

ORIGINAL ARTICLE

Factors affecting the surface and release properties of thin polydimethylsiloxane films

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Polydimethylsiloxane (PDMS) elastomers are commonly used as dielectric electroactive polymers (DEAPs). DEAP films are used in making actuators, generators and sensors. In the large-scale manufacture of DEAP films, the release of films from the substrate (carrier web) induces some defects and prestrain in the films, which affects the overall performance of the films. The current research is directed toward investigating factors affecting the peel force and release of thin, corrugated PDMS films used as DEAP films. It has been shown that doping the PDMS films with small quantities of perfluoroether allylamide ($F(CF_2CF_2O)_7CF_2CONHCH_2CH=CH_2$) lowers the surface energy, which could facilitate release. This idea is further investigated, and the resultant change in the film performance is evaluated. The relationship between the adhesive energy, surface energy, Young's modulus and peel force of the films is also analyzed.

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INTRODUCTION

In the large-scale manufacture of dielectric electroactive polymer (DEAP) films, the polydimethylsiloxane (PDMS) elastomer mixture is applied to the substrate (carrier web) and then peeled off the substrate after the elastomer has cured completely.¹ The process of release from the substrate is not as smooth as desired, and the process induces considerable prestrain and defects in the film, which affects the performance of the films as actuators. To ease the process of release, release agents in the form of sprays or liquids cannot always be used. Modifying or replacing the web for the processing of the microstructured films is not possible at present because of the commercial unavailability of such products in the large quantities required for the continuous process of producing microcorrugated DEAP films.

The surface energy of PDMS is 19–21 mJ m⁻² and contact angle with water is 110° (Supplementary Figure S1), which explains its inert nature toward many chemical species and its poor adhesion to many substrates. Through the addition of suitable surface-active block copolymers, it has been shown that the surface energy can be decreased further or that the film can be made selectively nonadhesive to a particular substrate.³ A polymeric additive with a lower surface energy than its host matrix is known to adsorb preferentially at the free surface and consequently decrease the adhesion of that surface toward a particular substrate.³ The substrate on which the PDMS films are made is a polyethylene terephthalate coated with microscale corrugations of methyl acrylate. This is the substrate used in the large-scale industrial manufacture of DEAP films.¹

Surface tension, peeling and release

The molecules on a free surface of the film will have lesser binding energy than those molecules in the bulk. This missing (negative) binding energy can be viewed as a positive energy added to the free surface. External forces must perform positive work against internal surface forces to increase the area of a surface. These internal surface forces are called surface tension, which is defined as the normal force per unit area.⁴ Surface tension is present at all surfaces and interfaces. The surface energy density associated with an interface between a solid or liquid and a gas is always positive because of the missing negative binding energy of surface molecules. Interfaces between solids and liquids or between solids and solids are not required to have positive interfacial energy density. The sign depends on the strength of the cohesive forces holding molecules of a material together compared with the strength of the adhesive forces between the opposing molecules of the interfacing materials. If the interfacial energy between two liquids is negative, then a large amount of energy can be released by mixing them, and hence, the liquids get mixed instead of staying separate. Immiscible liquids such as oil and water have a positive interfacial energy density, which makes them seek a minimum interfacial area.⁴

The peel force required to peel a film from a substrate is a complex function of geometry, the mechanical properties of the film and the substrate, the thickness of the film, the interfacial cohesive properties and the friction between the surfaces. A detailed study and analysis of the peel test and the factors that govern the peel force has been presented by Thouless *et al.*⁵

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There are many theories to determine the peel force,^{5,6} adhesive fracture energy or the interfacial adhesion⁷ when a film is being peeled from a substrate. The thin-film peeling theory developed by Kendall,⁸ which explains the peeling of a thin elastomer film from a rigid substrate, will be used to analyze the peeling in this context.

Consider an elastomer film of thickness d that is being peeled from a rigid substrate at a peel angle θ with a force F (Figure 1). The elastomer has a Young's modulus E . The adhesive energy between the film and the glass substrate is R . Consider a unit length Δc of the film being peeled from the substrate between the points A and B.⁸ The width of the film at any given point is b . The three contributions to the energy changes involved in the peeling process are

- (1) Surface energy due to the creation of new surfaces [$-bR\Delta c$].
- (2) Potential energy due to the movement of the applied force [$F(1 - \cos \theta)$].
- (3) Elastic energy due to the extension of the film in the direction of the applied force [$F^2\Delta c/2bdE$].

Adding up these contributions and assuming energy conservation, the following equation is obtained:⁸

$$-bR\Delta c + F(1 - \cos \theta)\Delta c + \frac{F^2\Delta c}{2bdE} = 0 \quad (1)$$

In our case, the corrugations on the surface of the carrier web make the release process all the more difficult because they increase the surface area, meaning a larger force is needed to peel the film.

From Equation (1), the adhesive energy R can be calculated. R is different from the surface energy, work of adhesion or any thermodynamic quantity, and R gives an estimate of the adhesive force between the substrate and the film. If the R between the substrate and the elastomeric film is high, then the force F required to peel the film will also be high. The smaller the peel angle, the smaller the potential energy contribution will be. Because the standard peel tests are performed at $\theta = \pi/2$ or π , the peel angle will not be modified. The elastomeric contribution to peel is governed by E , which is a material property. The surface energy contribution to peel force can be lowered if the surface energy can be decreased. This should preferably be done without affecting the bulk properties of the material (mechanical and dielectric) properties.

To decrease the surface energy of the PDMS, it will be modified with small quantities of a perfluoroether additive that contains low-energy trifluoromethyl (CF_3) groups.⁹ These low-energy CF_3 groups will migrate to the two surfaces of the silicone film and will segregate. Because of the high density of the CF_3 groups in the perfluoroether, it is possible to obtain a silicone surface with very low surface energy simply by adding very small quantities of perfluoroether. Perfluoroether allylamide ($(\text{F}(\text{CFCF}_3\text{CF}_2\text{O})_7\text{CFCF}_3\text{CONHCH}_2\text{CH}=\text{CH}_2$, PFE) added to curing PDMS (0.3–1.5

weight%) lowered the surface energy of PDMS from 19 to 8 mJ m^{-2} .⁹ The PFE molecules added to the PDMS are chemically bonded to the silicone network by the platinum-catalyzed hydrosilation reaction.⁹ The PDMS films were doped with 1% PFE (that is, 1% of the mass of PDMS), and the films were examined to determine whether they yield favorable results.

To investigate the influence of PFE on the mechanical properties, the linear rheological properties of the pure PDMS and PDMS doped with PFE will be examined. Furthermore, to investigate the changes induced on the surface of the films by the PFE, contact-angle measurements and peel tests on these surfaces have been performed. Dielectric permittivity tests are also conducted to investigate the influence of PFE on permittivity because an increase in the dielectric permittivity would be an advantage for DEAP films and would make it possible to address additional issues beyond process-related problems.¹⁰

The allyl groups ($-\text{CH}=\text{CH}_2$) of PFE react with the hydride groups ($-\text{Si}-\text{H}$) of the crosslinker (methyl hydrogen siloxane) and compete with PDMS in the hydrosilation reaction. To ensure proper bonding of PFE to the silicone network and complete crosslinking of PDMS, sufficient crosslinker has to be added to the reaction mixture. The effects of the additional crosslinker and platinum catalyst in the reaction mixture are also investigated. Hence, various compositions of crosslinker and catalyst are used in the reaction mixture.

EXPERIMENTAL PROCEDURE

Materials

The PDMS elastomer, oil and inhibitor used for the following set of experiments are: (1) Elastosil RT-625 (a commercially available RTV silicone); (2) Powersil Fluid TR50; and (3) Inhibitor PT 88, respectively, obtained from Wacker Chemie AG (München, Germany). The PFE (KDP-4645) was supplied by DuPont Krytox Performance Lubricants (Wilmington, DE, USA).

Carrier web. The carrier webs on which the release property of PDMS film were tested are made of a temperature-stabilized polyethylene terephthalate band (0.2 mm) coated with methyl acrylate UV resin. The surface of the carrier web has microscale corrugations (Figure 2). There are two types of carrier webs used at Danfoss PolyPower A/S (Nordborg, Denmark), depending on whether the corrugation lines are along the length of the web (down-web) or perpendicular to the length of the web (cross-web). In addition, there are two types of carrier webs defined by the wave depth and period. One has a depth of $5 \mu\text{m}$ and a period of $10 \mu\text{m}$ (30% web) and is capable of stretching to approximately 35% strain, and the other has both a depth and a period of $7 \mu\text{m}$ and is capable of stretching up to approximately 80% strain (100% web).^{1,11} The carrier web has negligible strain under the present process conditions, and the peel dynamics are not affected.

Instrumentation and specifications

Rheological experiments. Rheological measurements (time sweep and frequency sweep) on the silicone networks were performed with a controlled-stress rheometer AR-2000 (TA Instruments, New Castle, DE, USA).

Contact angle. Contact-angle experiments are performed using the Contact-Angle system (OCA Data Physics, Stuttgart, Germany). The probe liquids are water and hexadecane (product no. H0255) from Sigma-Aldrich (St Louis, MO, USA).

Peel test. Peel tests were performed in Danfoss PolyPower A/S using the Zwick/Roell (Zmart.pro, Ulm, Germany) material tester.

Dielectric permittivity. The dielectric permittivity tests on the samples were performed using a Novocontrol Alpha-A (Novocontrol Technologies GmbH & Co. KG, Hundsangen, Germany), a high-performance frequency analyzer.

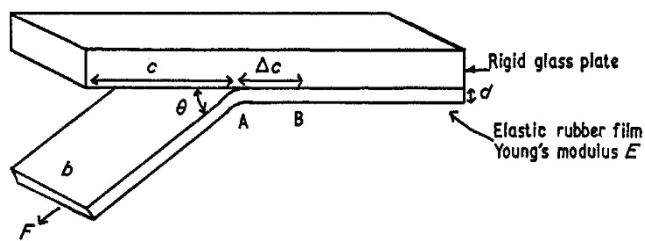


Figure 1 Peeling an elastomer film from a rigid substrate.⁸

Procedure

Preparing the addition-curing PDMS mixture and films. Elastosil RT-625 is supplied as premixes A and B. Premix A is a mixture of vinyl-terminated PDMS and crosslinker, while premix B is a mixture of vinyl-terminated PDMS and catalyst among other components such as fillers. Premixes A and B were mixed in the prescribed proportions (9:1) using the speed mixer DAC 150FVZ-K (Synergy Devices Ltd, Buckinghamshire, UK) for 2 min at 1000 r.p.m. Similarly, samples containing Elastosil RT-625 (9:1), 1% PFE (of the mass of Elastosil RT-625), 15% oil (of the mass of Elastosil RT-625) and 0.8% inhibitor (of the mass of Elastosil RT-625) were also mixed.

Another set of samples was made with Elastosil RT-625, in which A and B are mixed in the ratio 10:1, so that the resultant mixture contains more crosslinker. The amount of PFE added to this mixture was also 1 w/w%. The crosslinker percentage was increased in the second set of samples so that the PFE would have sufficient crosslinker to react with. Oil (15%) and inhibitor (0.8%) were also added to some elastomer samples of this set to investigate the influence of these constituents as well.

To examine the effects of additional catalyst, a new set of samples was made, in which Elastosil RT-625 A and B were mixed in the ratio 9:1.1. PFE (1%), oil (15%) and inhibitor (0.8%) were also added to this set of samples.

Mixing the premixes A and B of Elastosil RT-625 resulted in the hydrosilation reaction of the vinyl-terminated polydimethylsiloxane (PDMS) ($-\text{CH}=\text{CH}_2$) with the hydride crosslinker ($-\text{Si}-\text{H}$) in the presence of the platinum catalyst, resulting in a PDMS network. PFE also reacts with the crosslinker when it is present in the mixture (Figure 3). In total, 18 samples were made with varying the composition (Table 1) to investigate the effects of PFE, additional crosslinker and oil on the silicone films. The segregation of PFE in the surface is the same in the film–air interface and the film–substrate interface. The PFE migrates to the surfaces/interfaces of the PDMS film uniformly. Fourier transform infrared spectroscopy curves showing the peaks

with the same intensity for PFE on both the interfaces of the PDMS film are presented in Supplementary Figures S2–5.

Rheological tests. To investigate the effects of PFE, additional crosslinker and catalyst on the mechanical properties of Elastosil RT-625, rheological tests were performed.

Time sweep. Time sweeps of 18 samples (Table 1) were performed at 2% strain, at temperatures of 23 and 80 °C and at a frequency of 1 Hz. The applied strain (2%) was designed to be within the linear regimen of the material. Samples with the inhibitor were cured at 80 °C because the inhibitor inhibits curing at room temperature. Samples without the inhibitor were cured at 23 °C.

Frequency sweep. Mechanical characterization (frequency sweeps) of the 18 samples was performed by making LVE measurements. Films with a thickness of 1 mm (1000 μm) were prepared with the 18 sample mixtures. After complete curing, the films were cut to make 25 mm diameter discs. LVE measurements on these samples were performed from a frequency of 100–0.001 Hz with 2% strain (which we ensured was within the linear regimen of the material based on an initial strain sweep) using the 25 mm aluminum parallel plate geometry at 25 °C. The normal force applied by the aluminum disc on the sample was 5–10 N.

Contact-angle tests. As described above, addition-curing mixtures were prepared with the 18 different compositions. Films with a thickness of 1 mm were made on a flat substrate. Once fully cured, the contact angles of the films were tested.

Advancing and receding contact-angle experiments were performed on the 18 samples using water as the probe liquid. Static contact-angle measurements were made with hexadecane as the probe liquid. Because hexadecane swells the

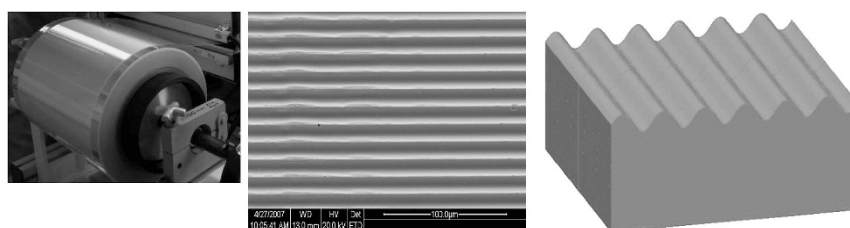


Figure 2 The microscale corrugations on the carrier web.¹ A full color version of this figure is available at *Polymer Journal* online.

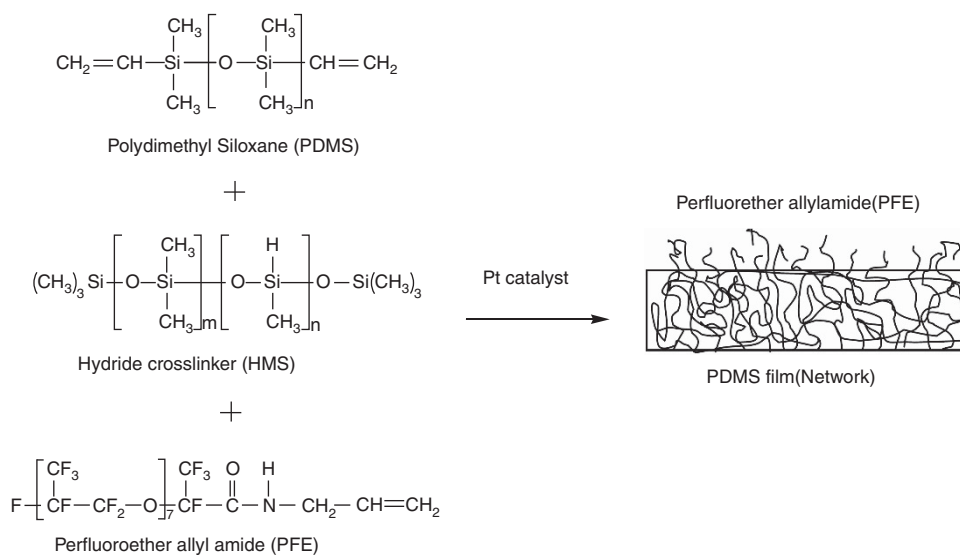


Figure 3 Modification of the polydimethylsiloxane (PDMS) network with perfluoroether allylamide (PFE) (hydrosilation reaction).⁹

Table 1 Composition of the 18 samples and their properties

Sample no.	PFE					GP		$G'(\omega \rightarrow 0)$	$E = 3G$	Contact angle with <i>n</i> -hexadecane	Surface energy	Max. peel force F'	Average peel force	Adhesive energy
	Elastosil (A:B)	(ww % ⁻¹)	Inhibitor (ww% ⁻¹)	Oil (ww% ⁻¹)	Temperature (°C)	(min)	t_{97} (min)							
1	9:1	—	—	—	23	65.8	780	0.19	0.57	32.5	23.3	0.015	0.007	16.1
2	9:1.1	—	—	—	23	53.9	317	0.06	0.18	28.2	24.3	0.017	0.009	27.0
3	9:1	1	—	—	23	50.5	406	0.06	0.18	55.0	17.0	0.018	0.008	34.2
4	9:1.1	1	—	—	23	54.7	789	0.05	0.15	60.1	15.1	0.016	0.009	32.3
5	9:1	1	0.8	15	80	5.2	52.2	0.05	0.15	57.0	16.4	0.014	0.009	28.0
6	9:1.1	1	0.8	15	80	4.4	67.8	0.03	0.09	63.8	14.3	0.023	0.014	73.9
7	9:1	1	0.8	—	80	4.4	19.8	0.06	0.18	61.0	15.1	0.019	0.013	37.8
8	9:1	1	—	15	23	77.3	1012	0.025	0.08	55.2	16.9	0.011	0.007	39.7
9	9:1.1	1	0.8	—	80	5.5	42.6	0.05	0.15	56.3	16.5	0.018	0.010	41.4
10	9:1.1	1	—	15	23	80.3	826	0.09	0.28	52.5	17.8	0.012	0.007	15.4
11	9:1	—	0.8	15	80	4.7	37.5	0.04	0.12	28.7	24.2	0.012	0.006	20.9
12	9:1.1	—	0.8	15	80	3.4	30.3	0.034	0.10	44.0	20.3	0.022	0.013	61.7
13	10:1	—	—	—	23	45.9	454	0.04	0.12	47.8	19.4	0.016	0.010	26.9
14	10:1	1	—	—	23	51.5	1246	0.09	0.27	42.2	20.8	0.016	0.008	19.2
15	10:1	1	0.8	15	80	7.9	87.5	0.04	0.12	53.9	17.4	0.013	0.009	29.6
16	10:1	1	0.8	—	80	7.2	50.8	0.074	0.22	56.9	16.4	0.018	0.011	27.3
17	10:1	1	—	15	23	62	679	0.03	0.09	49.0	18.9	0.012	0.008	36.0
18	10:1	—	0.8	15	80	6.9	61	0.05	0.15	30.3	23.9	0.012	0.009	21.4

Abbreviation: GP, gel point.

surface of the PDMS films, the contact-angle measurements were made within 5–10 s of the drop coming into contact with the film.

Peel tests. Peel tests were performed on the 18 samples to estimate the peel force and the adhesive force. Films with a thickness of 100 μm were made on the 30% down-web with the addition-curing mixtures using a 3540 bird film applicator (Elcometer, Aalen, Germany). The dimensions of the samples were 30 \times 30 mm². Once the films were fully cured on the carrier web, they were tested for peel force with a peel angle of 90°. A peel curve was obtained from plotting the peel force versus the length of sample peeled.

Dielectric permittivity. For the permittivity tests, films with a thickness of 1 mm were made from the 18 addition-curing mixtures, and once fully cured, discs with a diameter of 25 mm were cut from the films. They were then tested for their dielectric permittivity.

RESULTS AND DISCUSSION

Rheological tests

Time sweeps. The time sweeps of samples 6 and 12 are shown in Figure 4; these samples show the greatest deviation because they contain the highest (1%) and lowest (0%) amounts of PFE, respectively. After the onset of the hydrosilation reaction, the PDMS network approaches chemical gelation. A crosslinking polymeric system is said to reach its gel point at a critical extent of the crosslinking reaction at which either the weight-average molecular weight diverges to infinity (infinite sample size) or the first macromolecular cluster extends across the entire sample (finite sample size).^{12,13} The gel point that is at the crossover between G' and G'' is an important processing parameter for DEAP materials; for more details, see Bejenariu *et al.*¹⁰ Chambon and Winter¹² and Winter.¹³ The addition of PFE to the Elastosil RT-625 does not modify the gel point significantly. From the results, it can be concluded that the addition of 1 w/w% PFE does not lead to any changes in the process conditions for the RTV rubber, which is very favorable for the process.

Frequency sweeps. The frequency sweeps of samples 6 and 12 are compared in Figure 5. Again, these are the samples showing the most deviation because they contain 1 and 0% PFE, respectively. From the frequency sweeps (Figure 5), it is evident that the storage modulus G' of pure Elastosil RT-625 film is not greatly influenced by the addition of PFE. The addition of PFE lowers the elastic modulus by a few percent, which is within the experimental uncertainty of the measurements, but the increase in G'' with the addition of PFE confirms that there is a small but still fairly insignificant decrease in the elasticity of the material with PFE. The deviation in G'' is much clearer because the magnitude of G'' is dominated by the very small sol fraction. In contrast, G' is dominated by the elastically active material.¹⁴

In Table 1, the elastic moduli of all the samples $G'(\omega \rightarrow 0)$ and other process-related properties, such as the time for the material to obtain 97% of its final strength (t_{97}) and the crossover point of G' and G'' , are tabulated. The t_{97} is also an important parameter for many processing considerations because it may be overly expensive to wait for the last 3% of reaction, which proceeds very slowly, and in the case of fairly stoichiometric networks, does not influence the elasticity significantly.

Contact-angle and surface energy

Advancing and receding water drop. Advancing and receding water contact-angle experiments were performed on the 18 samples. The Elastosil RT-625 samples without PFE had an average water contact angle of 110°, which is characteristic of PDMS.² Samples with 1 w/w% PFE had an average water contact angle of 115°. The results/experimental data from the contact-angle tests are presented in Supplementary Figure S1.

Static contact angle with *n*-hexadecane. The static contact angles measured with *n*-hexadecane are tabulated in Table 1.

*Fowkes' method to calculate the surface energy.*¹⁵

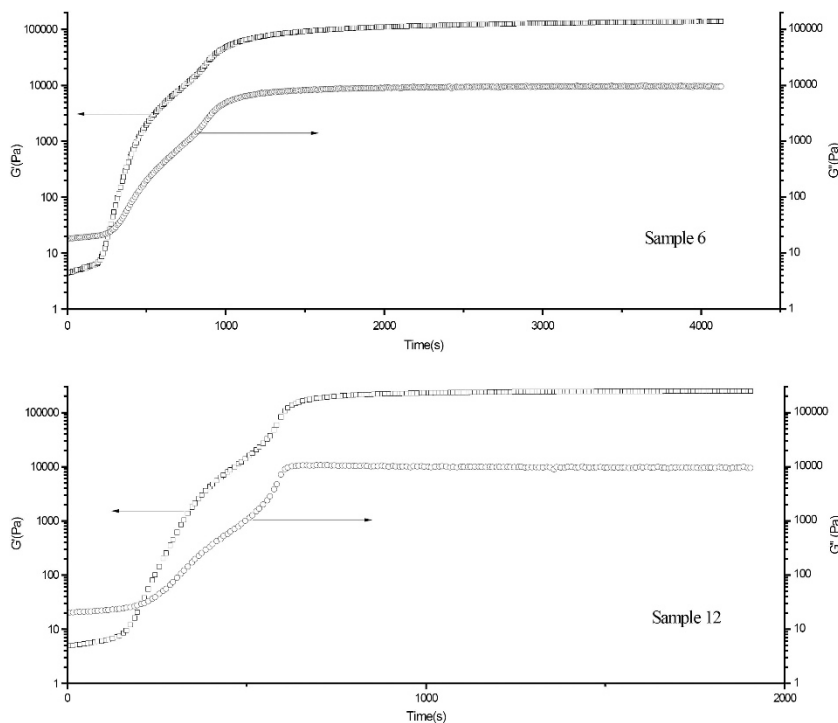


Figure 4 Curing profiles of sample 6 (Elastosil (9:1.1)+oil (15%)+inhibitor (0.8%)+perfluoroether allylamide (PFE) (1%)) and sample 12 (Elastosil (9:1.1)+oil (15%)+inhibitor (0.8%)) at 80 °C. A full color version of this figure is available at *Polymer Journal* online.

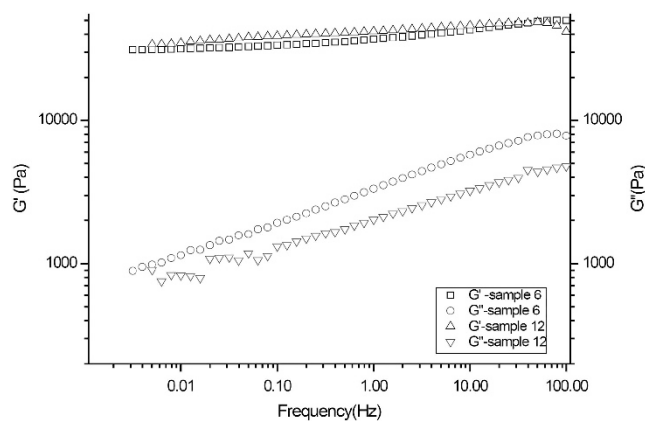


Figure 5 Frequency sweeps of sample 6 (Elastosil (9:1.1)+oil (15%)+inhibitor (0.8%)+ perfluoroether allylamide (PFE) (1%)) and sample 12 (Elastosil (9:1.1)+oil (15%)+inhibitor (0.8%)) at 23 °C. A full color version of this figure is available at *Polymer Journal* online.

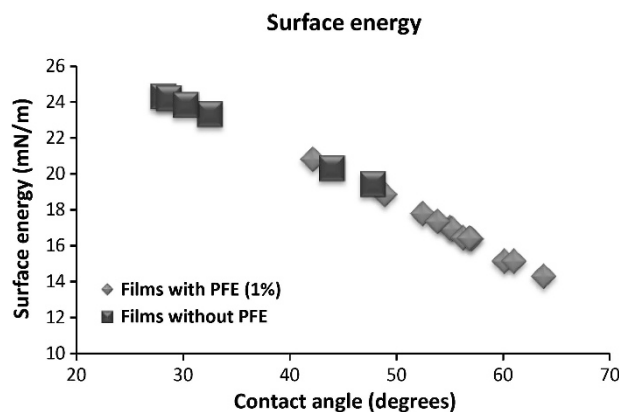


Figure 6 Surface energies calculated using Fowke's method. A full color version of this figure is available at *Polymer Journal* online.

According to the Fowkes' method, the surface energy of a solid surface (S) can be calculated using the contact angle of a liquid (L) using a simple formula:

$$\frac{\sigma_L(\cos\theta + 1)}{2} = \sqrt{\sigma_S^P \sigma_L^P} + \sqrt{\sigma_S^D \sigma_L^D} \quad (2)$$

where $\sigma_L = \sigma_L^D + \sigma_L^P$, σ is the surface energy (surface tension), P denotes the polar component of the surface tension and D is the dispersive component of the surface tension.

When we use a liquid whose polar component is zero, the surface tension of the liquid will then be $\sigma_L = \sigma_L^D$. Hence, Equation (2) will

become

$$\sigma_S^D = \frac{\sigma_L(\cos\theta + 1)^2}{4} \quad (3)$$

Thus, using the contact angles of a polar liquid, such as water, and a nonpolar liquid, such as *n*-hexadecane, one can estimate the surface energy of a solid using Equations (2) and (3). The results are tabulated in Table 1. The samples doped with PFE have a markedly lower surface energy than the pure Elastosil RT-625 samples (Figure 6).

Figure 6 shows a comparison of the surface energy with the contact angle of *n*-hexadecane. Films with PFE have a higher contact angle with *n*-hexadecane and a lower surface energy compared with the films without PFE. However, samples 12, 13, 14 and 17 (20.3, 19.4, 20.8 and 18.9 mN m⁻¹, respectively) have almost similar surface energy values; samples 14 and 17 contain PFE, and samples 12 and 13 do not contain PFE.

Peel tests

A peel curve obtained from the peel test of sample 3 is shown in Figure 7. The peel force F (N mm⁻¹), which is the force used in peeling a width of 1 mm of the sample, is plotted against the length of the sample that is being peeled. From the peel curve, we see that F increases to a maximum value and then falls to a low constant value. From examining the F values of the samples, it is observed that samples 6 and 12 with 15% oil and 0.8% inhibitor have the maximum F values. Samples 5, 8, 10, 11, 15, 17 and 18, all of which contained 15% oil, have low F values compared with the average of 0.0157 N mm⁻¹. In contrast, samples 7, 9 and 16, which contain 0.8% inhibitor (without oil), had higher F values than the average of 0.0157 N mm⁻¹. The presence of oil in the addition curing mixtures certainly makes the release easier because it acts like a release agent. The values of the maximum and constant peel force of all samples are tabulated in Table 1.

In Figure 8, the surface energy and the F of the samples are plotted. It is observed that the samples with a low surface energy and those with a high surface energy both have similar patterns with F . The reduction of surface energy of the samples did not reduce or affect their F . Because the carrier web is very thick and rigid, the stretching of the web is never observed during the experiments.

Calculation of adhesive force 'R'. Using the peel Equation (4), the adhesive energy R is calculated [8]

$$E\left(\frac{F}{b}\right)^2 \frac{1}{2dE} + \left(\frac{F}{b}\right)(1 - \cos\theta) - R = 0 \quad (4)$$

$$E = 2G(1 + \nu)l \quad (5)$$

where G is the static shear modulus and ν is the Poisson ratio.

F is obtained from the peel tests, and E is calculated from Equation (5). The value of G ($G = G'(\omega \rightarrow 0)$)¹⁶ is obtained from

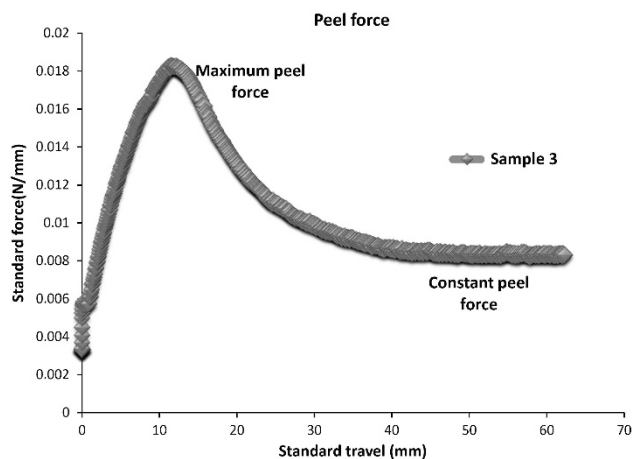


Figure 7 Typical peel curve. A full color version of this figure is available at *Polymer Journal* online.

the frequency sweeps (Table 1) of the rheological tests and the Poisson ratio (ν) for silicone elastomers (rubber), which was taken as 0.5. The maximum value of F is used for all calculations to estimate the maximum R between the film and the substrate during the processing. A comparison of samples 2 and 12 clearly shows that as E increased, R decreased. (Sample 2 has $F = 0.016$ N mm⁻¹, $E = 0.18$ MPa and the calculated $R = 27$ N m⁻¹. Sample 12 has $F = 0.022$ N mm⁻¹, $E = 0.10$ MPa and the calculated $R = 61.7$ N m⁻¹.)

From Figures 9 and 10, one can understand the relationship between E , R and F . From Figure 9, it is observed that as F increased, the value of R also increased. To lower the peel force, the adhesive energy needs to be lowered. From Figure 10, it is observed that R and E are inversely related, in accordance with Equation (1). As the E value of the samples increased, their R value decreased, thus making their release easier. Although the surface energy, F and R at the interface are related through the action of surface forces, the relationship is not obvious.¹⁷ The relationship between F , R , E and release can be understood further from the theory proposed by Johnson *et al.*¹⁷ Every surface has a surface energy resulting from the action of surface forces. When two surfaces are in intimate contact, these surface forces act as attractive (adhesive) forces. The strength of the attractive (adhesive) force between two surfaces depends on the contact surface area.¹⁷ Interfacial gaps due to surface asperities or dust particles will strongly influence the adhesion between the surfaces because the attractive forces decrease rapidly with increasing separation.¹⁷ Materials with a low elastic modulus will even out easily against a substrate and make very good contact. Therefore, they are strongly adhered to the substrate.¹⁷ Hence, to peel the film off, one needs to overcome the adhesive forces and apply a high F . To make the release easier, the contact at the interface needs to be reduced to prevent the surfaces from adhering strongly. That is what release agents such as oils and surfactants do, namely, reducing the strength of the contact to ease the release of a surface from another surface. In contrast, materials with a high Young's modulus will not flatten out against another surface and cannot make intimate contact and hence experience a lower adhesive force.

Dielectric permittivity

In Figure 11, the dielectric permittivity ($\epsilon = \epsilon' + i\epsilon''$) of samples 1, 3, 5, 13, 14 and 15 is shown. The dielectric constant is essential

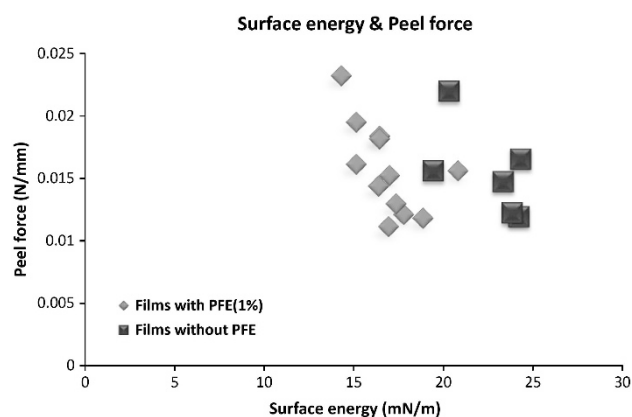


Figure 8 Comparison of the surface energies of the samples with peel force F . A full color version of this figure is available at *Polymer Journal* online.

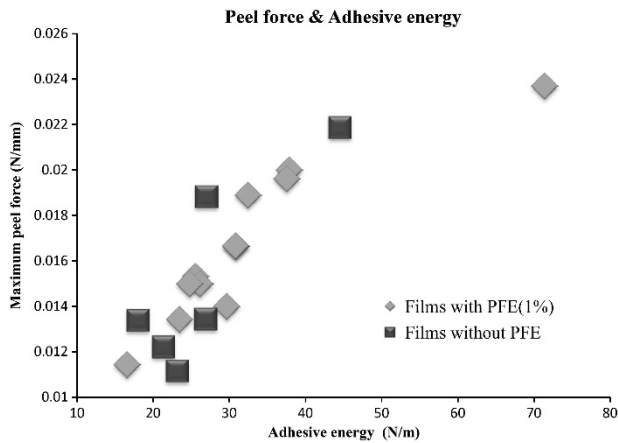


Figure 9 Relationship between the adhesive energy R and the peel force F . A full color version of this figure is available at *Polymer Journal* online.

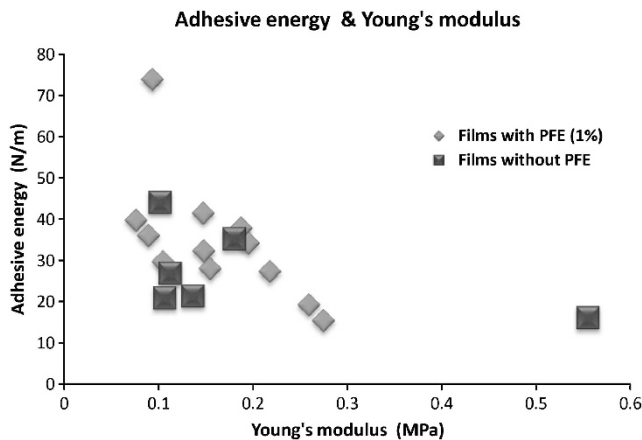


Figure 10 Relationship between the adhesive energy R and Young's modulus E . A full color version of this figure is available at *Polymer Journal* online.

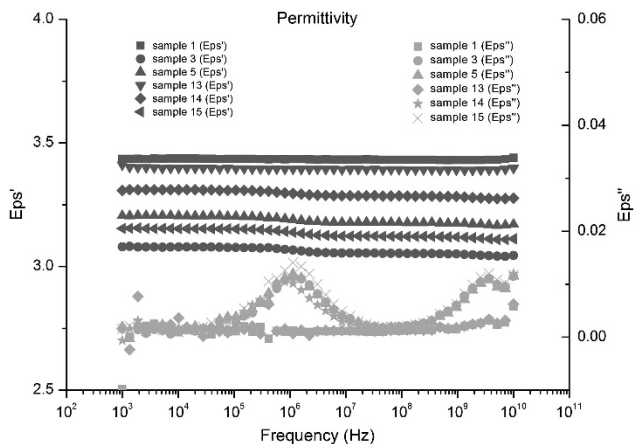


Figure 11 Dielectric permittivity test ($Eps' = \epsilon'$ and $Eps'' = \epsilon''$). A full color version of this figure is available at *Polymer Journal* online.

information when designing capacitors or, in our case, an actuator, which operates according to the same principle as a capacitor.

The permittivity (ϵ) value of PDMS films used in our experiment is usually approximately 3.2.¹⁰ The tests conducted confirmed that the ϵ values of the samples were not greatly influenced by PFE addition and were well within the allowed limits. The variation in ϵ was in the range of 3.1–3.4 (Figure 11). In addition, the low variation in permittivity values can be attributed to the addition of oil and inhibitor.

CONCLUSIONS

PDMS was doped with 1% PFE to lower the surface energy, and as expected, the surface energy was lowered. The addition of PFE to the Elastosil RT-625 did not greatly influence the storage modulus G' , as observed from the time sweeps. The dielectric permittivity of the samples was also not modified by the addition of PFE. Elastosil RT-625 doped with PFE has a high contact angle in the advancing drop. When the drop is receding, the surface behaves more hydrophilic than the pure Elastosil RT-625 films. This can be due to the presence of high-energy moieties near the surface. The amide groups immediately beneath the surface are highly polar and hence become exposed when water (polar liquid) comes into contact with the surface.⁹

Although the surface energy decreased with the addition of PFE, the peel force values did not decrease. The reason for this is that the elastic contribution to the peel force is markedly higher than the potential and the surface energy contribution. For the investigated elastomer films, the Young's moduli are low, which causes the films to easily smooth out over surfaces and make good contact. This contact increases the adhesive forces, making the release difficult. The samples with a low Young's modulus have a higher adhesive energy between the film and the substrate than those with a higher Young's modulus. Therefore, tuning the elasticity of the networks is actually the easiest path for decreasing the release problems of thin silicon films when other solutions such as using a release agent are not an option.

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