Mechanical Properties of Graded Polydimethylsiloxane for Flexible Electronics

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Abstract. The extensive use of polydimethysiloxane (PDMS) in the context of flexible systems such as flexible display has grown an issue of its flexibility upon high loadings and rupture. In previous study, PDMS substrates failed through the rupture and delaminated completely at 120% at global strain. Graded PDMS samples consisting five different layers of mixing ratios was designed to effectively fulfill the mechanical function. Each layer was differ in color to accentuate the difference of mixing ratios. The first layer had the mixing ratio of base polymer to curing agent 2:1, followed by 5:1, 10:1, 15:1 and 20:1 respectively. The graded PDMS samples were prepared using a simple casting, curing, stacking and cutting process. This study presents the measurement of the mechanical properties of Graded PDMS based on the mixing ratio of base polymer to curing agent. A tensile test and fracture test were performed with a Universal Testing Machine at room temperature. The new finding is that when the graded PDMS were stacked together varying the layers from hardest (2:1) to softest (20:1) PDMS substrates, the range of flexibility is very wide. It can be seen from the stress-strain curves that the graded PDMS sample showed flexibility and yet can elongate up 170% strain before failing. It could be concluded that by designing and improving the graded PDMS combining the hardest (2:1) to the softest (20:1) PDMS substrates can vary its mechanical properties to approach the hardest biological materials such as bone and tooth enamel, whereas still can be reversibly stretchable up to 170% strain without failure. This study will be of use to all the technological communities who are in used of PDMS polymers for their specific investigations and applications in flexible system.

1. Introduction
Flexible electronics, also known as flex circuits, is a technology for assembling electronic circuits by mounting electronic devices on flexible plastic substrates, such as polyimide, PDMS or transparent conductive polyester film. Flexible electronic assemblies may be manufactured using identical components used for rigid printed circuit boards, allowing the board to conform to a desired shape, or to flex during its use [1].
Flexible electronics such as flexible displays, wearable electronics, electronic textile, sensors skins, and thin-film transistors (TFTs), are among the few applications used in thin substrates. Substrate thickness can be used for such devices as it can be as low as 25 μm while traditional microelectronic devices are manufactured on thick, hard substrates [2].
New generation nowadays are demanding modern technologies especially in telecommunications such as mobile phones and computers. They want a technology that is thin, light and preferably foldable but at the same time not reducing any of its best qualities. In this matter of developing the new demanding technologies, the flexibility of flexible electronics needs to be improved.
The design of flexible electronics involves the development of specific architectures that would combine high global stretchability with minimum local strains. Using graded Polydimethylsiloxane (PDMS), the flexibility of this electronics can be expended. The creation of 3D polymer-based composites can approach hard materials such as bone and tooth enamel, but at the same time allow the structures to be reversible and stretchable [3].

2. Fabrication Of Graded PDMS
The materials used for this study includes base and curing agents purchased from Sylgard 184 Silicone Elastomer Kit, product by Dow Corning, USA. Vinyl-terminated base dimethylpolysiloxanes are available in various viscosities. It is a base polymer for addition curing and can be used as a base polymer in order to create the desired hardness.

The investigation was on a large range of cross linker agent concentrations, mixing ratios by weight ranging from 20:1 to 2:1. Graded PDMS mixtures were first prepared by mixing the base and the curing agent at the specific mixing ratios by weight. Both base and curing agents were weighed by using Mettler Toledo Weighing Balances from Mettler Toledo, USA. The mixing ratios studied here consisted of 20:1, 15:1, 10:1, 5:1 and 2:1. The different layers of the different mixing ratios were differentiated by colors. Mixtures were then carefully poured into the dishes using a specific volume of the mixture to have the desired thickness of each layer.

The preparation involved mixing two liquids of different viscosities which incurred the formation of trapped bubbles. Gases trapped needed to be removed in order to avoid air or oxygen sensitive and bubble formation at the interfaces that might become a problem. The formation of gas bubbles was undesirable, necessitating degassing. This process is called degasification. Thus, the samples were finally degassed in a low-vacuum chamber in order to remove the air bubbles formed during mixing. Then the next layer was stacked on to the previous layer. The readily prepared graded PDMS sample was shown in Figure 1.

For elasticity test, 24x3 mm tensile rectangular shaped samples were produced from the graded PDMS using sample cutter. Figure-2 and Figure-3 showed the schematic drawing of samples for tensile test and fracture test respectively. In the present study, 5 mm/min speed was used and the distance of gauge length was 3.00 mm. The break load drops was 20%. The specimens were going to be examined and the average results were reported. The Nexygen software that is applicable on the Universal Tensile Machine (UTM) showed the results in mechanical properties such as maximum load, deflection at max load, stiffness and also stress and strain.

Meanwhile for fracture test, 24×3 mm rectangular shaped were produced using the same process as elasticity samples but with notch of 1/3 of overall length. In the present study, and also the same process done in tensile test, 5 mm/min speed was used and the distance of gauge length is 3mm. The break load drops is 20%. Three samples were examined and their average results were reported.

3. Experimental Results And Discussion

The mechanical performance tests were conducted after the fabrication of the samples were done. In the current study, the mechanical performance tests that had been conducted were tensile and fracture test on the graded PDMS samples.

3.1. Tensile Strength

The effect of stacking different layers of different ratios of base and curing agent in the stress-strain behavior was observed where the y axis was the exercised engineering stress and strain, \( \varepsilon = \frac{\Delta L}{L_0} \) where \( L_0 \) was the initial sample length and \( \Delta L \) is the elongation. The graded PDMS samples were quite uniform and showed typical behavior of stress strain curve. The mean values of tensile strength was 275 kPa together with the values of Young’s Modulus (E) of 28.4 kPa, calculated from the slope of the initially linear part of the curves for stress-strain region below 40%.
The average of Young’s Modulus value was 28.4 kPa neglecting the graded PDMS sample 2. Graded PDMS sample 2 was neglected due to an error where the slip phenomenon happened caused by the UTM machine while testing causing premature failure of the graded Sample 2 upon stretching. Graded PDMS sample 1 and 3 showed very similar behavior where the value of Young’s Modulus was quite similar indicating that the test is successive.

The first deal was to analyze the resulting stress-strain curves. Figure 2 presented experimental stress-strain curves obtained on the graded PDMS samples. Loading strains and stresses reached 170% and 0.5 MPa respectively before failing. The stress-strain behavior was observed to be rate dependent and highly non-linear. The curves showed an initially relative stiff behavior and followed by a rollover to a more compliant behavior when it is at low strains. The stress level were seen to ranging from 0 MPa to nearly 0.5 MPa depending on the magnitude of strain. As expected and stated earlier, the low and high rate data were fully consistent and display typical linear elastic region up to strain values of 40% (see Figure 3) followed by a nonlinear region before failure.

![Stress-Strain curves of graded PDMS samples.](image1)

Figure 2. Stress-Strain curves of graded PDMS samples.

![Detail of averaged tensile stress-strain curves up to 40% strain for graded PDMS samples.](image2)

Figure 3. Detail of averaged tensile stress-strain curves up to 40% strain for graded PDMS samples.
It was observed that a very large range of behavior from elastic-linear behavior, to a hyper elastic and even brittle on the graded PDMS samples. A systematic sample hardening was observed from the graded PDMS samples. This was caused by a phenomenon where a systematic increase of low strain and high strain stiffness.

It was observed that the graded PDMS sample stiffness was quite lower to be compared to the largest observed tensile stiffness, using 10:1 PDMS to date which is less than 4 MPa [4]. However, the loading strain was higher than 170% that indicates an improvement than nongraded polyurethane (PU) substrates where it failed at strains > 120% [3].

The comparison was made between the graded PDMS sample and non-graded PU sample with an out-of-plane gradient in elastic modulus spanning over five orders of magnitude of different volume fractions from other study. The previous study was to determine the unique set of mechanical properties by tuning the local reinforcement level of a thermoplastic elastomer in 3D. The comparison was made with the volume fraction of M0040 undergo tensile strain.

The non-graded M0040 PU module consisted of a prismatic patch with designed $E_l$ profile deposited onto a ribbon of stretchable substrate with 50:50 weight ratio mixture of hard and soft monomers in N,N- Dimethylformamide (DMF). The non-graded PU failed and delaminates completely at 120% strain upon stretching the whole construct [3].

The acknowledgement is of graded PDMS being quite brittle with low value of Young’s Modulus, however can be elongated up to 170% strain, 50% higher than non-graded PU. The achievement can be further studied in order to obtain a graded PDMS that can approach the hardest biological materials such as bone and tooth enamel but still can be reversible stretch up to 350% strain without failure.

To complete the analysis of material stiffness and rupture, Figure 4 presented the evolution of graded PDMS rupture as a function of stiffness. It showed a particularly interesting for applications where both high rigidity and good flexibility were required since they clearly constraining the limits of normal PDMS mixtures.

![Rupture Strain (%) vs Tangent Modulus (MPa)](image)

**Figure 4.** Variation of rupture strain of graded PDMS samples function of PDMS stiffness.

3.2. Fracture Toughness

The next detailed discussion is fracture energy. The fracture energy were determined from stress-strain area under graph. The accuracy of this method was verified by taking video footage of the crack propagation for selected samples.

The properties of the graded PDMS samples were listed in Table 1 below where $W_o$ is area under graph determine from the stress-strain curve of fracture test, $l_0$ is initial length of sample and $G_c$ is fracture energy.
Table 1. Properties of fracture energy of graded PDMS sample.

| Sample | $W_0$  | $I_0$  | $G_c$  |
|--------|--------|--------|--------|
| 1      | 11.0504| 3.06   | 38.8142|
| 2      | 15.9076| 3.16   | 50.2680|
| 3      | 9.7923 | 3.00   | 29.3770|
| Average| 8.2501 | 3.0733 | 25.1798|
| Standard Deviation | 3.8130 | 0.0808 | 11.3319 |

Stress-strain curves for fracture test was displayed in Figure 5. The graded PDMS samples showed an upturn in stress at high strain. Each of the graded PDMS samples broke at maximum stress values, however graded PDMS Sample 2 extended to higher elongation ratio before rupture as it had the highest maximum stress value before breaking. Graded PDMS Sample 1 only showed moderate increase in toughness, however the toughness values for each series began to diverge in the more ductile networks.

In simple extension, non-graded PDMS sample usually reached higher elongation ratio before fracture with similar values of elastic modulus. These results reiterated that enhanced mechanical properties in graded PDMS sample were manifested at high strains.

When a pre-cut sample is prepared, network fracture begins at a lower elongation ratio as the stress is concentrated at the crack tip [5]. While it is difficult to determine on the sharpness or bluntness of natural imperfections, enclosed flaws are considered less serious stress raisers than edge cuts [6]. Regarding the pre-cut samples in this tests reported to be always utilize edge cuts and the crack tip was expected to be sharp, it was not most graded PDMS samples except for graded Sample 2 that had slightly lower values. It was suspected that the shape of the crack tip as the crack grow play an important role in these results.

Graded PDMS sample showed outstanding mechanical properties in simple extension. It was extended to a large elongation ratio before fracture and had large stress at high elongation resulting in higher toughness value with similar elastic modulus. The graded PDMS was not quite tough and there was no other results from other studies to be compared with as there was no investigation yet on fracture...
toughness of graded PDMS. Each pre-cut sample was extended at a constant crosshead speed until the crack had propagated through its entire width. The elongation ratio at which the crack began to grow in these samples was determined and verified by taking video footage of the crack propagation. It can be seen that the crack propagated uniformly through its entire width until breaking. The stress was concentrated at the crack tip resulting in fracture begins at much lower elongation ratio.

4. Conclusion
The presented experimental data described the variation in material properties of graded PDMS with its number of layers consisting different mixing ratio. This demonstrated that by modifying the cross-linker agent concentration (mixing ratio), one can tune the stiffness of 2.5 mm thick graded PDMS samples varying each ratio of 2:1, 5:1, 10:1, 15:1 and 20:1 respectively. No problems associated with moulding had been observed. Indeed from the results, it was speculated that the graded PDMS ranging from the brittle behavior in the stiffest material (2:1) to the hyper elastic behaviour surprising that the value of elongation ratio of these samples differed from each other. The resulting $G_c$ values were quite similar for the material (20:1) enabled the user to explore the stiffness range from 26.1 kPa to 30 kPa and more loading strain reaching up to 170%.

The relationship between rupture strain and material stiffness had been systematically and statistically investigated. The results indicated that the rupture strain of the Graded PDMS evolved as the square of the material stiffness. Graded PDMS was extended to a large elongation ratio before fracture and had a high upturn in stress at elongation, resulting in toughness value that were quite high.

It was believed that by increasing the range of mechanical properties of graded PDMS, a material with more compatibility with technological processes was rendered. This method could potentially be explored to produce flexible electronics with extreme mechanical gradients and tougher graded adhesives that would reduce huge failure of current fiber-reinforced composites.

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