Characterization of Polydimethylsiloxane Dielectric Films for Capacitive ECG Bioelectrodes

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***Abstract*—Capacitive ECG bioelectrodes are potentials for wearable and long-term physiological monitoring applications. In non-contact ECG recordings, the dielectric material sets limit to smooth bioelectric signal acquisition. Previously used dielectrics are rigid, unconformable on the skin, induce artifact and triboelectric noise, and becomes unstable when they absorb skin exudates. Recently, polymeric materials such as PDMS have gained different biomedical applications because it is biocompatible, flexible, and easy to fabricate. However, its use as a dielectric for capacitive ECG sensing is poorly reported. In this study, 15 samples of thin PDMS films of various thicknesses were fabricated by varying the proportion of the Sylgard 184™ silicone elastomer to the crosslinker from Dow Corning Corporation, and manually deposited on acrylic glass substrates. The composition ratio and thickness were used to tune the structure and dielectric properties of the films. The effects on the capacitance generated by each dielectric film were measured using the parallel plate method, and their corresponding values of relative permittivity was also estimated. The results obtained reveal that PDMS films made from a composition ratio of 10:2 yielded the maximum capacitance and relative permittivity. In contrast, the film with 0.14 mm thickness revealed the highest value of capacitance (31 pF). The recorded values of capacitance demonstrate the feasibility of PDMS dielectrics for capacitive ECG bioelectrodes.**

Keywords—polydimethylsiloxane, composition ratio, dielectric films, dielectric parameters, capacitive ECG bioelectrode, capacitance

I. Introduction

Electrocardiography can reveal the health status of the human heart through bioelectrical signals that are collected using non-invasive electrodes[1]. The wet silver/silver chloride is the ubiquitous clinical electrode with incomparable signal quality for the short-term recording of electrocardiogram (ECG). However, in a long-term application, dehydration of the gel can result in severe signal attenuation and noise interference[2]. Capacitive bipotential electrodes are exciting wearable bioelectronics for long term monitoring of human physiological status. They are capable of non-invasively recording heart bioelectric signals via dielectrics[3]. Metal oxides and fabrics have impressive dielectric properties, but in prolonged use, they induce electrical interference strong enough to corrupt weak biopotential signals such as the ECG[4].

Polydimethylsiloxane, known as PDMS, is a synthetic polymer of silicone that has gained many biomedical applications because of its unique electrical properties[5, 6]. However, no previous study has extensively explored the dielectric capability of pure PDMS for capacitive bioelectrodes. It is of interest to characterized PDMS to examine its dielectric performance for potential applications in wearable heart monitoring devices. High mortality due to heart problems has influenced the demand for a flexible and comfortable ECG electrode for constant monitoring and early detection of cardiac malfunction[7]. Non-contact biopotential electrodes can operate based on capacitive coupling without the need for electrolytic paste or gel[3]. Separate from the sensing element, the dielectric is another vital material that could influence the effective performance of any capacitive ECG bioelectrode[2].

II. Literature Review

Dielectrics are materials with weak ability to conduct electric current. They are insulators which polarised when subjected to an electric field[8]. Materials, whether insulators or conductors, have separate dielectric and electrical characteristics. It is imperative to select an appropriate dielectric material that can generate sufficient electric charges for the sensing conductor[9]. Previously, oxide of metals such as barium[10], tantalum[10, 11], aluminum[12], and silicon[10] have been tested as dielectrics for capacitive ECG sensing. The disadvantages of these dielectric materials in bioelectrodes are their rigid structure, nonconformity with skin, proneness to noise artifacts, and require special shielding against electrical interferences[2, 13].

Alternative materials used as dielectrics are natural and synthetic fabrics[4, 14]. Dielectric fabrics are soft, breathable, porous, and skin conformable. However, in long-term ECG measurements, they exhibit unstable dielectric behavior due to sweat absorption, surface deterioration, and induce frictional noise. A literature review has shown the research conducted on non-contact bioelectrodes reveal that cotton is the most widely used fabric as dielectrics[7]. [4] characterized fabric dielectrics made from cotton, rayon, linen, polyester, nylon, and polyvinyl cloths for the design of capacitive copper-based electrodes to detect bioelectric signals due to muscular movement. Their findings did not include any reason for the low noise recorded and the effect of moisture on the dielectric materials. In another study, [15] made an attempt to optimize the ECG signal quality using a fabric-based electrode. Another concern is the friction between the skin and fabric dielectric can create triboelectric charges strong enough to corrupt ECG signals.

Generally, natural and synthetic polymeric materials have low dielectric constant. In microelectronics, polymers serve as insulators against current leakage and dielectrics between conductors[16]. PDMS belongs to the class of synthetic polymeric silicones that is flexible and harmless to human tissue. It is biocompatible, soft, elastic, and flexible to provide better ergonomic support to the skin[17-19]. Even at giga-hertz test frequency for antennas, its dielectric constant is seldom above 3[20]. Notwithstanding, there is feasibility for dielectric enhancement of silicone PDMS[5, 21]. Also, pure PDMS has not been reported to induce triboelectric charges[22].

In this study, we characterize manually fabricated PDMS films for dielectric applications in capacitive ECG bioelectrodes. In Section I, an introduction to ECG bioelectrodes is given. Section II is a concise review of related studies. Section III describes the materials and method employed while Section IV contains results and discussion. The last stage is Section V which presents the conclusion reached and suggestions.

III. Materials and Method

A. Chemicals and Materials

1.1kg silicone elastomer kit comprising of the Sylgard 184 (Part-A) and curing-agent (Part-B) from Dow Chemical Co, USA, were the main components of the polymer liquid. A transparent multipurpose polymethyl methacrylate (PMMA) acrylic glass films of size 21.0 x 29.7 cm and 3 mm thickness from Cytron Technologies, Malaysia, was used as substrate plates. The transparency of the substrates was to allow physical observation of possible defects that could occur during the arrangement of substrates. A commercial 0.125 mm thick plain PET polyester (mylar) film of 48 x 255 cm size plus an adhesive tape of 0.14 mm thickness were also used as supporting substrate and spacer.

B. Fabrication of Thin PDMS Films

Fifteen samples of thin PDMS dielectric films of 0.14, 0.28, 0.42, 0.56, and 0.60 mm thickness were fabricated by mixing different proportions of the silicone elastomer (Part-A) and the curing agent (Part-B). The manual deposition technique for fabricating the PDMS dielectrics are described in Fig 1. Spin-coating method has been largely used by researchers to fabricate thin PDMS films because it can produce films with homogeneous surface[23, 24]. The disadvantages of spin-coating are that it requires standard laboratory facilities, technical training, and wastes raw materials. The idea of the fabrication technique used in this experiment was primarily established by[25]. This method was found to be simple, low-cost, reproducible, and the needed materials are easily obtainable. To fabricate thin PDMS films of thickness 0.14, 0.28, 0.42, 0.56, and 0.60 mm, a mixture of PDMS Part-A and Part-B in a ratio of 10:1 was manually stirred for 3 minutes at room temperature.

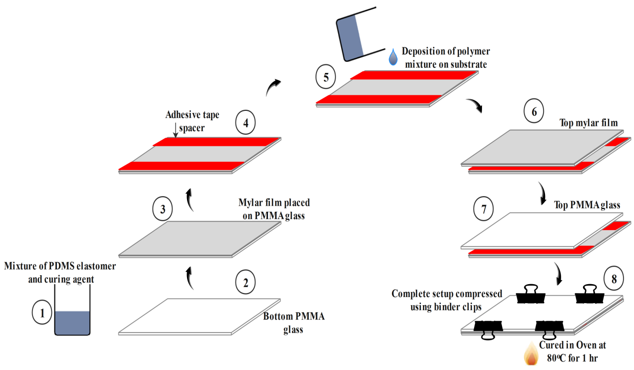


Fig. 1. Schematic procedure for fabrication of thin PDMS using manual deposition technique

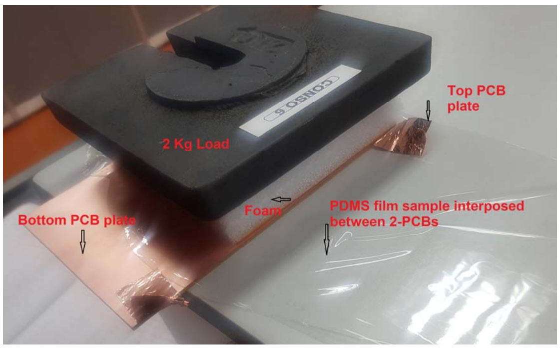
The composition was then degassed to remove any trapped air that was added during the stirring. The mylar film was cut to the size of the PMMA glass and was arranged, as shown in Fig 1. Adhesive tape of thickness 0.14 mm was used as a spacer to set the thickness of the fabricated PDMS film. After that, the mixture was deposited while another set of mylar film and PMMA glass were used to sandwich the polymer liquid, and then compressed tightly using binder clamps. Lastly, the complete setup was placed inside a pre-heated oven at 80 ºC for 60 minutes, as exemplified in Fig 2. The experiment was repeated for another mixture ratio of 10:1.5 and 10:2 for the same thickness.

Fig. 2. Samples of pre-baked PDMS films interposed between PMMA glass plates prior to baking inside oven

A. Electrical and Dielectric Parameters of PDMS Films

The standard technique recommended for the measurement of relative permittivity at low frequency, usually below 1 GHz, is the parallel-plates method. To demonstrate the effect of PDMS composites mix ratio and thickness on the dielectric parameters, each sample of the fabricated PDMS film was inserted between two single-sided printed copper boards (PCB). The bottom PCB had an area of 64.80 cm^2 while the top plate had a size of 43.20 cm^2.

(a)



(b)

Fig. 3. Capacitance measurement by parallel-plate technique (a) top view and (b) front view

A 2 kg load and a high-density foam were then placed on the final setup to provide adequate touching of the PDMS film and PCBs. The instruments were arranged as shown in Fig 3. A commercial LCR meter (UT612) was used to take the capacitance measurements for each sample of the PDMS film of the parallel-arranged copper PCBs. The LCR meter had a precision of ± (0.5% + 5) and default frequencies of 100 Hz, 120 Hz, 1 kHz, 10 kHz, and 100 kHz.

IV. Results and Discussion

The experiment has been performed to examine the behavior of the dielectric parameters of manually fabricated PDMS films. Table I provides a summary of the capacitances measured. To confirm the insulating property of the fabricated films, a digital multimeter was used to measure their resistances. All the samples produced a resistance value >200 MΩ, indicating they are pure insulators.

A. Measured Capacitance by Parallel Plate Method

TABLE I. Values of Capacitance Measured for Different PDMS Mixture Ratio, Thickness, and Test Frequency

Polymer Ratio	Thickness d (mm)	Resistance (MΩ)	Capacitances Measured at Different Test Frequency, C_E (pF)				
			100 Hz	120 Hz	1 kHz	10 kHz	100 kHz
10:1	0.14	> 200	28.00	28.00	25.70	24.02	22.03
	0.28	> 200	27.00	27.00	24.85	23.00	21.76
	0.42	> 200	26.00	26.00	23.89	22.90	20.78
	0.56	> 200	24.00	24.00	22.01	21.00	20.86
	0.60	> 200	24.00	23.00	21.54	20.48	19.85
10:1.5	0.14	> 200	29.00	29.00	27.47	26.10	24.34
	0.28	> 200	28.00	28.00	26.00	24.55	23.78
	0.42	> 200	27.00	27.00	25.07	23.80	22.76
	0.56	> 200	26.00	26.00	24.20	23.00	20.88
	0.60	> 200	26.00	25.00	23.85	22.76	20.48
10.2	0.14	> 200	31.00	31.00	28.90	27.56	25.60
	0.28	> 200	30.00	30.00	27.28	26.12	24.88
	0.42	> 200	29.00	29.00	26.98	25.00	23.71
	0.56	> 200	28.00	28.00	26.04	23.84	22.51
	0.60	> 200	28.00	28.00	25.89	23.52	22.15

The capacitance values measured are provided in Table I and displayed in graphical forms in Fig. 4, Fig. 5, and Fig. 6 for three prepolymer composition ratios and different thickness. The lowest default frequency of the LCR meter (UT612) used for measurement is 100Hz. Changes in the capacitance generated by each film can be seen as the composition ratio and thickness are increased for different test frequency.

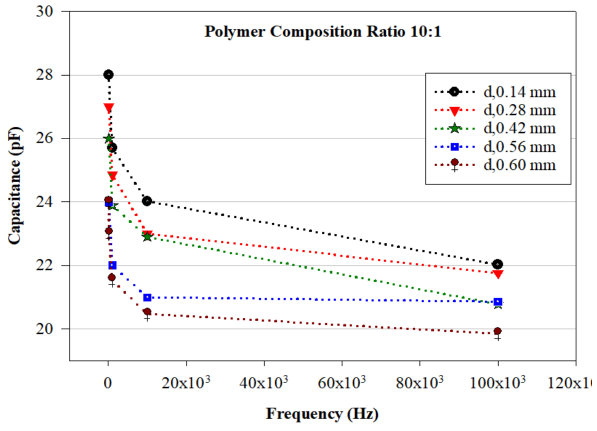


Fig. 4. Effect of thickness and mixture ratio of 10:1 on dielectric capacitance at different test frequency

The highest values of capacitance generated at the frequency were 28pF for a mix ratio of 10:1, 29pF for 10:1.5, and 31pF for a ratio of 10:2. These values were obtained for a dielectric thickness of 0.14mm. The findings of the experiment showed that for a parallel plate of conductors, the capacitance is in inverse proportion to the dielectric thickness. The higher the thickness of the dielectric, the lower the capacitance value. For the three polymer composition ratios and frequencies, the lowest values of capacitance were recorded at a dielectric thickness of 0.60 mm. Surprisingly at the constant thickness, it was observed that the capacitance was decreasing with an increase in frequency. The test frequency of 100 kHz produced the lowest values.

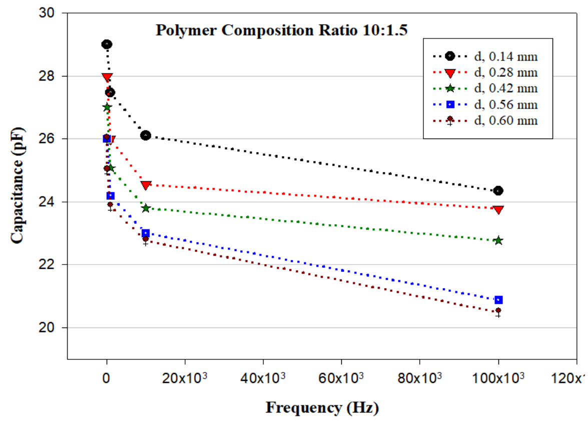


Fig. 4. Effect of thickness and mixture ratio of 10:1.5 on dielectric capacitance at different test frequency

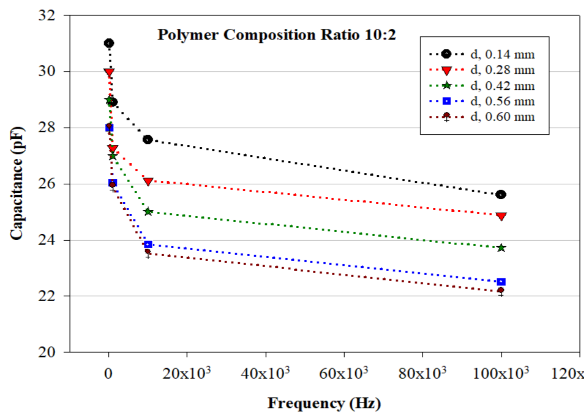


Fig. 5. Effect of thickness and mixture ratio of 10:2 on dielectric capacitance at different test frequency

For instance, at a thickness of 0.42 mm, the capacitance value dropped from 26 pF to 20.78 pF for the mixture ratio of 10:1, 27 pF to 22.76 pF at 10:1.5, and 29 pF to 23.71 pF at 10:2.

Theoretically, for any two parallel plate conductors, their capacitance can be described as,

$$C = \varepsilon_o \varepsilon_r A/d \qquad (1)$$

C_E is capacitance measured in (pF); the relative permittivity of the PDMS dielectric material is ε_r; ε_o is the permittivity of free-space in (F/m); A is the size of the upper conductor plate in (cm^2). Assuming other parameters at the denominators are constants, then the capacitance would largely depend on the dielectric thickness and test frequency. The same (1) was modified to estimate the values of the relative permittivity of PDMS film samples, as given in Table II.

In Fig 4, there was a significant decrease in capacitance from the test frequency of 1 kHz to 100 kHz. Likewise, the drop in capacitances was observed graphically in Fig 5 and Fig. 6 when the mix ratio was increased 0.5. The highest values of capacitance measured were 28 pF, 29 pF, and 31 pF at a dielectric thickness of 0.14 mm, while the least capacitances recorded were 19.85 pF, 20.48 pF, and 22.15 pF at 0.60 mm. Interestingly, the polymer mix ratio of 10:2 yielded the highest capacitance out of the three polymer compositions. Therefore, this result validates that the proportion of crosslinker in the prepolymer has an effective impact on the dielectric parameters of the PDMS films.

TABLE II. TESTIMATED VALUES OF RELATIVE PERMITTIVITY AT DIFFERENT TEST FREQUENCY

Polymer Ratio	Thickness d (mm)	Estimated Relative Permittivity at Different Test Frequency, ε_r				
		100 Hz	120 Hz	1 kHz	10 kHz	100 kHz
10:1	0.14	0.102	0.102	0.094	0.088	0.081
	0.28	0.198	0.198	0.182	0.168	0.159
	0.42	0.285	0.285	0.262	0.251	0.228
	0.56	0.351	0.351	0.322	0.307	0.305
	0.60	0.376	0.361	0.338	0.321	0.311
10:1.5	0.14	0.106	0.106	0.101	0.096	0.089
	0.28	0.205	0.205	0.190	0.180	0.174
	0.42	0.296	0.296	0.275	0.261	0.250
	0.56	0.381	0.381	0.354	0.337	0.306
	0.60	0.408	0.392	0.374	0.357	0.321
10:2	0.14	0.113	0.113	0.106	0.101	0.094
	0.28	0.220	0.220	0.200	0.191	0.182
	0.42	0.318	0.318	0.296	0.275	0.260
	0.56	0.410	0.410	0.381	0.349	0.330
	0.60	0.439	0.439	0.406	0.369	0.347

Table II presents the calculated relative permittivity for each PDMS film for the frequency range of 100 Hz to 100 kHz. The lowest values of relative permittivity attained were 0.102 at a thickness of 0.14 mm and 0.376 for 0.60 mm for a constant frequency of 100 Hz. For a maximum frequency of 100 kHz, the relative permittivity calculated progressively improved from 0.081 to 0.311, 0.089 to 0.321, and 0.094 to 0.347. A likely reason could be an increase in the ratio of the crosslinker alters the chemical structure of PDMS, thereby making it more polarizable, hard, but less elastic. The optimum relative permittivity achieved was 0.439 for 0.60 mm thickness at low frequencies of 100 Hz and 120 Hz.

TABLE III. COMPARISON OF DIELECTRIC PARAMTERS OF DIFFERENT MATERIALS

Material	Thickness (mm)	Capacitance at 1 kHz (pF)	Relative Permittivity, ε_r	Ref
Cotton	0.23	101.19	3.004	[4]
Rayon	0.58	67.88	3.118	
PVC-Textile	0.24	36.09	5.082	
Polyester	0.16	57.04	1.178	
Nylon	0.48	19.72	1.222	
Linen	0.40	77.61	4.007	
PDMS	0.14	31 pF	0.439[a]	Current study

a. Maximum relative permittivity for PDMS was obtained at 0.60 mm dielectric thickness

While the results of this study prove the initial assertion that dielectric parameters of PDMS can be altered by tuning essential factors such as thickness and mix ratio, the highest capacitance and relative permittivity generated are comparably lower than that of cotton and other synthetic fabric dielectric materials as recorded in Table III. Likely reasons for this difference are that the dielectric properties of fabrics are affected by chemical structure and permeability. Though the thickness, material size, and test frequencies used in both scenarios were not the same.

V. CONCLUSION

In summary, for the first time, the impact of polymer ratio, thickness, and frequency on dielectric parameters on PDMS films was evaluated. The results obtained from this experimental study research proves that thickness, polymer composition ratio, and test frequency have noticeable effects on the dielectric parameters of manually fabricated PDMS films. An increase in the proportion of the silicone elastomer curing agent causes the relative permittivity of PDMS dielectrics to rise. The optimum value achieved was for a polymer component mix ratio of 10:2. In contrast, the maximum value of the relative permittivity recorded was 0.439 at a maximum thickness of 0.60 mm, and 4.48 pF was the electrode-skin capacitance obtained at the minimum dielectric thickness of 0.14 mm. This finding suggests that the active electrode area should be made large enough to balance the low capacitance of the PDMS dielectrics. Two possible ways are to reduce the thickness of the dielectric and integrate a sensor element of high conductivity. The proposed PDMS films have dielectric parameters to support capacitive bipotential measurement applications effectively.

ACKNOWLEDGMENT

The first author gratefully acknowledged the support of the Tertiary Education Trust Fund (TETFund) of Nigeria for scholarship award and intervention. Also, the authors express the appreciations to Universiti Teknologi Malaysia (UTM) for providing access to research facilities during the COVID-19 movement control order (MCO).

REFERENCES

[1] K. K. Reddy, K. Abilash, S. Sridevi, M. Pappa, and A. Sateesh, "Development of Non Invasive cardiovascular risk diagnosing system," in *2020 4th International Conference on Trends in Electronics and Informatics (ICOEI)(48184)*, 2020, pp. 961-967.

[2] A. Searle and L. Kirkup, "A direct comparison of wet, dry and insulating bioelectric recording electrodes," (in English), *Physiological Measurement,* Article vol. 21, no. 2, pp. 271-283, May 2000.

[3] A. J. Portelli and S. J. Nasuto, "Design and Development of Non-Contact Bio-Potential Electrodes for Pervasive Health Monitoring Applications," *Biosensors (Basel),* vol. 7, no. 1, Jan 1 2017.

[4] C. L. Ng and M. B. Reaz, "Characterization of Textile-Insulated Capacitive Biosensors," *Sensors,* vol. 17, no. 3, Mar 2017, Art. no. 574.

[5] A. A. Chlaihawi, B. B. Narakathu, S. Emamian, B. J. Bazuin, and M. Z. Atashbar, "Development of printed and flexible dry ECG electrodes," *Sensing and Bio-Sensing Research,* vol. 20, pp. 9-15, 2018.

[6] K. Raj M and S. Chakraborty, "PDMS microfluidics: A mini review," *Journal of Applied Polymer Science,* vol. 137, no. 27, p. 48958, 2020.

[7] S. Majumder, L. Chen, O. Marinov, C. Chen, T. Mondal, and M. J. Deen, "Noncontact Wearable Wireless ECG Systems for Long-Term Monitoring," *IEEE Reviews in Biomedical Engineering,* vol. 11, pp. 306-321, 2018.

[8] R. N. P. Choudhary and S. K. Patri, *Dielectric materials: introduction, research and applications*. Nova Science Publishers, 2009.

[9] K. Vlach, J. Kijonka, F. Jurek, P. Vavra, and P. Zonca, "Capacitive biopotential electrode with a ceramic dielectric layer," (in English), *Sensors and Actuators B-Chemical,* Article vol. 245, pp. 988-995, Jun 2017.

[10] R. M. David and W. M. Portnoy, "Insulated electrocardiogram electrodes," *Medical & Biological Engineering,* Article vol. 10, no. 6, pp. 742-751, 1972.

[11] C. H. Lagow, K. J. Sladek, and P. C. Richardson, "Anodic Insulated Tantalum Oxide Electrocardiograph Electrodes," *IEEE Transactions on Biomedical Engineering,* vol. BME-18, no. 2, pp. 162-164, 1971.

[12] A. Lopez Jr and P. C. Richardson, "Capacitive electrocardiographic and bioelectric electrodes," *IEEE Transactions on Biomedical Engineering,* Article vol. 16, no. 1, p. 99, 1969.

[13] C. Chen, W. Chang, and T. Y. Xie, "Shielded capacitive electrode with high noise immunity," in *2017 IEEE International Conference on Consumer Electronics - Taiwan (ICCE-TW)*, 2017, pp. 157-158.

[14] B. Babusiak, S. Borik, and L. Balogova, "Textile electrodes in capacitive signal sensing applications," *Measurement,* vol. 114, pp. 69-77, 2018.

[15] W. Wu, S. Pirbhulal, A. K. Sangaiah, S. C. Mukhopadhyay, and G. Li, "Optimization of signal quality over comfortability of textile electrodes for ECG monitoring in fog computing based medical applications," *Future generation computer systems,* vol. 86, pp. 515-526, 2018.

[16] G. Maier, "Low dielectric constant polymers for microelectronics," *Progress in Polymer Science,* vol. 26, no. 1, pp. 3-65, 2001.

[17] X. Wang, Z. Liu, and T. Zhang, "Flexible Sensing Electronics for Wearable/Attachable Health Monitoring," *Small,* vol. 13, no. 25, Jul 2017.

[18] F. Stauffer *et al.*, "Skin Conformal Polymer Electrodes for Clinical ECG and EEG Recordings," *Adv Healthc Mater,* vol. 7, no. 7, p. e1700994, Apr 2018.

[19] K. Zheng, S. Chen, L. Zhu, J. Zhao, and X. Guo, "Large Area Solution Processed Poly (Dimethylsiloxane)-Based Thin Film Sensor Patch for Wearable Electrocardiogram Detection," *IEEE Electron Device Letters,* vol. 39, no. 3, pp. 424-427, 2018.

[20] N. J. Farcich, J. Salonen, and P. M. Asbeck, "Single-Length Method Used to Determine the Dielectric Constant of Polydimethylsiloxane," *IEEE Transactions on Microwave Theory and Techniques,* vol. 56, no. 12, pp. 2963-2971, 2008.

[21] L. Wu, T. Xu, G. Li, X. Zhu, and J. Zhao, "A facile alignment of PDMS layers for the fabrication of 3D microfluidic devices using synergistic zipper structure," in *2018 IEEE Micro Electro Mechanical Systems (MEMS)*, 2018, pp. 1265-1268.

[22] T. Wartzek, T. Lammersen, B. Eilebrecht, M. Walter, and S. Leonhardt, "Triboelectricity in capacitive biopotential measurements," *IEEE Trans Biomed Eng,* Article vol. 58, no. 5, pp. 1268-77, May 2011, Art. no. 5672585.

[23] D. Wang *et al.*, "A novel approach for PDMS thin films production towards application as substrate for flexible biosensors," *Materials Letters,* vol. 221, pp. 228-231, 2018.

[24] R. K. Pal, S. Pradhan, L. Narayanan, and V. K. Yadavalli, "Micropatterned conductive polymer biosensors on flexible PDMS films," *Sensors and Actuators B: Chemical,* vol. 259, pp. 498-504, 2018.

[25] R. Martinez-Duarte, "Easy and inexpensive fabrication of PDMS films of different thicknesses « Chips and Tips," ed: Royal Chemical Society, 2012.