Electronic Supplementary Information for Explaining the spread in measurement of PDMS elastic properties: influence of test method and curing protocol

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1 Connectedness of existing literature

Litmaps software was used to create a connectedness diagram of all the studies used in this review shown in Figure ESI.F1. Lötters¹ is seen to be particularly commonly cited with work on PDMS.

L Litmaps



Fig. ESI.F1 Included studies show citation links between articles. Markers are sized based on relevance within the network and sorted chronologically from left to right, and vertically on a log scale by total citations in any field per the Litmaps Database²

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2 Additional data

Previously unpublished data included in this paper was collected with the method described by Chockalingam et al.³ via cyclic loading between a radius of a = 0.06 mm and 0.08 mm with 400 seconds of relaxation. A neo-Hookean material model is considered and a non-linear least squares fitting is used to determine an initial defect size *A* (used to calculate a stretch λ) and shear modulus μ .

All samples are SYLGARD 184 mixed via a THINKY planetary mixer and cured at 100 °C for 2 hours. Sample identifiers indicate the composition used as $S[w_{B+oil}]$ -[(w_{oil}/w_{B+oil})100] e.g. S40-10 is a 40:1 base to curing agent ratio with 10% non-reactive PDMS oil (μ MicroLuburol, 350cSt). Reported shear modulus in Table ESI.T1 is the mean $\bar{\mu}$ of results collected for each rate of \dot{a} = 0.01, 0.02, 0.04, 0.08, 0.16, and 0.32 [mm/s] with a standard deviation calculated from all six rates for all trials, *N*, within a sample.

Sample ID	$w_B + : w_{oil} : w_C$	N _{trials}	$ar{\mu} \pm \sigma$ [kPa]	$\bar{A} \pm \sigma$ [mm]
\$55-10	50+5.6:1	2	3.65 ± 0.11	0.326 ± 0.01
S50-00a	50+0:1	4	5.71 ± 0.16	0.319 ± 0.01
S50-00b	50+0:1	3	4.71 ± 0.51	0.321 ± 0.01
S50-00c	50+0:1	2	5.47 ± 0.29	0.342 ± 0.007
S50-10	45+5:1	3	7.38 ± 0.37	0.366 ± 0.02
S50-20	40+10:1	3	7.24 ± 0.34	0.395 ± 0.006
S45-00a	45+0:1	1	11.9 ± 0.24	0.448 ± 0.02
S45-00b	45+0:1	4	10.6 ± 0.81	0.414 ± 0.03
S40-00	40+0:1	4	14.4 ± 1.8	0.460 ± 0.02

Table ESI.T1 Additional data presented in this work with associated composition and fit parameters for shear modulus and initial defect size

3 Included studies

A summary of all studies included in the manuscript is listed in Table ESI.T2. Curing temperature listed as 25 °C for all room temperatures cures and specified as RT in the Heating Method column. Unspecified methods are abbreviated as UNS throughout.

Table ESI.T2 Summary of all included studies	

Author	Test Type	Cure Temp [C]	Cure Time [hr:min]	Mix Ratio [base+oil: curing agent]	Heating method	Mixing and degas
Chockalingam 2021 ³	VCCE	100	2:00	variable, Si Oil	Oven	Planetary mixer, vac-
Raayai-Ardakani 2019 ⁴	VCCE	40	72:00	variable	Oven	Planetary mixer, vac-
Raayai-Ardakani 2019 ⁵	VCCE	40	72:00	variable	Oven	Planetary mixer, vac-
This work	VCCE	100	2:00	variable, Si Oil	Oven	Planetary mixer, vac-
Milner 2021 ⁶	Cavitation Rheology	25	144:00		RT	Planetary mixer, vac-
Milner 2021 Thesis ⁷	Cavitation Rheology	25	144:00	variable	RT	Planetary mixer, vac-
Yang 2019 ⁸	Cavitation Rheology	25	144:00	variable, Si Oil	RT	Planetary mixer, vac-
Numer 20119	Cimple Cheer		144.00			uunii uegas
Upadhyay 2010 ¹⁰	Simple Shear	23 60	3.00	10.1	Oven	UNS mixing vacuum
			5.00			degas 2 hr
Brown 2005	Tension	60	20:00	10-50:1	Oven	UNS mixing, vacuum degas 30 min
Fuard 2008 ¹²	Tension	100	1:30, 2:00	variable	Oven	UNS
Johnston 2014 ¹³	Tension†	25-200	variable	10:1	Oven	Flocculator, degas 30 min
Khanafer 2008 ¹⁴	Tension	65	12:00	6-10:1	Oven	UNS mixing, vacuum degas >2 hr
Kim 2011 ¹⁵	Tension			5-15:1	UNS	UNS
Liu 2009 ¹⁶	Tension†	100 or 200	variable	10:1	Hot plate	UNS
Mills 2008 ¹⁷	Tension‡	150	12:00	10:1	UNS	UNS
Moučka 2021 ¹⁸	Tension†	25-150	variable	10:1, Si Oil	UNS	Vacuum mixer 100 RPM 10 min
Schneider 2008 ¹⁹	Tension	150	0:15	10:1	Oven	Hand mix 6 min, de- gas
Seghir 2015 ²⁰	Tension	10 or 160	2:00, 144:00	variable	Hot plate	UNS with 5 low vac-
Upadhyay 2021 ²¹	Tension	60	3:00	10:1	Oven	UNS mixing, vacuum
Wang 2019 ²²	Tension	65	4:00	10-30:1	Oven	Planetary mixer, vac-
Wang 2014 ²³	Compression	65	12:00	5-33:1	Oven	UNS mixing, vacuum
Carrillo 2005 ²⁴	Compression &	25	336:00	10-30:1	RT	UNS mixing 10 min
	Nanoindentation				Tlat mlata	
$Cao 2005^{-1}$	Nanoindentation	/0 65	24:00	10:1	Hot plate	UNS
Cheng 2011	Nanoindentation	05	1:30	10:1	cure	spin coated, vacuum
Gupta 2007 ²⁷	Nanoindentation	25	0:20, 2:40	variable	RT	UNS
Liao 2010 ²⁸	Nanoindentation	85	2:00	variable	UNS	UNS
Mata 2005 ²⁹	Nanoindentation	95	0:30	5.7-21:1	Oven	UNS mixing, spin coated
Patel 2019 ³⁰	Nanoindentation	70	5:00	10:1	UNS	UNS mixing, vacuum degas 30 min
Peng 2011 ³¹	Nanoindentation	60	20:00	50:1	Oven	UNS mixing, vacuum degas
Armani 1999 ³²	Beam Bending		0:15	5-15:1	UNS	 UNS
Du 2010 ³³	Beam Bending	65	1:30	10:1	Hot plate	UNS
Li 2024 ³⁴	Cylindrical Cavity	- 100	2:00	variable	Oven	Planetary mixer, vac-
Thangawng 2007 ³⁵	Membrane	110	0:15	10:1	Hot plate	UNS, spin coated

4 Distribution of predictor variables

Spearman Correlations were chosen to describe the data for two main reasons: first, we wanted to to avoid imposing linearity in the relationship between the predictors (cure time, cure temperature, or mix ratio) and the resulting stiffness. Second, all three predictors are not normally distributed (rejecting the null hypothesis that the data are from a normally distributed population at a confidence level of p < 0.05 for the Kolmogorov-Smirnov test). Ordinal scale plots of the predictor variables are included in Fig. ESI.F2.



Fig. ESI.F2 Ordinal plot of predictor variables.

5 PDMS reaction chemistry

PDMS is formed from a base with bi-vinyl terminated -R²Si-O- units combined with a curing agent containing S-H silane groups to form siloxane repeating units -Si-O- between cross links in the presence of a platinum catalyst ^{36–38}. The average molecular weight between cross links M_c is determined by the curing agent ratio, with the higher weight, and therefore longer, chains resulting in softer material. SYLGARD 184 is not pure PDMS, however the reaction pathway remains the same ^{36,39}. Schweitzer et al.⁴⁰ report M_c increases 3-6 fold between w_B:w_C of 10:1 and 20:1 with SYLGARD 184 (2400 to 7600 g/mol). Measuring five curing agent ratios between 10:1 and 25:1, the increasing M_c increases exponentially. This supports the nonlinear relation between μ and w_B:w_C observed in Fig. 2 given ⁴¹ $M_c \propto 1/\mu$.

6 Predictor variable contour plots

To visually examine the inter dependencies of the predictor variables, Fig. ESI.F3 shows the contour plots for the permutations of mix ratio, cure time and cure temperature. Contour plots were generated in MATLAB 2023b using the Curve Fitting Toolbox and linearly interpolated surfaces. Fig. ESI.F3(a) shows how the combined effects of higher cross linker concentration and hotter cure temperature lead to the stiffest material while Fig. ESI.F3(b) and (c) confirm that total cure time does not have a strong effect on shear modulus.



Fig. ESI.F3 Contour plots of the effect of mixing ratio, cure temperature and cure time on shear modulus. The effect of mixing ratio is seen to outweigh the effect of cure time. (a) compares the effect of mixing ratio and cure temperature on shear modulus with all points shown as black circles. Both predictors are seen to have an effect on the resulting stiffness. (b) compares cure time and mixing ratio while (c) compares cure time and temperature with mix ratio and temperature dominating over time, respectively.

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