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.](#page-0-0) Lötters 1 1 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 Database^{[2](#page-5-1)}

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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Previously unpublished data included in this paper was collected with the method described by Chockalingam et al.^{[3](#page-5-3)} via cyclic loading between a radius of *a* = 0.06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](#page-1-0) 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.

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.](#page-2-0) 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.

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.](#page-3-0)

Fig. ESI.F2 Ordinal plot of predictor variables.

5 PDMS reaction chemistry

PDMS is formed from a base with bi-vinyl terminated $-R^2$ Si-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](#page-5-36)[–38](#page-5-37). 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,](#page-5-36)[39](#page-5-38). Schweitzer et al. [40](#page-5-39) 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](#page-5-40)} $M_c \propto 1/\mu$.

6 Predictor variable contour plots

To visually examine the inter dependencies of the predictor variables, Fig. [ESI.F3](#page-4-0) 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\(](#page-4-0)a) shows how the combined effects of higher cross linker concentration and hotter cure temperature lead to the stiffest material while Fig. [ESI.F3\(](#page-4-0)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.

Notes and references

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