ANALYSIS AND CHARACTERIZATION OF POLYDIMETHYLSILOXANE (PDMS) SUBSTRATE BY USING UNIAXIAL TENSILE TEST AND MOONEY-RIVLIN HYPERELASTIC MODEL

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ABSTRACT: Polydimethylsiloxane (PDMS) is the most widely used silicone polymer in the development of microfluidic devices, sensors, medical appliances and stretchable electronic devices. PDMS Sylgard 184 from Dow Corning is obtained by mixing only base polymer and cross-linking agent in a certain ratio which is more expensive than other types of PDMS and difficult to cure when reacting with rubber like materials and plastics. Also, the tensile testing of such type of substrate is impossible to carry out at higher cross linking ratio (30:1) and above. To overcome these issues, this study proposed a new formulation to fabricate PDMS substrate by mixing base polymer PDMS-OH (PDMS hydroxy terminated) with different chemicals that act as cross-linking agent, organic solvent, viscosity controller and catalyst. The mixing is done at 200-250 rpm using magnetic or mechanical stirrer. The prepared formulation is then poured into three dog bone shaped molds (based on ASTM D412 Type C standard) and allowed to cure at room temperature for 24 hrs. The tensile strength is then characterized by uniaxial tensile test on UTM machine. Mooney-Rivlin (5 and 9 parameters) model is implemented on engineering stress strain data in ANSYS to validate the obtained strain rate and material constants associated with this hyperelastic model are obtained by curve fitting damped least squared method. Also, the maximum stress obtained by UTM testing is 2.18 MPa before failure at 300% strain rate. The MR models are validated by calculating coefficient of determination R^2 values. The R^2 values of MR5 and MR9 parameters models are 0.9914 and 0.9994, respectively.

KEYWORDS: *Uniaxial Tensile Test; Characterization of Material; PDMS Substrate; Hyperelastic Models*

1.0 INTRODUCTION

The most widely used polymeric organic compounds as substrate in many stretchable electronic systems is PDMS or dimethicone. PDMS is composed of inorganic chain of silicon and oxygen atoms. The chemical formulation with two methyl groups attach to silicon atoms is $SiO\ (CH_3)_2$. In early ages, PDMS worked as an insulator that has low tension and high voltage but nowadays, it has been used in the making of several microsystems [1], actuators [2], stretchable and flexible electronic systems [3], contact lenses, medical appliances, sanitary items, lubricants and heat resistant tiles. It is also biocompatible in nature that has low glass transition temperature and becomes more elastic if it combines with cross linked agent. PDMS also exhibits excellent gas permeability and due to the lack of toxicity and good biocompatibility, it is widely used in manufacturing of pharmaceutical and medical appliances. As compared to other elastomers, like silicon and rubber, PDMS is more thermally stable, chemically inert, easy to handle, isotropic structure, homogenous properties and less expensive. The different forms of PDMS are available in the market such as Sylgard 184, Dow Corning, PDMS-SiH (polydimethylsiloxane silica hydrate) and PDMS-OH. The most common type of PDMS is Sylgard 184.

There are two states of PDMS surface: hydrophobic and hydrophilic. PDMS acts as a hydrophobic elastomer after reacting with cross-linking agent. Due to this behavior, not a single drop of polar solvent like water can easily spread on the surface of PDMS film. Wettability of PDMS substrate is controlled by different surface treatments like plasma discharging [4], passing ultra violet rays, ozone and corona rays, thermal aging [5], applying different chemicals on its surface. The duration of exposures of UV, ozone and corona rays depend on the surface activation of PDMS. Large exposures more than 1 min oxidizes the PDMS surface in the presence of oxygen and helps to make the surface brittle and break after mechanical deformation [6], while small duration of exposures make the surface ductile, therefore, PDMS surface activation is achieved by short exposures of UV, ozone and corona rays. The polar functional groups (SiOH) are also introduced by plasma oxidation treatment on the surface of PDMS which help in changing the state of PDMS surface from hydrophobic to hydrophilic state. Since, hydrophilic state is unstable, therefore, wettability increased and low molecular weight chains (LMW) spread out over large area of PDMS surface [4]. This state can also be changed during thermal aging which helps to volatize and remove LMW particles from the PDMS surface [5].

The mechanical properties of PDMS have been previously studied by many scientists mainly focused on particular applications such as thin membranes used in sensors [7], elasticity of material used in accelerometers [8], non-linear behavior of PDMS in standard and modified formulations [9]. The common ratio for PDMS Sylgard 184 is 10 parts base polymer and 1 part cross-linking agent (10:1). Generally, PDMS substrate can be cured at normal room temperature such as ≤ 25^0C and possesses 1.3 MPa Young's modulus while on the other hand, PDMS film cured above 25^0C and less than 200^0C then modulus of elasticity will be 2.97 MPa [10]. Also, the stiffer PDMS has elastic modulus below 5 MPa, while the elastic modulus for soft PDMS is below 1 MPa, observed during tensile test [11]. Moreover, Someya [12] provided the range of elastic modulus (0.1-3.5 MPa) of PDMS by altering curing temperature, ratio of base polymer and cross-linking agent, loading and thermal conditions. Hence, the softness of PDMS substrate depends on low curing temperature and increased ratio of PDMS base polymer and cross linked agent (10:1, 15:1 or 20:1) [13].

PDMS is also chemically inert silicon rubber that has low polarity. Several aqueous solvents cannot react with PDMS except organic solvents. The reaction of PDMS with organic solvents shows that PDMS starts swelling as in photolithography procedure [14]. In last few decades, a lot of work has been done in the measurement of mechanical behavior of common type of PDMS that is PDMS Sylgard 184. Different ratios of base polymer and cross linking agent are taken into account. The mechanical properties of PDMS Sylgard 184 from Dow Corning are measured by mixing different ratios of base polymer and curing agent [15]. Also, Kim et al. [9] studied the non-linear mechanical behavior of Sylgard 184 by using 3 different ratios of polymer and curing agent. In recent times, Schneider et al. [16] observed the mechanical properties of Sylgard 184 (ratio 10:1) by adding different concentrations of thinner into Sylgard 184 against temperature and also calculated modulus of elasticity against strain rate without adding thinner. Vaicekauskaite et al. [17] mapped the mechanical and electrical properties of Sylgard 184, Sylgard 186, Ecoflex 00-10, Ecoflex 00-30 and Ecoflex 00-50 by blending them into certain proportions and fabricated stretchable transducers. Also, Glover et al. [18] investigated the effect of uncrosslinked material from Sylgard 184 of low modulus of elasticity on mechanical properties. Despite of these properties of PDMS Sylgard 184, it also possesses some disadvantages like it is very expensive and in some cases, this type of PDMS cannot be able to fully cure when in contact with any plastic or rubber like structure. Therefore, in order to reduce this problem, other chemicals such as cross-linking agents, organic solvents and catalysts are added to the base polymer PDMS-OH.

In point of fact, PDMS is one of the hyperelastic materials which is ideally incompressible and does not obey simple Hooke's law. To characterize and evaluate the mechanical properties of such type of materials, different hyperelastic models have been used by many researchers on the basis of their deformation in stress-strain analysis. All these models are generally handled by ABAQUS and FEA software. The most popular non-linear hyperelastic models are Neo-Hookean (NH), Mooney-Rivlin (MR), Yeoh and Ogden models [19]. Kim et al. [19] compared NH, MR and Ogden models with each other and concluded that NH and MR models are the simplest and easiest one to apply than other models but they do not provide good results for large deformations. Yeoh and Ogden models are suitable for large deformations. Similarly, NH and MR models are applied by Noor et al. [20] in order to calculate the stiffness of hyperelastic material. Xie et al. [21] compared the elastic material model with NH model for intervertebral disc (IVD) experimental data and found that NH model gives better results. Gajewski et al. [22] calculated the mechanical properties of elastomeric bridge bearing with steel reinforcement under compression and shear loading by using ABAQUS. The curve fitting of experimental data obtained by uniaxial tensile test and shear test is done by NH and Yeoh models. As a result, it is concluded that Yeoh model predicts the best mechanical behavior of elastomeric bridge bearing. Rathod et al. [23] used linear and non-linear constitutive models to calculate material properties of PDMS in ABAQUS. NH and Arruda-Boyce models are applied on experimental data and the results show that NH model gives more accurate results than Arruda-Boyce model. Similarly, the stress-strain analysis of silicon elastomer PDMS films are carried out by UTM and the curve fitting of obtained experimental data is done by using NH, MR, Yeoh, Ogden, Arruda-Boyce and Van de Waals models in ABAQUS [24]. As a result, it is found that MR, Ogden 2-terms and Yeoh 3rd order are in a good agreement with experimental data.

The main objective of this study is to propose the new formulation of PDMS substrate using PDMS-OH as a base polymer, cross linking agent and different chemicals in a certain ratio and also examine the mechanical properties by performing uniaxial tensile test on Universal testing machine (UTM-INSTRON 3366). Moreover, Mooney-Rivlin hyperelastic model is used to characterize the material properties. This model is implemented on engineering stress-strain data in ANSYS Workbench software.

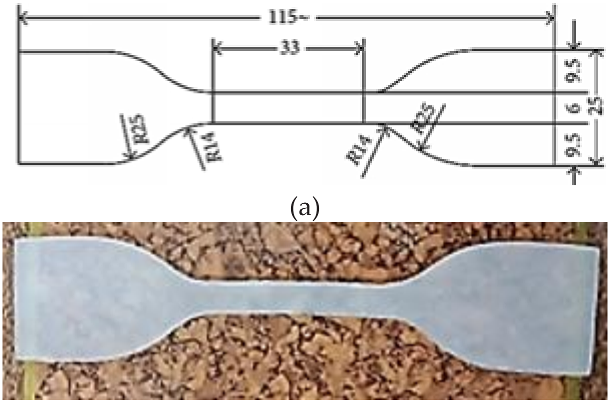
2.0 METHODOLOGY

2.1 Materials

Poly (dimethylsiloxane) hydroxy terminated (PDMS-OH) of 110 x 10^3 g/mol molecular weight and viscosity of 50 x 10^3 cSt, (3-glycidyloxypropyl) trimethoxysilane (ETMS) (236.34 g/mol molecular weight ≥ 98% purity, 1.07 g/ml @ 25^0C specific gravity) performances as cross-linking agent, toluene (92.14 g/mol molecular weight, 99% purity, 0.867 g/ml density) used as a solvent, fume silica (5-50 nm particle size, 2.2 to 2.3 g/ml specific gravity) acts as a viscosity controller and dibutyltin dilaurate (DBDTL) (631.56 g/mol molecular weight, 1.066 g/ml density, 95% purity) functions as catalyst. All these chemicals are purchased from Sigma Aldrich.

2.2 Sample Preparation

PDMS substrate is obtained by mixing PDMS-OH with fume silica and toluene for approximately 20 mins. The mixing is done by mechanical stirrer or magnetic stirrer at 150-200 rpm. After that, ETMS is added and again stirred for 5-10 mins. Finally, DBDTL is poured into that mixture and stirred for approximately 1-2 mins. The final mixture is then poured into a dog bone shaped mold (ASTM D412 Type C standard) [25] and followed by curing for 24 hrs at room temperature. Figure 1 shows the specifications of ASTM D412 Type C mold and final sample, respectively.



(a)

(b)

Figure 1: (a) Specification of ASTM D412 type c mold and (b) final sample

2.3 Uniaxial Tensile Test

Universal testing machine (UTM-INSTRON 3366) is used to analyze the mechanical behavior of PDMS substrate. The test is conducted on three dog bone shaped samples as shown in Figure 1. The samples are gripped at gauge length of 25 mm and 10 mm/min loading rate (cross -head speed) with 10 kN load cell at room temperature is applied. The experimental results used to calculate the engineering stress and strain values.

2.4 Mooney-Rivlin Hyperelastic Material Model

The non-linear stress-strain relation of hyperelastic materials such as rubbers and elastomers are explained by different hyperelastic models. One of the most commonly used hyperelastic models is Mooney-Rivlin (MR) model. In this model, material is considered to be isotropic, incompressible, and come back to its original shape after unloading. Hence, flexibility is also independent on the strain rate. MR model is the extension of Neo-Hookean model that is used to improve the accuracy. It is limited to small strain deformations and uniaxial loading only. There are 2, 3, 5 and 9 parameters of MR models available for the characterization of materials. In this study, MR5 and MR9 parameters models are opted for validation of engineering stress-strain data. The general formation of strain energy and stress of MR model for incompressible materials explains the elastic behavior of rubber like materials are given such as

$$W = \sum_{i,j=0}^{N} C_{ij}(I_1 - 3)^i (I_2 - 3)^j \tag{1}$$

$$\sigma_i = \lambda_i \frac{\partial W}{\partial \lambda_i} \tag{2}$$

where W is Helmholtz free strain energy per unit reference volume, J is the Jacobean determinant or determinant of elastic deformation gradient, C_{ij} is material constant, I_1, I_2, I_3 are invariants ($I_1 = \lambda_1^2 + \lambda_2^2 + \lambda_3^2$; $I_2 = \lambda_1^2\lambda_2^2 + \lambda_2^2\lambda_3^2 + \lambda_3^2\lambda_1^2$; $I_3 = \lambda_1^2\lambda_2^2\lambda_3^2$) and λ is stretch ratio calculated as $\lambda = 1+\varepsilon = Lo/L$.

3.0 RESULTS AND DISCUSSION

The stress-strain engineering curve is obtained by performing uniaxial tensile test on ASTM D412-C PDMS substrate as shown in Figure 2. From the graph, it is observed that the maximum stress achieved by substrate before failure is 2.18 MPa at 300% strain and modulus of elasticity is 0.48 MPa. The formulated PDMS substrate has 33:1 ratio of base polymer and crosslinking agent, while the modulus of elasticity of Sylgard 184 is 0.036 MPa at the same ratio as that of formulated PDMS as illustrated in Figure 3 [12]. It is also observed that formulated PDMS substrate is more stretchable and flexible than Sylgard 184 because when the un-crosslinked polymer increases in Sylgard formation then the modulus of elasticity dramatically decreases for higher ratios such as 30:1 and above. Also, more than 92% increment is observed in modulus of formulated PDMS. Sylgard 184 cannot be handled in tensile testing due to extremely viscous nature, therefore, the formulated PDMS substrate gives good modulus of elasticity and tensile strength at higher cross linking ratios.

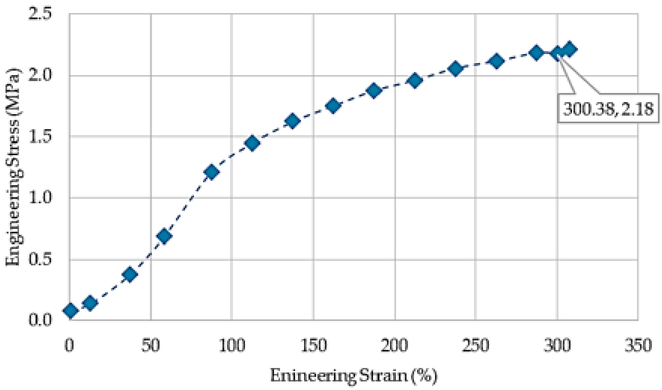


Figure 2: Engineering stress-strain curve

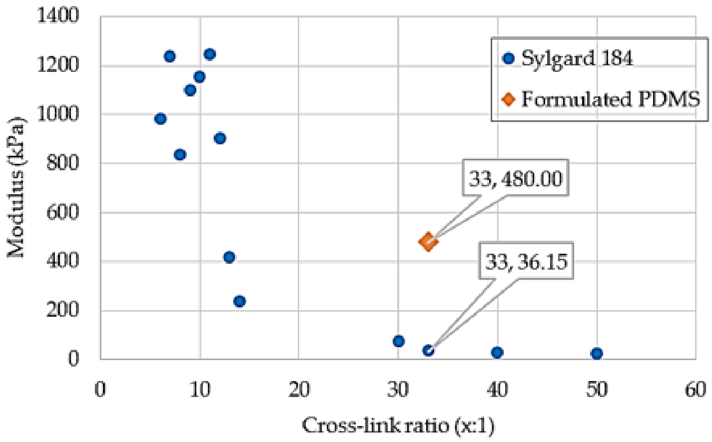


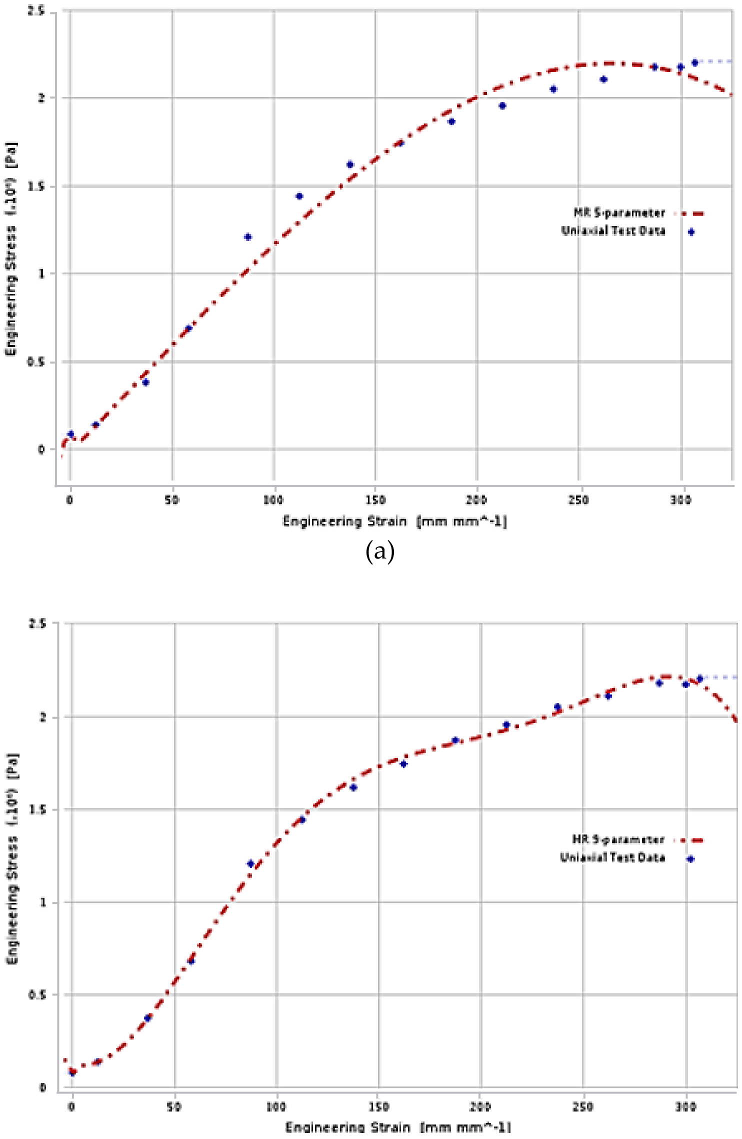
Figure 3: Young's modulus of sylgard 184 related to the cross-link ratios [12]

PDMS substrate is hyperelastic material and shows non-linear behavior in stress-strain graph, MR5 and MR9 parameters models are applied on experimental uniaxial tensile data. These models are estimated by applying Levenberg-Marquardt also known as damped least squared method [26-27]. Figure 4 shows the curve fitting results of experimental data with different hyperelastic models.

Table 1: Material constants and R^2 of curve fitting of MR hyperelastic models

Constant	C_{10}	C_{01}	C_{20}	C_{11}	C_{02}	C_{30}	C_{21}	C_{12}	C_{03}	R^2
5 Parameter	-0.301	0.442	$-1.53x10^{-8}$	$2.09x10^{-7}$	0.077	-	-	-	-	0.9914
9 Parameter	-0.967	1.194	-0.0011	-0.438	0.901	$-2.51x10^{-12}$	$1.26x10^{-9}$	0.0003	0.11	0.9994

Furthermore, the material constants associated with hyperelastic models and the validation of these models are checked by calculating R^2 values on the basis of FEM simulation as shown in Table 1. By increasing number of parameters in MR model, the accuracy of material model increases. MR9 model works on three or more inflection points while MR5 only used for two inflection points. According to Figure 2, the material observed 3 inflection points during tensile test. Also, the R^2 values of MR9 and MR5 models are very close to each other (0.9994 and 0.9914, respectively) but differ on the basis of number of parameters and inflection points, therefore, MR9 parameter model performs better than MR5.



(a)

(b)

Figure 4: Curve fitting results for (a) MR5 and (b) MR9 parameters

4.0 CONCLUSION

This research focuses on the development of PDMS substrate by stirring PDMS-OH elastomer with toluene and fume silica for 20 mins followed by ETMS and DBDTL. The formulated PDMS substrate is more stretchable and flexible than Sylgard 184 at higher ratios. Uniaxial test is carried out to determine the strength of the substrate.

The proposed formulation has a tensile strength of 2.18 MPa before failure at about 300% elongation rate. Its modulus of elasticity is obtained as 0.48 MPa (at curing temperature 25^0C). The elasticity can be increased by adding certain ratio of plasticizer into the formulation and can also increase or decrease the curing temperature. Furthermore, PDMS substrate is characterized by Mooney-Rivlin (MR) (5 and 9 parameters) hyperelastic model. The calculated R2 value of MR9 is maximum that is 0.9994 than MR5. As a result, MR9 model gives more accurate results than MR5 because of higher order parameters and more than 2 inflection points obtained in stress-strain curves.

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