_1wt_1(t_1 + t_2)}$$

Equation 8

where $EI$, $t$, $R$, and $w$ represent flexural stiffness, thickness, radius of curvature, and width, respectively. Subscripts 1 and 2 represent electrode and electrolyte layer, respectively. Flexural stiffness represents the resistance of the beam to bending, and is equal to [8, 11]:

$$EI = \frac{E_1w((2t_1 + t_2)^3 - t_2^3)}{12} + \frac{E_2wt_2^3}{12}$$

Equation 9

In addition to the generated strain parameter, Alici et al. [44] established a maximum displacement model for PPy actuators (Equation 10). This model was derived based on the assumption that the bending moment was solely the result of expansion and contraction of the electrode layers. This model (Equation 10) described the bending motion in terms of applied voltage, capacitance, geometrical parameters, and mechanical properties.

$$y'' - [1 + (y')^2] \frac{3\alpha VC(t_1 + t_2)}{2wL[E_1(t_1^3 - t_2^3) + E_2t_1^3]}$$

Equation 10

Mukai [11] and Ceseracciu [8] have adapted the generated strain parameter to characterize the mechanical response of BGA. Generated strain characterizes the bending motion of BGA better than the strain defined only by geometry, since it takes in consideration the
mechanical properties of the actuator too. However, using the displacement model suggested by Alici et al. (Equation 10) for BGA had several shortcomings. In Alici model, the effect of the applied frequency, which showed a significant influence on BGA displacement, was neglected. Moreover, capacitance was calculated based on the current-time response of the actuator. Hence, this model if used would require additional current measurements. In addition, obtaining maximum displacement with this model would dependent on solving a nonlinear second-order differential equation. Therefore, establishing a displacement model for BGA was essential to facilitate the development of novel applications.

In this study, a new model was established using nonlinear regression to predict displacement of BGA. NLREG (Version 6.4) software (Sherrod, 2008) was utilized for the regression analysis. Different regression methods such as multivariate, linear, polynomial, exponential, logistic, and general nonlinear regression could be performed using NLREG. NLREG employs an adaptive nonlinear least-squares algorithm to determine the values of the parameters that minimize the sum of the squared residual values for the set of observations [45].

Based on the experimental observations, it was concluded that voltage, frequency, thickness and thickness ratio influenced BGA displacement. These parameters were used in the regression analysis. Different linear and nonlinear functions were fitted to all sets of observations. Among the tested functions, Equation 11 fitted the data very well (Figure 26).

\[
\left( \frac{Y}{L} \right)_{max} = a \left( EI \right)^{-b} \left( \frac{V}{t} \right)^{c} \left( \frac{t_{r}}{f} \right)^{d}
\]

Equation 11

where \( Y, L, V, f, t, \) and \( t_{r} \) are displacement, length, applied voltage, frequency, thickness and thickness ratio, respectively. \( a, b, c, \) and \( d, \) are material constants.
For BGA fabricated with regular weight fractions of SWNT and MWNT, the material constants and adjusted coefficient of multiple determination ($R^2$ value) are presented in Table 8. This model fitted the SWNT BGA experimental data accurately ($R^2 = 0.9793$) whereas the same model did not fit the MWNT BGA samples very well ($R^2 = 0.7573$).

An adaptable displacement model would be a major advancement in BGA application, since BGA performance enhancement by using different constituents is an ongoing field of research. Therefore, this model was fitted to the results reported by Fukushima [4], Mukai [11], and Biso [16] to verify the model adaptability. The reported results selected for the verification were picked from references in which different types of SWNT (pristine SWNT, SWNT-COOH, SWNT-amide, Super growth CNT) and ionic liquids (EMIBF$_4$, BMIBF$_4$, BMIMBF$_4$) were utilized in BGA fabrication.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Material</th>
<th>a</th>
<th>b</th>
<th>c</th>
<th>d</th>
<th>$R^2$-value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ghamsari et al.</td>
<td>Pristine SWNT + EMIBF$_4$</td>
<td>5528.271</td>
<td>0.238</td>
<td>1.500</td>
<td>0.713</td>
<td>%97.93</td>
</tr>
<tr>
<td></td>
<td>MWNT-COOH + EMIBF$_4$</td>
<td>3.424</td>
<td>-0.158</td>
<td>0.956</td>
<td>0.871</td>
<td>%76.28</td>
</tr>
<tr>
<td>Fukushima et al.[4]</td>
<td>Pristine SWNT + BMIBF$_4$</td>
<td>4.123</td>
<td>1.218</td>
<td>-0.045</td>
<td>0.593</td>
<td>%97.40</td>
</tr>
<tr>
<td>Biso et al. [16]</td>
<td>Pristine SWNT + BMIMBF$_4$</td>
<td>1.653</td>
<td>-0.694</td>
<td>-0.423</td>
<td>0.658</td>
<td>%96.90</td>
</tr>
<tr>
<td></td>
<td>SWNT-COOH + BMIMBF$_4$</td>
<td>1.469</td>
<td>-0.667</td>
<td>-0.407</td>
<td>0.762</td>
<td>%98.15</td>
</tr>
<tr>
<td></td>
<td>SG-CNT + BMIMBF$_4$</td>
<td>3.262</td>
<td>-0.501</td>
<td>-0.608</td>
<td>0.679</td>
<td>%96.26</td>
</tr>
<tr>
<td></td>
<td>SW-amide + BMIMBF$_4$</td>
<td>3.013</td>
<td>-0.483</td>
<td>-0.677</td>
<td>0.649</td>
<td>%92.64</td>
</tr>
</tbody>
</table>
Figure 26 The model fitted to the (a) regular SWNT and (b) MWNT BGA displacement tests conducted with different applied voltage, frequency, and thickness.
Table 9 The assumptions used for comparing the model prediction to other reported results.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Thickness (µm)</th>
<th>Thickness ratio</th>
<th>Length (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mukai et al. [11]</td>
<td>465,95</td>
<td>0.03,0.19</td>
<td>10</td>
</tr>
<tr>
<td>Biso et al. [16]</td>
<td>350</td>
<td>0.5</td>
<td>10</td>
</tr>
<tr>
<td>Fukushima et al. [4]</td>
<td>280</td>
<td>1</td>
<td>10</td>
</tr>
</tbody>
</table>

There were two challenges in conducting model verification. First, in most of the reported studies, the displacement was reported graphically and in the form of strain. Therefore, data synthesis had to be carried out to obtain the values of displacement. The displacement values were obtained by digitizing the reported graphs using Engauge Digitizer (Version 4.1) software (Mitchell, 2002). Second, since not all the required parameters were accurately reported, some assumptions had to be made. For example, since the individual thickness of each layer was not reported by Biso [16], the thickness of electrode and electrolyte layers was assumed to be equal. The assumed values used for different references are presented in Table 9.

The material constants obtained for different cases are presented in Table 9. The suggested model (Equation 11) fitted the reported results well with a high $R^2$ value (Table 8). Another source of error could be the inaccuracies in the assumed values used for this reference. However, the capability of the model to fit different cases well; at a wide range of frequency (Figure 27), suggested that this model can be used for BGA samples fabricated with different types of ionic liquid, single-walled carbon nanotubes, fabrication method, size, etc. The only requirement would be to conduct a few experiments on samples to obtain the material constants. Moreover, this function with minor changes in the current effective parameters and addition of new ones could be useful in characterizing the response of similar electroactive actuators.
Figure 27 Comparing the model prediction to the other reported results [16-18].

Here is the highlight of the displacement study:

- Average maximum displacement changed linearly with applied voltage
- Maximum displacement decreased by frequency in a power function trend
- Effect of weight fraction of components on BGA performance proved to be negligible.
- BGA displacement decreased linearly with increase in BGA total thickness, regardless of the used CNT type
- Thickness ratio was effective on BGA performance, and is improved for thickness ratios between 0.9 and 2.5.
• Lifetime of BGA was a function of applied voltage and the Carbon nanotube type. MWNT samples showed remarkably lower lifetime.
• There is no degradation in BGA performance
• A new model was established to predict the BGA displacement
• The model was verified by other reported results in the literature
CHAPTER 5. BGA APPLICATION IN MICROFLUIDICS

5.1 Different actuation mechanisms in microfluidics

Microfluidics chips (lab-on-a-chip) and devices are one of the most attractive fields of application for BGA and other electro-active materials. Various types of MEMS-based microfluidic mechanisms (micropump, micromixer, and microvalve) have been utilized in drug delivery and other biomedical applications [46]. Due to the rapid development of biocompatible polymers and microfabrication techniques, the design and fabrication of low-cost, easy-to-use and disposable microstructures has become more feasible [26]. However, from a practical solution standpoint, the successful miniaturization and commercialization of fully integrated microfluidic systems have been delayed due to the lack of reliable on-chip components, such as micropumps and microvalves [27].

Different materials employing different mechanisms and stimuli have been investigated to be utilized in microfluidic applications. Hydrogel [47], Ionic Polymer Metal Composite (IPMC) [48-51], and conducting polymers [52-54] (Figure 28) are examples of electroactive materials that have been used in microfluidics. Despite remarkable reported results in each case, each approach has shortcomings that limit their use in real life applications. Hydrogel actuators in microfluidics are limited because of bubble generation, slow response time, and difficulties in precise and rapid motion control [47] (Figure 29). Utilizing IPMC as a diaphragm micropump is also limited by complicated fabrication process and degradation in performance over time [49]. Conducting polymers major shortcoming is their typical slow response time and short lifetime, since their bending motion is caused by a reduction-oxidation (redox) process [53].
Piezoelectric actuation is another electroactive effect which is widely used in microfluidics. Bu et al. employed piezoelectric disks to design and fabricate a novel peristaltic micropump for DNA amplification [55]. Piezoelectric actuation can yield high forces, but very small deflections even with very large voltages. The drawback of small deflection was overcome by Piezoelectric microvalve [56-58], hydraulically amplified piezoelectric [59], stacked piezoelectric discs [60, 61], and piezo bimorphs [62, 63]. However, piezoelectric actuated microstructures still require high voltage for operation. In addition, the use of piezo-elements motion generator in some micropumps has resulted in complicated structures [52].

Figure 28 (a) Micropump assembly (b) Conjugated polymer membrane used as diaphragm pump (modified from [53]).

Figure 29 (a) Schematic view of electrically-driven hydrogel sorter (b) examples of droplet sorting (water phase flow rate = 6µL/min) (modified from [47]).
Considering the disadvantages of these actuation mechanisms, BGA’s fast response time, low power consumption, simple fabrication and low driving voltage were compelling reasons to study the possibility of utilizing this electrically-actuated smart material in microfluidics.

5.2 BGA morphing diaphragm for possible application in micropump

The possibility of fabrication and use of BGA in large scales was explored to evaluate the capability of this actuator for microfluidic applications. All the tested BGA samples in mechanical characterization and displacement study were fabricated in the form of rectangular stripes and size of ~40x10 mm. Attempts were made to make BGA samples in square shape to be used as a morphing diaphragm. However, the maximum displacement of these samples was remarkably lower than what was expected. Therefore, the major challenge was to study and enhance the performance of BGA in diaphragm form. Fabrication of a morphing BGA diaphragm would have been very useful in facilitating its application in microfluidic devices, motion sensors, etc.

High surface resistance was suspected to be the source of less efficient performance of BGA in diaphragm form. Surface conductivity had to be studied and evaluated to confirm this hypothesis. A thin layer of gold was deposited on BGA diaphragms to improve surface conductivity.

<table>
<thead>
<tr>
<th>Table 10 Parameters used in gold-coating process.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Adhesion layer</td>
</tr>
<tr>
<td>Conductive layer</td>
</tr>
<tr>
<td>Deposition rate</td>
</tr>
<tr>
<td>Working pressure</td>
</tr>
<tr>
<td>Maximum output power</td>
</tr>
</tbody>
</table>
The gold layer was deposited using Temescal Model BJD-1800 E-Beam Evaporator (Livemore, USA). However, the deposited gold layer was easily displaced during testing negating its potential use, since the adhesion of the gold layer to BGA samples was not strong. Depositing a layer of Cr on the samples as “adhesion promoter” solved this issue. The final thickness of the Cr and Au layers was around 100Å and 1000Å, respectively (Figure 30). The parameters used in deposition process are presented in Table 10.

The coated BGA diaphragms showed ~2-3 times higher maximum displacement compared to non-coated samples. Although this proved the hypothesis regarding the effect of surface resistance, the maximum displacement of BGA diaphragm was still not sufficient for the micropump application that was originally considered for BGA diaphragm.

**5.3 BGA microvalve**

Observation of the low maximum displacement in BGA diaphragms, led to focusing on the application of BGA in smaller scales. Utilizing BGA as a microvalve was an application that was looked into in this study. Microvalves have been the key components to control microflow for reliable operation of microfluidic devices. The applications of the microvalves include flow
regulation, on/off switching, or sealing of biomolecules, micro or nano particles, chemical reagents, oils, water, air bubble, and many others. Many different approaches have been adopted in the design and fabrication of microvalves.

According to Oh and Ahn [27], microvalves could be roughly categorized in two major groups: active microvalves, using mechanical and non-mechanical moving parts, as well as external systems, and passive microvalves, using mechanical and non-mechanical moving parts (Figure 31). Since BGA microvalve falls under active mechanical group, common mechanisms used in this area is briefly reviewed in the following section. Most active mechanical microvalves actuate using thermal, magnetic, and electric mechanisms (Figure 32).
Figure 32 Different types of actuation mechanisms of active microvalves with mechanical moving parts: (a) electromagnetic; (b) electrostatic; (c) piezoelectric; (d) bimetallic; (e) thermopneumatic; (f) shape memory alloy (modified from [27]).

Different types of thermally-actuated active microvalves using bimetallic [64, 65], thermopneumatic [66-68], and shape memory alloy actuations [69-71] have been reported. Jerman et al. [64] developed a bimetallically driven microvalve providing proportional control of the flows in the range of $0 - 0.151$ L/min with minor leakage. This type of active microvalves can generate higher forces while providing large strokes. However, the response time in this group of microvalves is relatively slow and the power consumption is high. Moreover, thermal actuation may not be suitable for many fluids due to heat dissipation.
Shape memory effect has also been an attractive thermally-actuated approach, since high output force could be generated with rather simple design and compact structure [72, 73]. Shape Memory Alloy (SMA) thin films [69, 74] and more recently SMA wires [75, 76] have been applied in design and fabrication of microvalves. High force-to-volume ratio, large strain recovery, and low amount of heat required for operation makes SMA wires a better candidate for microfluidic applications. However, the response time of SMA microvalve is slower than other alternatives. Furthermore, high phase transformation temperatures lead to longer required cooling time and subsequently a limit in the maximum operation frequency [71].

Different active microvalves using magnetic actuation have also been reported [77, 78]. Micro ball valve reported by Oh et al. [78] showed the capability of being operated bidirectionally and also be easily integrated into microfluidic systems. However, the device showed a leakage flow rate of 30 µL/min at 2.1 kPa, and the power consumption was still relatively high. Passive microvalves have also displayed good capabilities to be employed in microfluidics. It is demonstrated that capillary forces can be utilized in passive actuation in microscale. The passive capillary effect utilizes dependence on the geometries [79-81], or surface properties (hydrophobic or hydrophilic surfaces) [82, 83]. Passive microvalves using the passive capillary effect: abrupt [79, 84], burst [81, 85, 86], hydrophobic surfaces have been reported [82, 83].

While active valves control flow rate by pressure difference and have complex structures, passive valves are only open to forward pressure and have simple structures, showing diode-like characteristics [27]. In the reverse flow direction the valving efficiency of passive valves is relatively poor, since the performance of these valves depends on the input pressure [27].
Despite being very effective, active mechanical microvalves of microvalves has several disadvantages; such as unavoidable leakage flow rate, relatively high cost due to their complicated structures, etc. Though leakage flow becomes a critical feature for on/off switching applications, it is not critical for flow regulation. Many micromachined microvalves have been used for gas or selected liquid regulations [59, 63, 77, 87-90]. BGA remarkable characteristics showed great potential to be used in microfluidics as an active microvalve. However, there were two primary concerns: bubble generation and capability in generating enough force.

A primary concern in utilizing BGA as an electroactive microvalve was the possibility of bubble generation. Bubble generation is a common issue when electroactive materials are employed in microfluidics. Thus, underwater performance of the BGA was tested to monitor displacement and bubble generation. The BGA samples in this study were tested under driving voltage of 2-10V, and no bubble generation was observed (Figure 33).

Figure 33 BGA sample being operated under water.
Capability of providing enough force for valving operation was another primary concern. BGA generated blocking force in each stroke had to be studied to verify its capability in being used as a microvalve. The BGA generated blocking force has been studied theoretically and experimentally [11, 18]. The reported results on BGA force were in the order of the reported forces required for microvalves in different references [27, 47] (Figure 34). Simple experiments were carried out to verify that BGA can generate enough force. As shown in Figure 35, BGA was able to lift objects weighing 1.2 grams, in addition to its own weight.
Rapid response time, low driving voltage, ease of fabrication, the lack of bubble generation, and capability of providing sufficient force make BGA a potential and promising candidate in flow regulation for microfluidic applications.

Microvalves have an important role in the operation of microfluidic devices. The desired characteristics of microvalves include no leakage flow, reduced dead volume, reduced power consumption, large pressure resistance, normally closed or open mode, insensitivity to particulate contamination, rapid response time, potential for linear operation, ability to operate with both liquids and gases and disposability [27, 91]. As discussed, various approaches have been explored in the development of microvalves. Apparently, there is no single microvalve that works for all applications ranging from life sciences to vacuum. Only a subset of the selected characteristics of a specific microvalve is important in a given application.

5.3.1 Design and modeling

After a convincing primary study of BGA capabilities, a device was designed to study the performance of the BGA as a microvalve. In addition to size, weight, and durability, the following parameters were considered in the design of this device:

- The device should have the capability to be easily disassembled, so that replacing the valve to test and study different BGA samples is feasible,
- Cross section of the channel should be adjustable to test different section areas,
- Device should have proper sealing to avoid any leakage,
- Capability of visual inspection
- While most BGA is in direct contact with the fluid, isolation between the BGA electrical contacts and passing fluid should be provided.
Based on these criteria a simple and yet practical device was designed. This device has two major parts: a base part (Figure 36) and a transparent PMMA cover. These two parts could be assembled using four M4x10 bolts (Figure 37). Instead of a fixed channel with certain shape and size, a tube-housing was considered in the device. Therefore, by using tubes with various inner diameters, different section areas could be tested and studied. The output fluid could be transferred to a container via a secondary tube for measurement purposes. A groove was also considered around the base main chamber for sealing purposes (Figure 36).

Electrical contacts and wiring was another issue that had to be addressed in the design of this device. Electrical contacts had to be isolated from the fluid passing through the microvalve device. Therefore, a part of BGA with electrical contacts had to be isolated from the fluid while the other part of BGA had to be in direct contact with the fluid. A secondary smaller chamber, isolated from the main chamber was included in the base part to solve this issue. Conductive silver paste (TED PELLA Inc., Redding, California) was used to connect the wires to BGA.

Figure 36 Base part of the designed microvalve device.
Figure 37 Exploded view of the microvalve device assembly.

Figure 38 3D printer major units.
5.3.2 Fabrication

The microvalve base part was fabricated using a 3D printing technique. 3D printing is a rapid prototyping method by which durable plastic parts can be produced fast and with high resolution. *V-Flash® FTI 320 desktop modeler* (Rock Hill, USA) was employed as the 3D printer (Figure 38). This 3D printer had a resolution of 768 x 1024 DPI (x-y plane) and could print layers with thickness of 102 microns. 3D printing process was accomplished in three stages: printing, washing, and curing.

Printing of designed microvalve base with the V-flash 3D printer required a 3D CAD model. The base part was modeled employing SolidWorks® 2009 software, and then imported into the machine software. Printing set-up, parts arrangement, and build optimization was managed through this software (Figure 39). Arrangement of the parts showed a great impact on the precision of the designed features and also the fabrication time. Printing of the microvalve base part was performed in two hours (Figure 40).

![Figure 39 Arrangement of the parts on the build pad in 3D printing.](image)
Washing the printed part was the next step which was carried out to remove the resin residue off the part features. Washing was essential for this specific part, since there were several small features. Printed base part was washed in the appropriate solvent and subsequently in water for thirty minutes.
Ultraviolet (UV) curing was the final stage in the 3D printing process. Exposure of the printed part to the UV energy causes the liquid monomer to harden instantly which facilitates the bonding and improves the scratch and solvent resistance. After washing, parts were dried and cleaned before being transferred to the curing UV oven. Curing time was set to 20 minutes based on the size of the part. The final part is presented in Figure 42.

The second part of the designed device was the transparent cover. The cover of the base was cut from a PMMA sheet with a thickness of 3 mm. Four M4x10 bolts were used to assemble this cover on the base part. Rubber flat washers were used to dampen the pressure caused by the bolts to prevent any damage to the PMMA cover. Graphite valve packing was utilized to seal the chamber and prevent any leakage.

5.3.3 Experimental setup

An apparatus was designed and set-up to study the efficiency of the BGA microvalve in manipulating the flow rate. This apparatus had to be able to provide a constant fluid flow. In addition, the output flow should be easy to measure.
Bartels MP6 micropump was utilized to generate the necessary flow rate measurement (Figure 43). This compact-size piezo membrane micropump was capable of transporting gases or liquids with low energy consumption and a broad performance spectrum [92]. Bartels controller MP-X was also used to drive the micropump by alternating either voltage (up to 250V) or frequency (0-300 Hz). Different flow rates could be achieved by varying these parameters and the signal shape. SRS-signal was recommended by the manufacturer to be typically the optimal setting for aqueous solutions [92]. Micropump specification is presented in Table 11.

A regular SWNT BGA with 2-2-2 layer combination was employed in this device to operate as a microvalve. The total thickness of the BGA strip was ~500 µm, with thickness ratio of 0.9. Based on the displacement study, the thicker 2-2-2 BGA samples would show lower displacement. However, thicker regular SWNT BGA was selected since higher generated blocking force was desirable rather than larger displacement. A layer of PDMS was put on the BGA samples to increase the durability. The PDMS layer would also increase the adhesion
between the microvalve and fluid inlet, providing better sealing. The PDMS layer was casted on the BGA utilizing SYLGARD® 184 Silicone Elastomer kit (Midland, USA).

After assembling the device, Tygon® tubing with inner diameter of 3 mm was used for connecting the reservoir, micropump, microvalve chip and the output container. Colored water was chosen as the fluid for all the flow rate measurements.

The micropump was driven by a SRS-shape signal at 100V and 80 Hz to provide the input flow rate. This configuration was kept constant for all tests. Assuming that the micropump provides constant flow, BGA was driven by a rectangular signal at two different voltages (8 and 10V) and six frequencies (250, 125, 100, 50, 25, and 10 mHz). Varying voltage and frequencies were applied to check the possibility of precise flow control with the BGA as a microvalve. BGA was driven at %50 duty cycle which means that BGA valve was “ON“ at half of the period and was “OFF” for the other half. Flow rate tests were also conducted at 0 Hz frequency (%100 ON) to measure the leakage of the valve. The flow rate measurement setup is presented in Figure 44.

<table>
<thead>
<tr>
<th>Table 11 MP6 micropump technical information [92].</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Pump Type</strong></td>
</tr>
<tr>
<td><strong>Dimensions</strong></td>
</tr>
<tr>
<td><strong>Weight</strong></td>
</tr>
<tr>
<td><strong>Power consumption</strong></td>
</tr>
<tr>
<td><strong>Pumping media</strong></td>
</tr>
<tr>
<td><strong>Signal shape</strong></td>
</tr>
<tr>
<td><strong>Typical flow rate</strong></td>
</tr>
<tr>
<td><strong>(250 V, SRS signal)</strong></td>
</tr>
<tr>
<td><strong>Typical back pressure</strong></td>
</tr>
<tr>
<td><strong>(250 V, SRS signal)</strong></td>
</tr>
</tbody>
</table>
Figure 44 Flow rate measurement setup (1) Bartels controller MP-X (2) Agilent 33220A function generator (3) fluid reservoir (4) Bartels MP6 mini-pump (5) Microvalve device (6) Output container (7) Agilent 54622A Oscilloscope.

Figure 45 Fabricated microvalve device.
Table 12 Sampling time selected for different frequencies.

<table>
<thead>
<tr>
<th>Frequency (mHz)</th>
<th>Period (sec)</th>
<th>Sampling time (sec)</th>
</tr>
</thead>
<tbody>
<tr>
<td>250</td>
<td>4</td>
<td>120</td>
</tr>
<tr>
<td>125</td>
<td>8</td>
<td>120</td>
</tr>
<tr>
<td>100</td>
<td>10</td>
<td>120</td>
</tr>
<tr>
<td>50</td>
<td>20</td>
<td>120</td>
</tr>
<tr>
<td>25</td>
<td>40</td>
<td>240</td>
</tr>
<tr>
<td>10</td>
<td>100</td>
<td>600</td>
</tr>
</tbody>
</table>

Flow rate was measured by weighing the output of the system over a certain sampling time (Figure 45). Sampling time (multiple of the time period) was selected based on the frequency at which BGA was being operated (Table 12). Each test was repeated three times to evaluate the repeatability of the data. The average of three flow rate readings was reported as the measured flow rate.

5.3.4 Results

The mass of the fluid passed through the microvalve during the sampling time was measured and then converted to volume. Based on the volume of the output fluid and sampling time, the flow rate was calculated. The average flow rate and standard deviation of each test is presented in Table 13.

The output flow rate of the system was varied by the driving voltage and frequency of the BGA microvalve (Figure 46). At the lower tested voltage (8V), the flow of fluid passing through the device decreased with frequency, since at lower frequencies the valve would be in ON mode for a longer time. The output flow rate displayed a polynomial relation with frequency. At the frequency of zero (%100 ON mode), the output of the system was significantly reduced but did not reach zero (Figure 46).
Table 13: Output flow rate measured at different voltages and frequencies.

<table>
<thead>
<tr>
<th>Test</th>
<th>BGA Voltage (V)</th>
<th>BGA Frequency (mHz)</th>
<th>Sampling Time (min)</th>
<th>Average Flow rate (mL/min)</th>
<th>Standard Deviation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>8</td>
<td>250</td>
<td>2</td>
<td>2.248</td>
<td>0.026</td>
</tr>
<tr>
<td>2</td>
<td>8</td>
<td>125</td>
<td>2</td>
<td>2.154</td>
<td>0.005</td>
</tr>
<tr>
<td>3</td>
<td>8</td>
<td>100</td>
<td>2</td>
<td>2.047</td>
<td>0.143</td>
</tr>
<tr>
<td>4</td>
<td>8</td>
<td>50</td>
<td>2</td>
<td>1.504</td>
<td>0.097</td>
</tr>
<tr>
<td>5</td>
<td>8</td>
<td>25</td>
<td>4</td>
<td>1.447</td>
<td>0.027</td>
</tr>
<tr>
<td>6</td>
<td>8</td>
<td>10</td>
<td>10</td>
<td>1.281</td>
<td>0.046</td>
</tr>
<tr>
<td>7</td>
<td>8</td>
<td>0</td>
<td>2</td>
<td>0.702</td>
<td>0.103</td>
</tr>
<tr>
<td>8</td>
<td>10</td>
<td>250</td>
<td>2</td>
<td>0.635</td>
<td>0.052</td>
</tr>
<tr>
<td>9</td>
<td>10</td>
<td>125</td>
<td>2</td>
<td>0.554</td>
<td>0.025</td>
</tr>
<tr>
<td>10</td>
<td>10</td>
<td>100</td>
<td>2</td>
<td>0.520</td>
<td>0.017</td>
</tr>
<tr>
<td>11</td>
<td>10</td>
<td>50</td>
<td>2</td>
<td>0.475</td>
<td>0.039</td>
</tr>
<tr>
<td>12</td>
<td>10</td>
<td>25</td>
<td>4</td>
<td>0.371</td>
<td>0.052</td>
</tr>
<tr>
<td>13</td>
<td>10</td>
<td>10</td>
<td>10</td>
<td>0.237</td>
<td>0.043</td>
</tr>
<tr>
<td>14</td>
<td>10</td>
<td>0</td>
<td>2</td>
<td>0.171</td>
<td>0.019</td>
</tr>
</tbody>
</table>

At the higher voltage (10V), same flow rate response to frequency was observed. However, output flow rate was reduced significantly (216% - 441% based on the frequency) at this voltage level (Figure 46). BGA microvalve showed minor leakage of 0.702 and 0.171 mL/min at 8 and 10V, respectively.

Based on these observations, BGA microvalve displayed the capability to manipulate the flow rate in microfluidic devices. BGA microvalve was determined to provide different flow rates by varying different parameters: driving signal shape, applied voltage, frequency, BGA thickness, and BGA thickness ratio.
Figure 46 Measured flow rate at different frequencies and two different voltages. BGA was driven under a rectangular shape signal with 50% duty cycle.

Most microsystems require consistent and precise flow rate to operate, something that is typically difficult at microscales [93, 94]. Traditionally, flow regulation in microfluidic devices takes place off-chip, since miniaturization and integration of such devices on chip is expensive and complex [94-96]. BGA microvalve with simplicity of design, low power consumption, and ease of integration can be immediately applicable in microfluidic devices. The flow rate range generated with this device is also suitable for controlling biological conditions in human health care including drug delivery [96]. This microvalve system can also be used in adjustable point-of-care devices equipped with programmable gating of biological fluids for life science analysis and research.
CHAPTER 6. CONCLUSIONS AND FUTURE WORK

6.1 Conclusions

This study was focused on an electroactive morphing nanocomposite: bucky gel actuator. Three major phases were completed to accomplish this study: (1) Mechanical characterization, in order to identify the effective parameters and obtain the maximum mechanical strength (2) Displacement study, with the aim of BGA performance optimization and modeling the output to facilitate its applications (3) Explore the possibility of BGA application in microfluidics.

6.1.1 Mechanical characterization

Mechanical characterization was carried out via DMA and nano-indentation. Based on DMA and nano-indentation results, all three parameters (thickness, carbon nanotube type, weight fraction of constituents) were found to influence the mechanical properties of bucky gel actuator. DMA tests revealed that BGA storage modulus was greatly influenced by strain rate, thickness and carbon nanotube weight fraction. Higher carbon nanotube weight fraction and thickness resulted in higher storage modulus of BGA. Viscosity of BGA also showed to be dependent on the weight fraction of constituents and strain rate, but not on the thickness. Based on DMA results, regular SWNT BGA samples exhibited the highest strength to weight ratio among the tested samples.

Based on nano-indentation tests, the weight fraction of all three constituents affected the values of hardness, elastic modulus and adhesion. Carbon nanotube type was determined to have a major effect on all mechanical properties. Among all tested samples, regular SWNT BGA with 22 wt% of carbon nanotubes and 45 wt% of ionic liquid exhibited the best mechanical properties. This set of samples displayed the highest storage and loss moduli, viscosity, hardness and
reduced elastic modulus. However, Regular SWNT BGA also had the highest amount of adhesion among the tested samples. Compared to SWNT samples, MWNT samples showed remarkably lower adhesion.

### 6.1.2 Displacement study

Displacement study was conducted by examining the effect of five selected parameters. Voltage and frequency proved to have remarkable impact on BGA displacement. For most cases, average maximum displacement changed linearly with applied voltage. However, maximum displacement decreased with increasing frequency as a power function. The other studied parameter; weight fraction of components, had negligible effect on BGA performance. On the other hand, it was observed that thickness and thickness ratio had influence on displacement.

In the case of thickness, regardless of the CNT type used for BGA, displacement decreased linearly with increase in BGA total thickness. It was observed that the newly defined thickness ratio played an important role in BGA response. All the SWNT samples with thickness ratio between 0.9 and 2.5 showed improvement in their displacement. Therefore, considering not only the total thickness but also the thickness ratio in fabrication of BGA is highly recommended.

BGA lifetime was observed to be a function of the applied voltage and CNT type. All samples showed shorter lifetime at elevated voltage levels. Also, compared to SWNT samples, MWNT samples displayed much shorter lifetime. The maximum displacement of BGA after being stored for a while was monitored to investigate possible degradation of BGA. The maximum displacement of the BGA samples displayed some variations over weeks. Despite this
variation in BGA displacement, no distinct decay or improvement in BGA performance was observed. Therefore, based on our observations there was no degradation in BGA.

A new model was established utilizing non-linear regression to predict displacement of BGAs. The established model demonstrated an excellent fit to the SWNT BGA samples. This model also displayed a very good fit with other reported results of BGAs made with different types of CNT and ionic liquid. The material constants for each case were obtained. Based on this study, utilizing this displacement model is recommended for BGA samples fabricated with different types of SWNT and ionic liquids.

6.1.3 BGA application in Microfluidics

Preliminary studies showed that BGA can generate enough blocking force to be employed in microfluidic applications. A flow regulator utilizing BGA as a microvalve was designed and modeled. BGA microvalve device was assembled and then coupled by a micropump to make an experimental apparatus for flow rate measurements. BGA microvalve displayed capability to be utilized in regulating the flow rate in different ranges. BGA microvalve could provide different flow rates by varying different parameters: driving signal shape, applied voltage, frequency, BGA thickness, and BGA thickness ratio. BGA show the capability to be employed in different biological applications.

6.2 Future work

The following steps are suggested as future work to expand this research.

6.2.1 BGA coupled with photovoltaic cells

BGA will always require a power source to operate. Solar cells are attractive as power sources for MEMS since they are easily integrated and can be utilized as on-chip power supply.
in applications such as implantable bio-sensing [97, 98]. Coupling BGA with photovoltaic cells is a step forward in applying this micro-actuator in low-cost, disposable, point-of-care devices. Powered by solar cells, BGA becomes a promising candidate for mini-robotics, sensing, etc. in terrestrial and space applications.

6.2.2 BGA as an active micromixer

Generally, application fields of microchannel based mixers encompass both modern issues such as sample preparation for chemical analysis and traditional, widespread usable mixing tasks such as reaction, gas absorption, emulsification, foaming, and blending [99]. Different active (ultrasound, small impellers, etc.) and passive (collision of jets, nozzle injection in flow, etc) micromixing approaches have been reported. BGA exhibited a promising potential to be employed as an active micromixer. BGA has to be fabricated in microchannel size, in order to be used in such applications. Fabricating and utilizing BGA in that size requires overcoming some challenges like electrical contacts and wiring.

6.2.3 Electroactive actuator employing Deep Eutectic Solvents

A Deep Eutectic Solvent (DES) is a type of ionic solvent with special properties composed of a mixture which forms a eutectic with a melting point much lower than either of the individual components. Compared to ionic liquids which share many characteristics but are ionic compounds and ionic mixtures, DESs are cheaper to make, much less toxic, and sometimes biodegradable [100-104]. Deep Eutectic Solvents formed between a range of carboxylic acids and choline chloride has shown conductivity and ionic mobility similar to imidazolium based ionic liquids. Replacing ionic liquid by deep eutectic solvents should be studied to investigate its effect on BGA performance and mechanical properties.
REFERENCES


APPENDIX A. PERMISSION TO REPRODUCE PUBLISHED WORK

To: Permissions <permissions@iop.org>
Cc: 
Bcc: 
Subject: Re: requesting permission for using figures and tables of our paper in my thesis
From: Ali Kadkhoda Ghamsari <akadkh2@tigers.isu.edu> - Wednesday 06/06/2012 14:48

Dear Ms. Ryder,

I would like to get permission to reproduce the following figures and tables in my thesis:
Tables: 1,3,4
Figures: 2-8
Thanks in advance
Ali K. Ghamsari

PERMISSION TO REPRODUCE AS REQUESTED IS GIVEN PROVIDED THAT:

(a) the consent of the author(s) is obtained,
(b) the source of the material including author, title of article, title of journal, volume number, issue number (if relevant), page range (or first page if this is the only information available), date and publisher is acknowledged.
(c) for material being published electronically, a link back to the original article should be provided (via DOI).

IOP Publishing Ltd
Temple Circus
Temple Way
BRISTOL BS1 6BE
Date

Sarah Ryder
Rights & Permissions

86
VITA

Ali Kadkhoda Ghamsari was born in Tehran, Iran, in the year 1983. He completed his schooling from Bahonar’s high school in his hometown. His interest in practically of engineering and its capability in solving real-world problems led him to join the best manufacturing program in his country at Amir Kabir University of Technology, Tehran, Iran. He earned his Bachelor’s degree in manufacturing engineering in 2004. He joined the graduate program in manufacturing engineering at K. N. Toosi University of Technology, Tehran, Iran on the same year. While pursuing his MS degree, he started working as a research assistant for Modern Metal Forming laboratory and succeeded in publishing several papers in respected journals in the field of high strain rate loading. After graduating in 2007, He decided to pursue higher education and he enrolled in PhD program in mechanical engineering at Louisiana State University. Currently, he is working as a research assistant for NextGenC³ Composite Crest Center. He will be graduating in August 2012 with Doctoral degree in Mechanical systems.