Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

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Mapping the mechanical and electrical properties of commercial silicone elastomer formulations for stretchable transducers

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S1. Experimental procedure:

S1.1 Gel fraction

To determine the gel fraction (W_{gel}), triplicates of disc samples, 500 ± 100 μm thick and 15 mm in diameter, are immersed in solvent, n-heptane (VWR International, France), for 72 hours at room temperature. After swelling for 3 days, samples are separated from the solvent and additional solvent is evaporated under low pressure (34 mbar) at 50°C for 4 hours. The gel fraction is calculated as the weight after extraction and drying (m_1) divided by the initial weight of the sample (m_0) as W_{gel} (%) = $(m_1/m_0) \times 100$.

S1.2 Sample thickness measurement

Silicone film sheets, of around 250 cm^2 in area, are prepared for each composition. Specimens of predefined shapes are cut and used for investigating the materials' properties. In order to determine the film thickness small strips (around 2 x 10 mm) are cut out from several locations of the sheet and the thickness of cross-sections of the strips is evaluated using optical microscopy technique. At least three strips are tested to calculate the average thickness and standard deviation.

S1.3 Mechanical properties

Stress–strain measurements are performed with Instron 3345 tensile tester equipped with a 500 N load cell. The applied crosshead velocity is 50 cm/min. Samples of 10 cm long, 2 cm wide and 300 ± 30 μm thick are used for tensile tests with Instron. When samples are clamped on the instrument, the area between the clamps is 5 cm in length and 2 cm in width, the thickness is 300 ± 30 μm, except for Elastosil Film 2030 250/200, which is 200 μm thick. Then samples are stretched with a rate of 50 cm/min (8.3 mm/s) until the samples break. Tests are made on 6 specimens for each sample. The software Bluehill Universal is used to calculate Y (at the strain of 10%), tensile strain, and tensile stress.

Typically, dog-bone shape of samples is recommended for stress-strain measurements. This particular shape serves many purposes. In our opinion, the two most important ones are: 1) enhanced clamping width compared to effective sample width in order to prevent slippage of specimens and 2) ensuring that samples snap in the middle of the specimen where the width is the smallest rather than at the contact points with the clamp. We have considered the possibility of using the dog-bone and rectangular specimens. Initial results showed that none of the aforementioned points constituted a problem in our measurements and therefore the squared samples are used out of convenience.

Hysteresis measurements are performed using the ElectroForce, with samples that are 4 cm long, 3 cm wide and 300 ± 30 μm thick, except for Elastosil Film 2030 250/200, which is 200 μm thick. Samples are clamped between two plastic holders and these holders are fixed to the instrument. After clamping, sample dimensions are 2 cm long and 3 cm wide. Samples are stretched to 50% with frequency of 1 Hz (stretch rate 10 mm/s), 1000 times. Measurements are repeated twice.

S1.4 Breakdown strength

Breakdown tests are performed on an in-house-built instrument following the international standards (IEC 60243-1 (1998) and IEC 60243-2 (2001))[1]. Film samples of thickness 100 ± 20 μm are placed on a plastic support frame during breakdown testing. The plastic frames with silicone film are placed into the breakdown instrument between two semi-spherical electrodes made of stainless steel. Subsequently, 100 V/s voltage ramp is applied until polymer film broke down. Measurements are repeated 12 times. Breakdown strengths are calculated from Weibull analysis[2].

S1.5 Shore hardness

Shore hardness is measured with AD-300 High End Durometers (Checkline Europe, Netherlands) A and 00 type. Samples at least 6.4 mm thick are used for this measurement and measurements are repeated 3 times per sample.

S1.6 Viscosity and pot life

Viscosity and pot life are measured with AR2000 rheometer, using 20 mm diameter stainless steel parallel plate (Smart Swap, UK) where the gap between plates is adjusted to 0.8 mm. The temperature is set to 25 ℃, and a shear rate 0.1 s^{-1} is used. Shear rate for Sylgard 186 and Sylgard MIX 1:3 is 0.524 s^{-1} because samples are too viscous to test at the rate of 0.1 s^{-1}. Viscosity is measured right after mixing part A and B and pot life is determined when the viscosity of the mixture doubled.[3]

S1.7 Transmittance

Optical Transmittance measurements are made using an UV−vis spectrophotometer POLARstar Omega microplate reader by BMG LabTech. Elastomer films of 100 μm thickness are used for measurements. Transmittance measurements are made from a wavelength of 220 nm to 1000 nm taking points at every 2 nm. Samples are pre-stretched by 20%, 50% and 100% and attached on plastic holder. Measurements are repeated 3 times.

S1.8 Dielectric permittivity

Novocontrol Broadband Dielectric Spectrometer, Germany, is used to investigate dielectric properties. Disc samples (500 ± 100 μm thick, 25 mm diameter) are sputtered with silver (silver layer is 5 nm) and placed between two gold electrodes. Tests are performed at room temperature in the frequency range from 10^{-1} to 10^{6} Hz at 1 V. Measurements are made on 4 specimens for each sample.

SI.9 Leakage current

Leakage current is measured with PK-SPIV17 high voltage leakage current, TSDC, and Pyroelectric test system (PolyK Technologies, USA). Samples of thickness 100 ± 20 μm are sputter coated with 5nm thick silver electrodes from both sides with a 2 X 2 cm active area. The coated sample is then placed inside a temperature control chamber, on a gold grounding electrode. The top electrode is lowered to contact the sample. Then a 500 V voltage is applied and tests are performed in 25 ^{o}C, 40 ^{o}C, and 60 ^{o}C temperature. When voltage is applied, the leakage current gradually decreases. When leakage current stabilizes and reaches a plateau, measurement is considered over. Measurements are made on 3 specimens for each sample.

S2. Results:

S2.1 Pot life

Pot life of the formulations are tabulated below.

Table S1: The pot life of the formulations

Sample name	**Pot life (min) at 25°C**
Sylgard 184 5:1	>60
Sylgard 184	**>60**
Sylgard 184 15:1	>60
Sylgard 184 20:1	>60
Sylgard 186 5:1	44
Sylgard 186	**56**
Sylgard MIX 1:3	27
Sylgard MIX 1:1	>60
Sylgard MIX 3:1	>60
Ecoflex 00-50	**16**
Eco MIX 00-50 1:3	>60
Eco MIX 00-50 1:1	>60
Eco MIX 00-50 3:1	>60
Ecoflex 00-30	**28**
Eco MIX 00-30 1:3	>60
Eco MIX 00-30 1:1	>60
Eco MIX 00-30 3:1	>60
Ecoflex 00-10	**28**
Eco MIX 00-10 1:3	>60
Eco MIX 00-10 1:1	>60
Eco MIX 00-10 3:1	>60

S2.2 Shore hardness

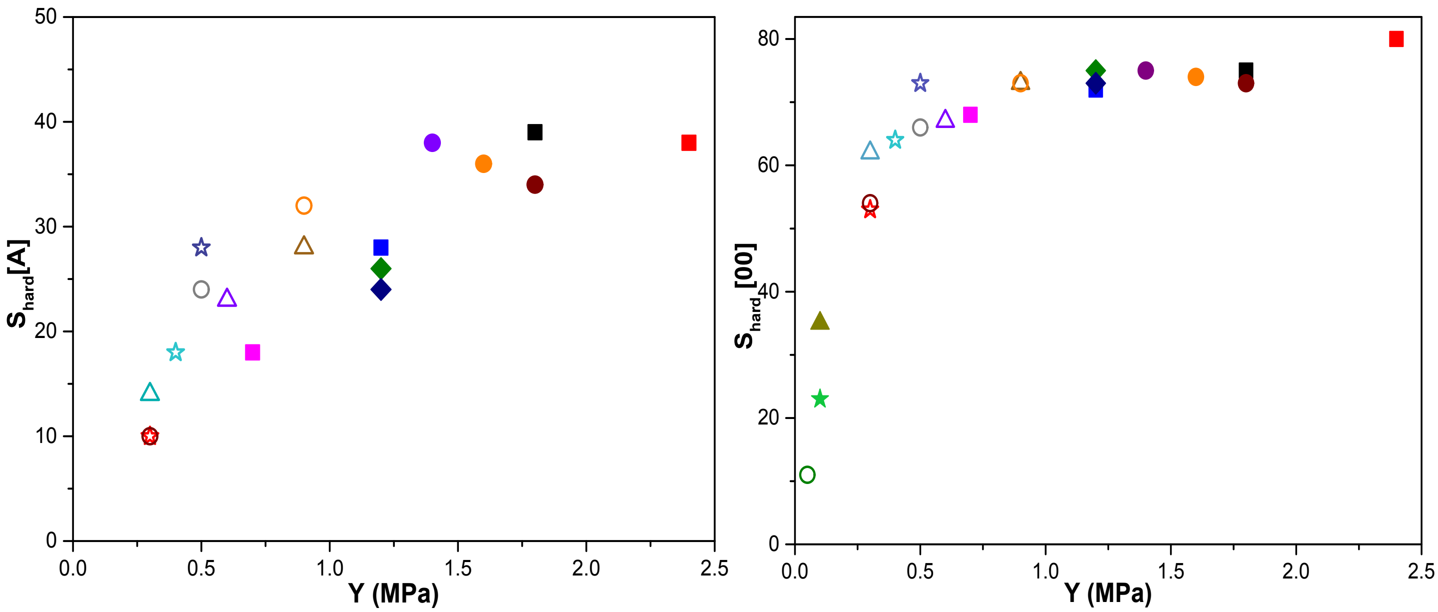

Figure S1: (left) Shore hardness [A], (right) Shore hardness [00] of elastomers plotted against the respective Young's modulus

S2.3 Dielectric permittivity

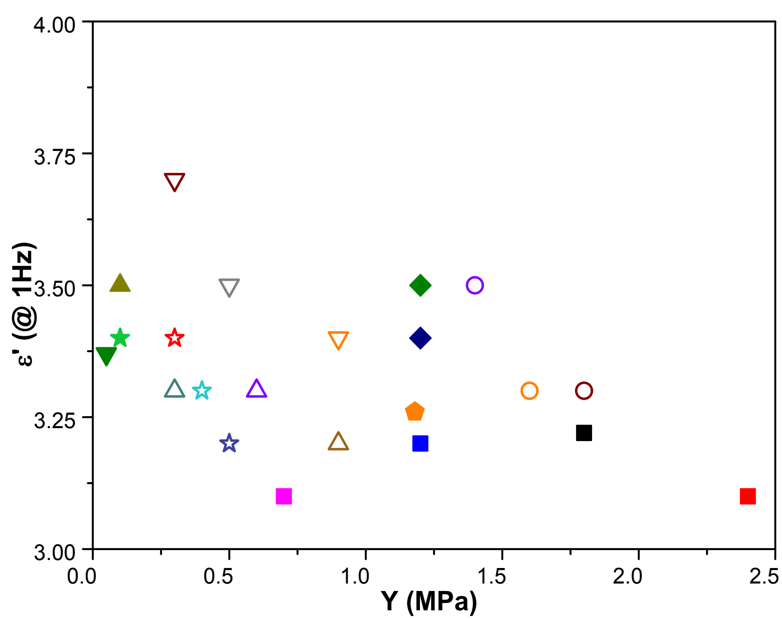

Figure S2: Dielectric permitivitty of elastomers plotted against the respective Young's modulus

Table S2: Leakage current (i_{leak}) at an electric field of 5 V/µm and at various temperatures

Formulation	**i_{leak}(·10^{-11}, A) @ 25°C**	**i_{leak} (·10^{-11}, A) @ 40°C**	**i_{leak} (·10^{-11}, A) @ 60°C**	**k (°C^{-1})**
Sylgard 184 5:1	2.16	3.22	12.9	0.052
Sylgard 184	**10.64**	**25.89**	**87.9**	**0.061**
Sylgard 184 15:1	1.6	2.6	5.32	0.034
Sylgard 184 20:1	5.46	7.52	15.1	0.029
Sylgard 186 5:1	2.1	5.25	27.2	0.074
Sylgard 186	**8.31**	**25.5**	**108**	**0.073**
Sylgard MIX 1:3	2.42	5.6	20.7	0.062
Sylgard MIX 1:1	2.04	4.02	12.8	0.053
Sylgard MIX 3:1	6.62	1.48	5.82	0.063
Ecoflex 00-50	**2.98**	**7.8**	**31.1**	**0.067**
Eco MIX 00-50 1:3	8.44	1.54	6.15	0.057
Eco MIX 00-50 1:1	4.6	1.21	4.06	0.062
Eco MIX 00-50 3:1	1.14	1.75	6.85	0.052
Ecoflex 00-30	**2.23**	**2.99**	**28.9**	**0.075**
Eco MIX 00-30 1:3	0.36	0.996	5.11	0.076
Eco MIX 00-30 1:1	2.34	5.91	26.1	0.069
Eco MIX 00-30 3:1	5.0833	10.2386	48.0	0.065
Ecoflex 00-10	**10.5**	**26.7**	**53.7**	**0.046**
Eco MIX 00-10 1:3	0.293	0.588	1.11	0.038
Eco MIX 00-10 1:1	0.124	0.437	1.61	0.073
Eco MIX 00-10 3:1	0.839	1.86	4.97	0.051
Elastosil	**4.01**	**10.0**	**37.9**	**0.064**

S2.4 Transmittance

The transmittance (T) of the elastomers at various pre-stretches at a wavelength of 550nm are tabulated in Table S3. The T of the elastomers over the entire visible spectrum is plotted further below.

Table S3: T (550nm) at different pre-stretch

Sample name	**0% pre-stretch**	**20% pre-stretch**	**50% pre-stretch**	**100% pre-stretch**
Sylgard 184 5:1	93.6	93	91	-
Sylgard 184	**94.4**	**94.2**	**91.4**	**-**
Sylgard 184 15:1	94	94	94	93
Sylgard 184 20:1	94	94	94	93.7
Sylgard 186 5:1	92.5	91.5	91	91
Sylgard 186	**93**	**93**	**91.7**	-
Sylgard MIX 1:3	93.3	92.2	93	-
Sylgard MIX 1:1	93.8	92.6	92.5	-
Sylgard MIX 3:1	94	93.05	91.5	-
Ecoflex 00-50	**78.2**	**69.4**	**78.2**	**45.5**
Eco MIX 00-50 1:3	78	72	57	-
Eco MIX 00-50 1:1	82	75	69	-
Eco MIX 00-50 3:1	87	84	80	-
Ecoflex 00-30	**78.4**	**75.8**	**65.7**	**54.6**
Eco MIX 00-30 1:3	77	68	64	-
Eco MIX 00-30 1:1	83	78	74	-
Eco MIX 00-30 3:1	89	86	84	-
Ecoflex 00-10	**70.5**	**53.2**	**38**	**26.5**
Eco MIX 00-10 1:3	76	62	48	38
Eco MIX 00-10 1:1	82	73	63	-
Eco MIX 00-10 3:1	88	82	80	-
Elastosil	**91**	**90**	**88.4**	**89**

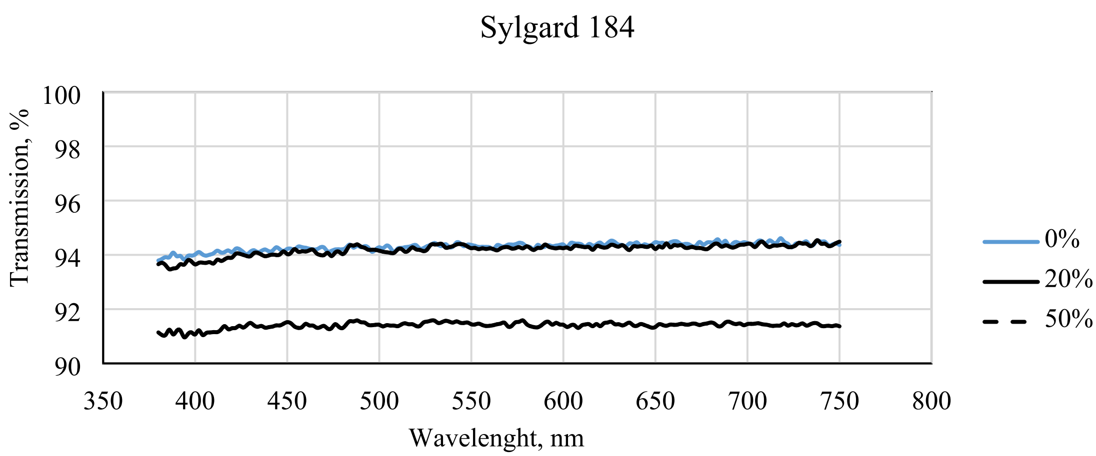

Figure S3: Transmittance of Sylgard 184 over the entire visible spectrum

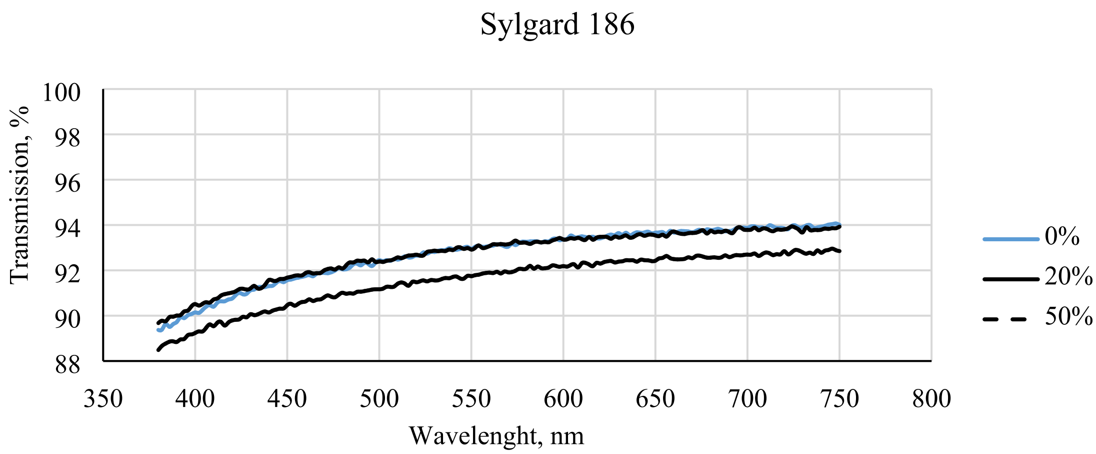

Figure S4: Transmittance of Sylgard 186 over the entire visible spectrum

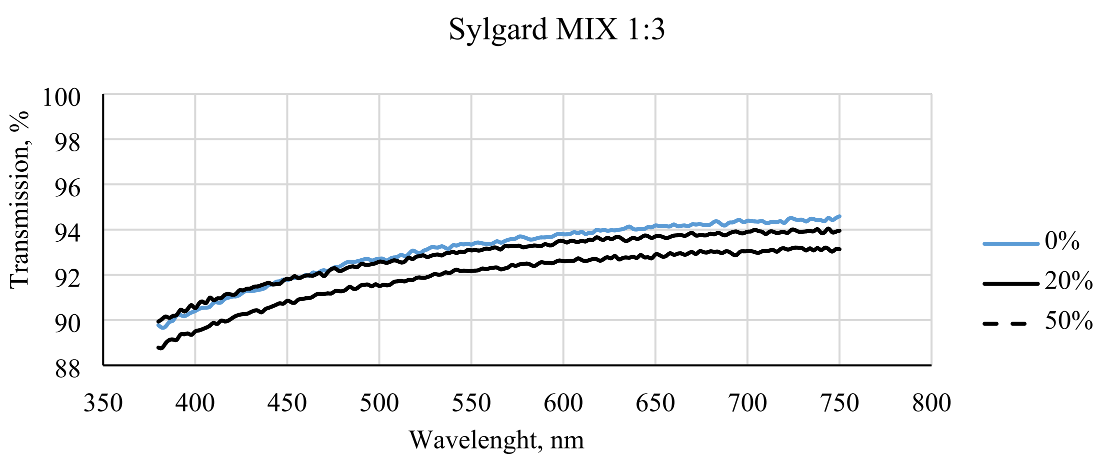

Figure S5: Transmittance of Sylgard MIX 1:3 over the entire visible spectrum

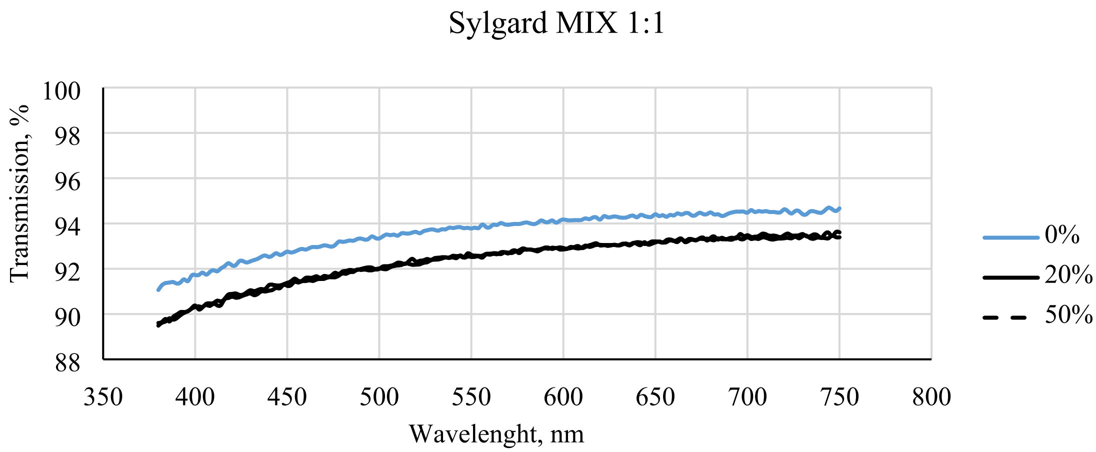

Figure S6: Transmittance of Sylgard MIX 1:1 over the entire visible spectrum

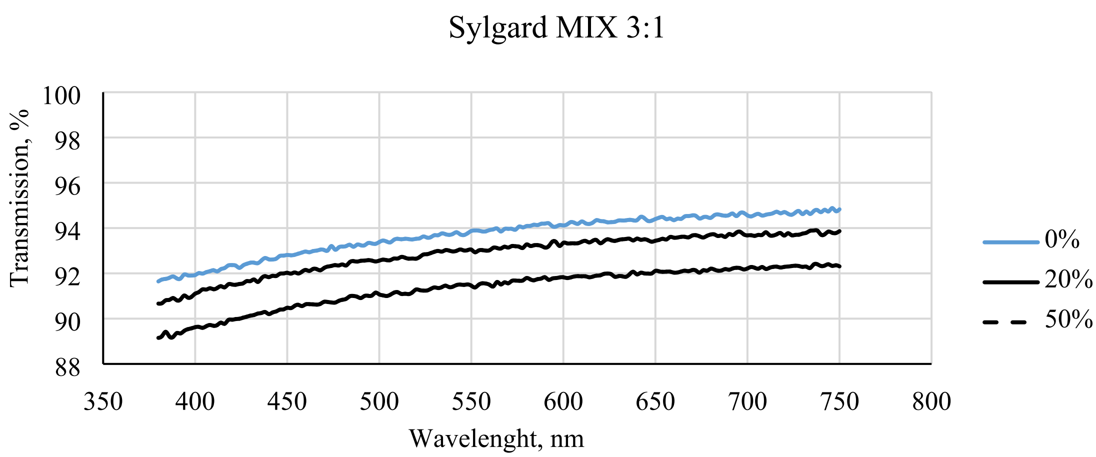

Figure S7: Transmittance of Sylgard MIX 3:1 over the entire visible spectrum

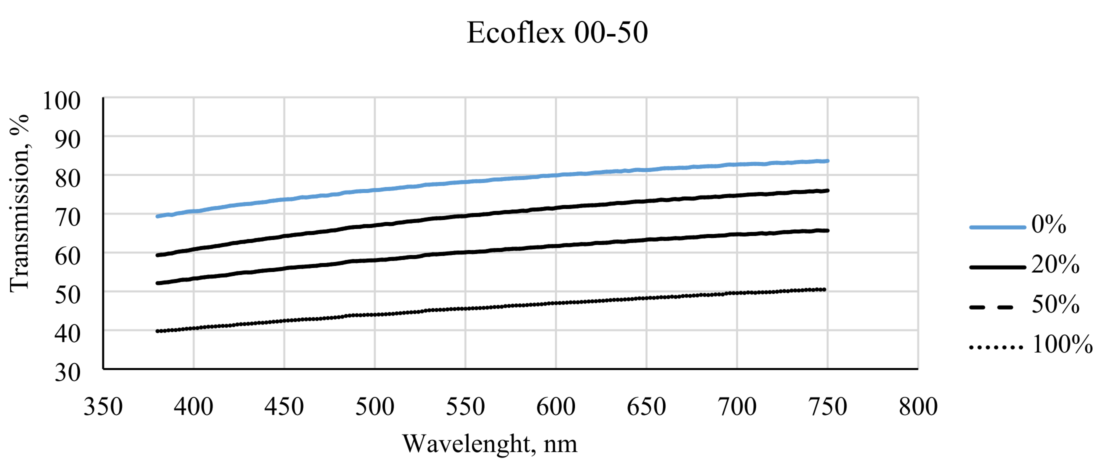

Figure S8: Transmittance of Ecoflex 00-50 over the entire visible spectrum

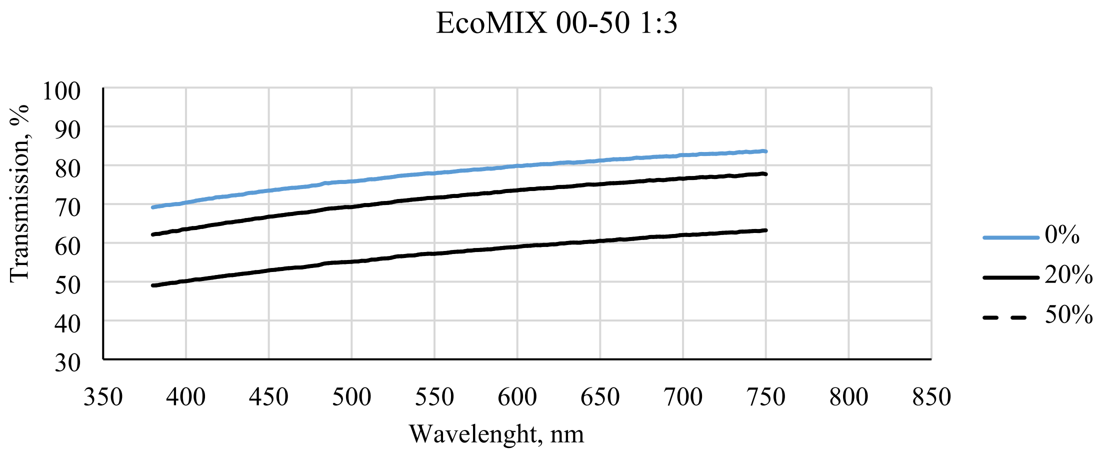

Figure S9: Transmittance of EcoMIX 00-50 1:3 over the entire visible spectrum

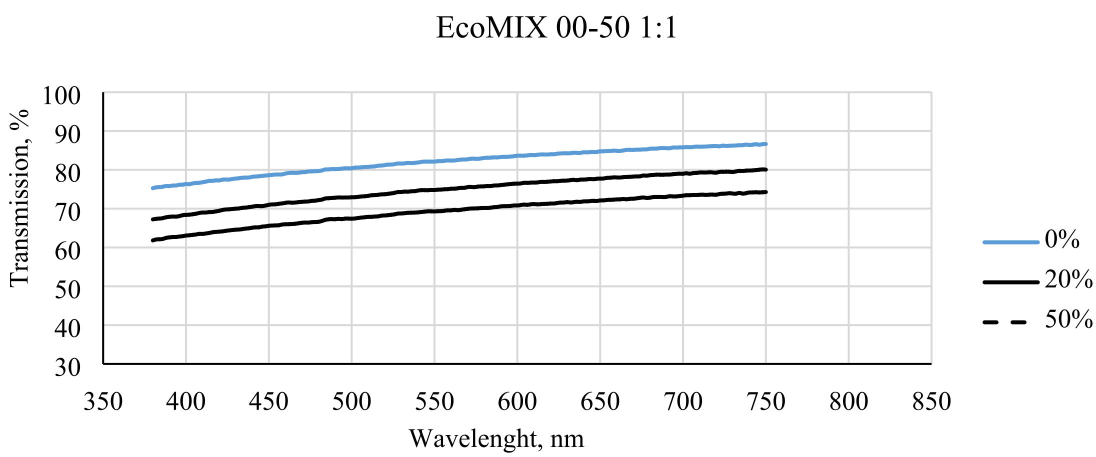

Figure S10: Transmittance of EcoMIX 00-50 1:1 over the entire visible spectrum

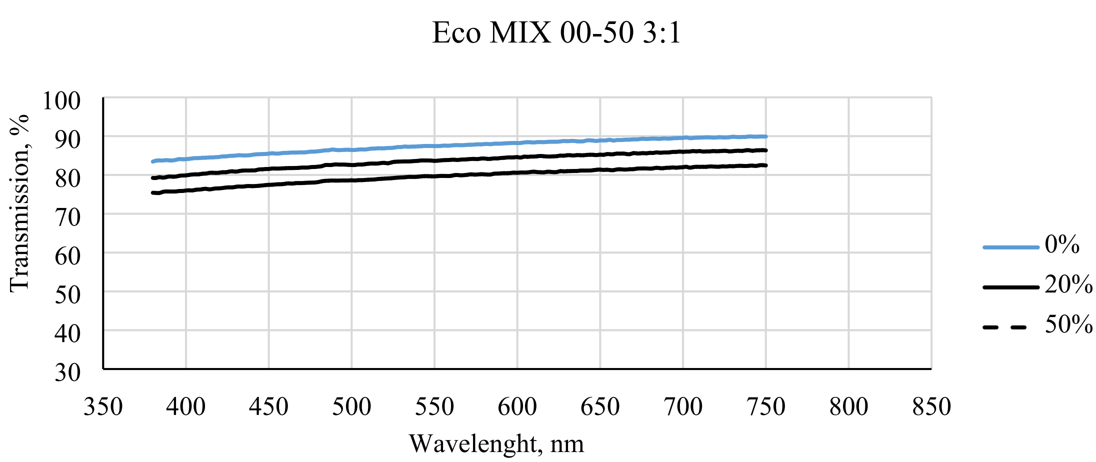

Figure S11: Transmittance of EcoMIX 00-50 1:3 over the entire visible spectrum

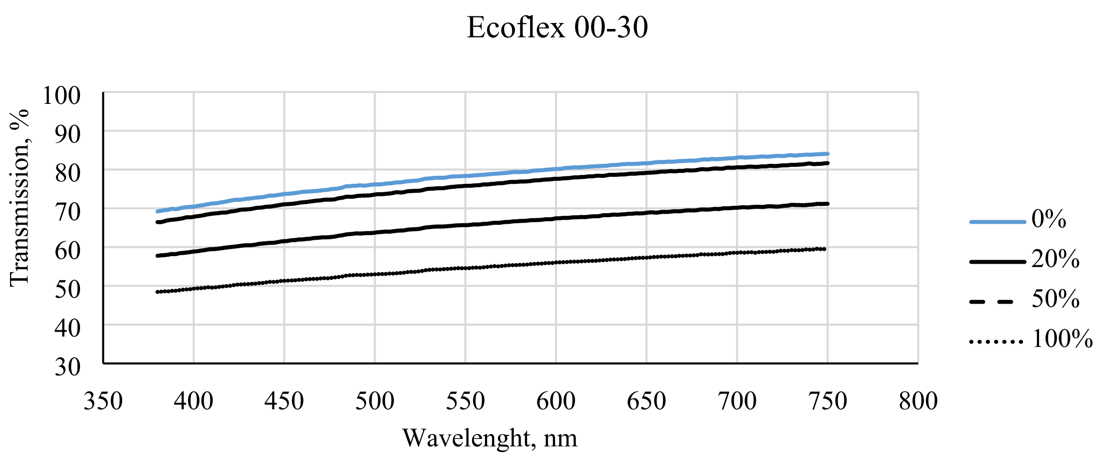

Figure S12: Transmittance of Ecoflex 00-30 over the entire visible spectrum

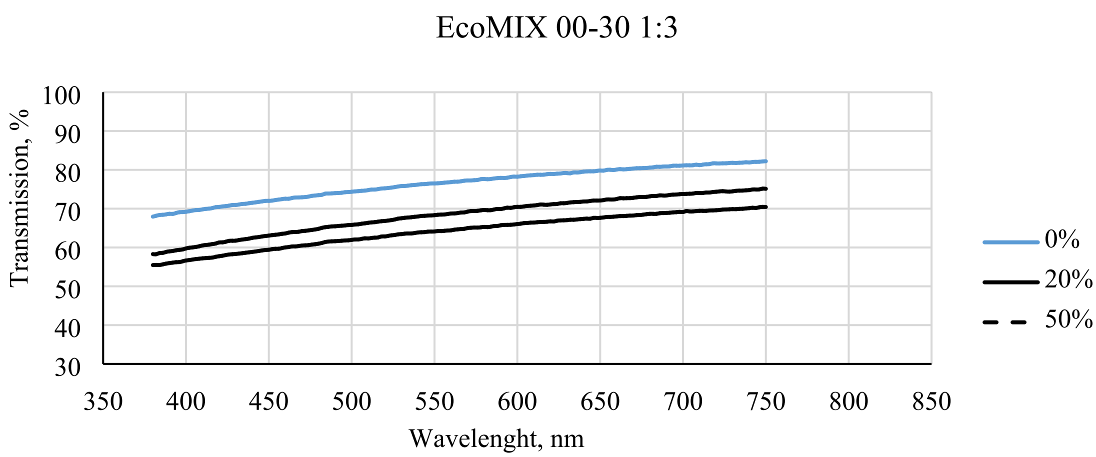

Figure S13: Transmittance of EcoMIX 00-30 1:3 over the entire visible spectrum

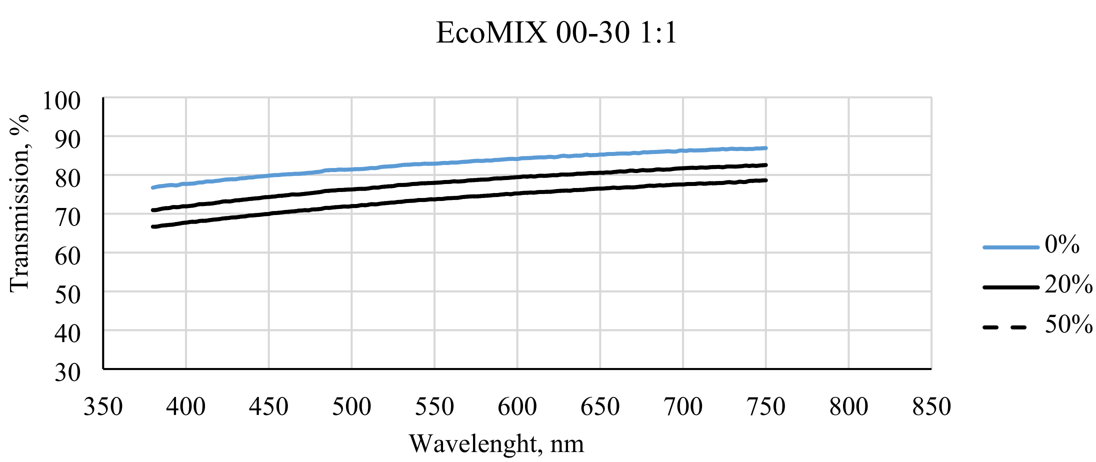

Figure S14: Transmittance of Eco MIX 00-30 1:1 over the entire visible spectrum

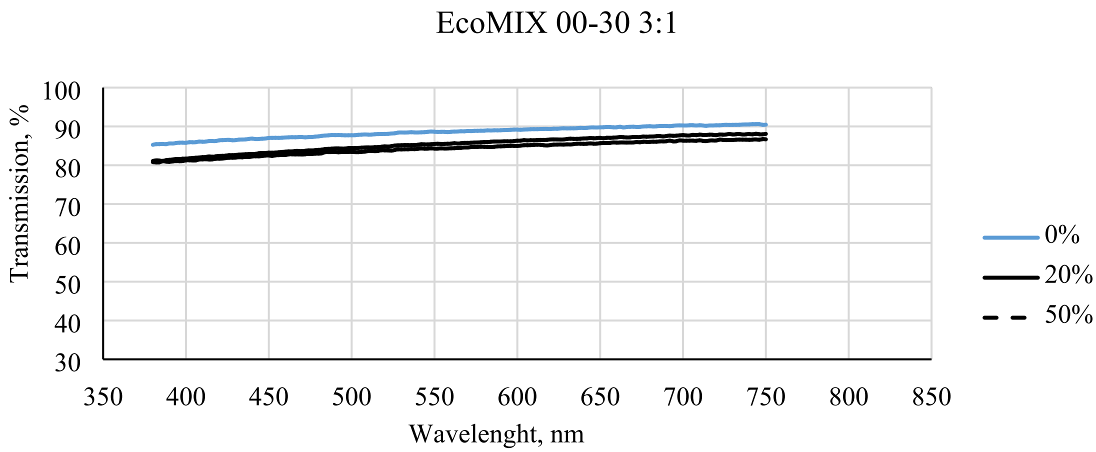

Figure S15: Transmittance of EcoMIX 00-30 3:1over the entire visible spectrum

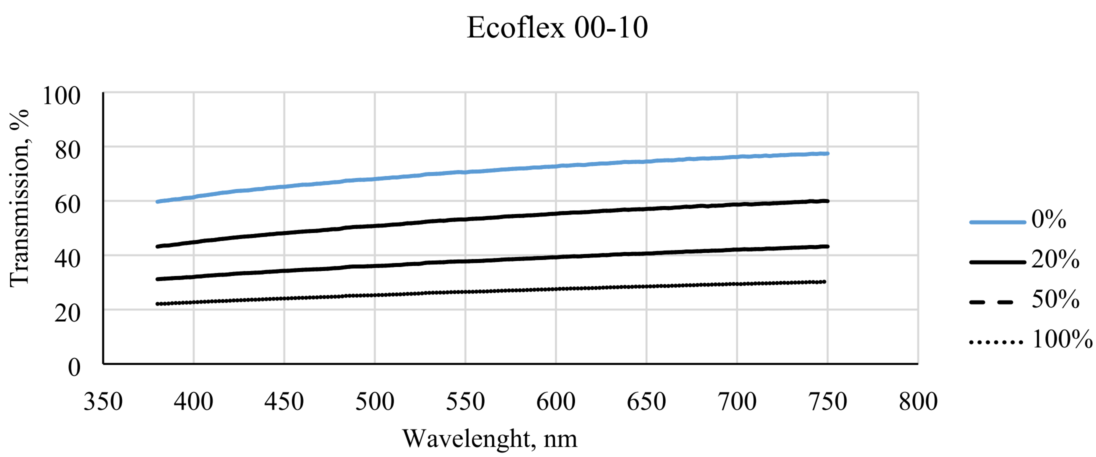

Figure S16: Transmittance of Ecoflex 00-10 over the entire visible spectrum

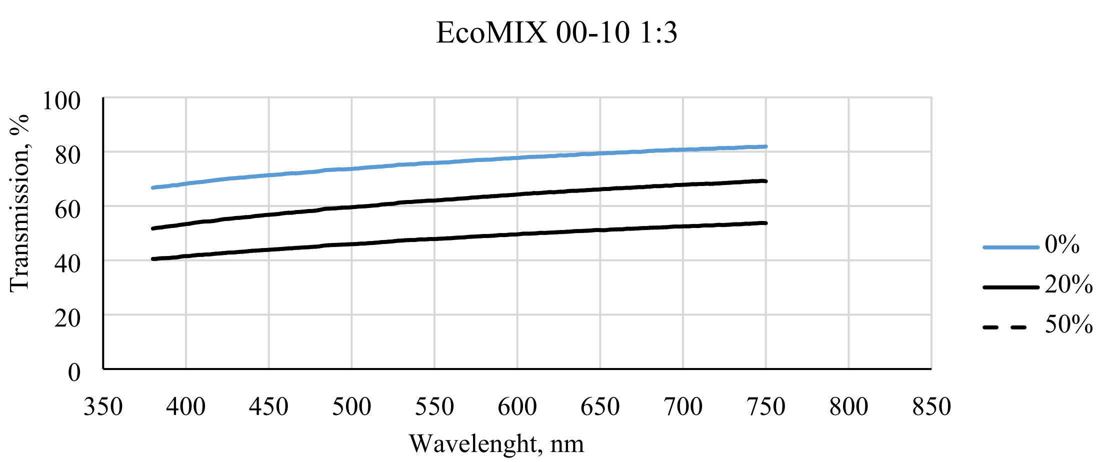

Figure S17: Transmittance of EcoMIX 00-10 1:3 over the entire visible spectrum

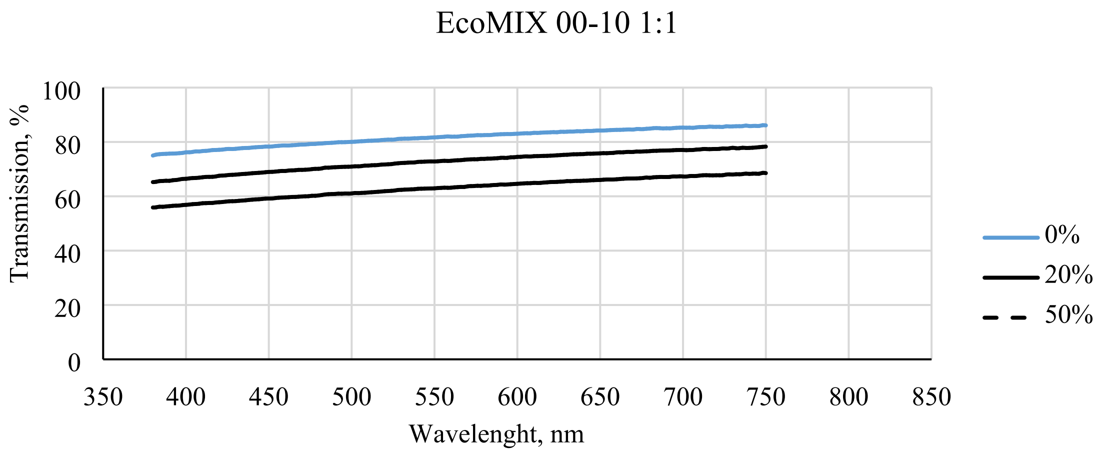

Figure S18: Transmittance of EcoMIX 00-10 1:1over the entire visible spectrum

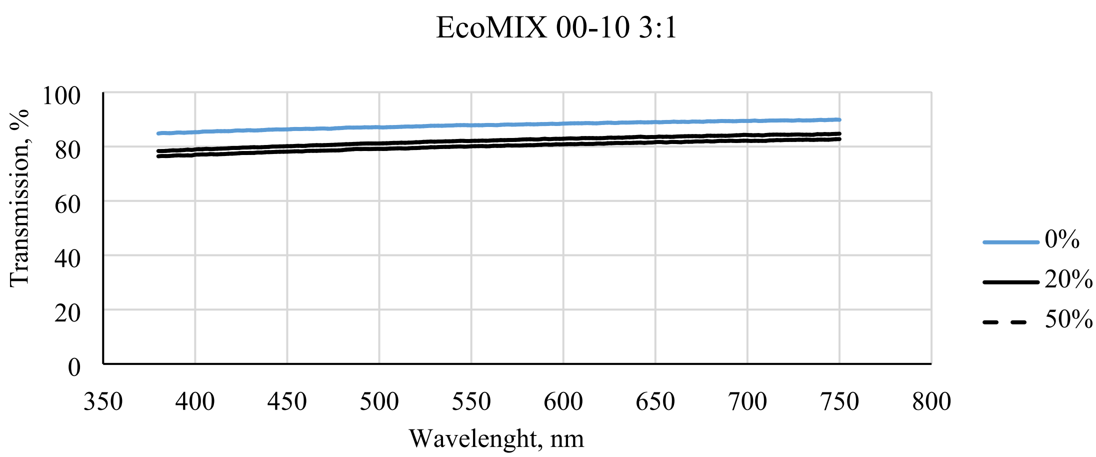

Figure S19: Transmittance of EcoMIX 00-10 3:1over the entire visible spectrum

S3. Properties of aged samples:

The properties of the elastomers are measured after 6 months and are tabulated and plotted below.

Table S4: Effects of ageing on Young's modulus (Y), Tensile stress(σ_{max}), Elongation at break ($\varepsilon_{tensile}$) for the formulations

Formulation	**Y (Mpa)**	**Y (MPa) after 6 months**	**$\varepsilon_{tensile}$ (%)**	**$\varepsilon_{tensile}$ (%) after 6 months**	**σ_{max} (MPa)**	**σ_{max} (MPa) after 6 months**
Sylgard 184 5:1	1.8	2.2	122	118	5.1	7.3
Sylgard 184	**2.4**	**1.6**	**136**	**140**	**7.1**	**6.5**
Sylgard 184 15:1	1.2	0.9	237	244	4.5	4.6
Sylgard 184 20:1	0.7	0.6	296	280	2.7	2.7
Sylgard 186 5:1	1.2	0.9	505	430	4.3	3.6
Sylgard 186	**1.2**	**1.3**	**567**	**394**	**5.6**	**3.9**
Sylgard MIX 1:3	1.4	1.5	365	278	4.2	3.6
Sylgard MIX 1:1	1.6	1.4	242	220	4.7	4.9
Sylgard MIX 3:1	1.8	2	177	184	5.6	6.6
Ecoflex 00-50	**0.1**	**0.1**	**860**	**812**	**1.7**	**1.3**
Eco MIX 00-50 1:3	0.3	0.4	351	302	1.6	1.6
Eco MIX 00-50 1:1	0.6	0.5	247	204	2.6	1.8
Eco MIX 00-50 3:1	0.9	0.9	204	147	3.7	3
Ecoflex 00-30	**0.1**	**0.06**	**835**	**685**	**1.2**	**0.8**
Eco MIX 00-30 1:3	0.3	0.25	373	284	1.6	1.2
Eco MIX 00-30 1:1	0.4	0.44	241	224	1.6	2
Eco MIX 00-30 3:1	0.5	0.9	228	155	1.8	3.4
Ecoflex 00-10	**0.05**	**0.07**	**573**	**534**	**0.4**	**0.6**
Eco MIX 00-10 1:3	0.3	0.2	342	325	1.6	1.6
Eco MIX 00-10 1:1	0.5	0.5	238	224	2.3	2.4
Eco MIX 00-10 3:1	0.9	0.8	218	163	3.9	3.2
Elastosil	**1.18**	**-**	**529.7**	**-**	**6.5**	**-**

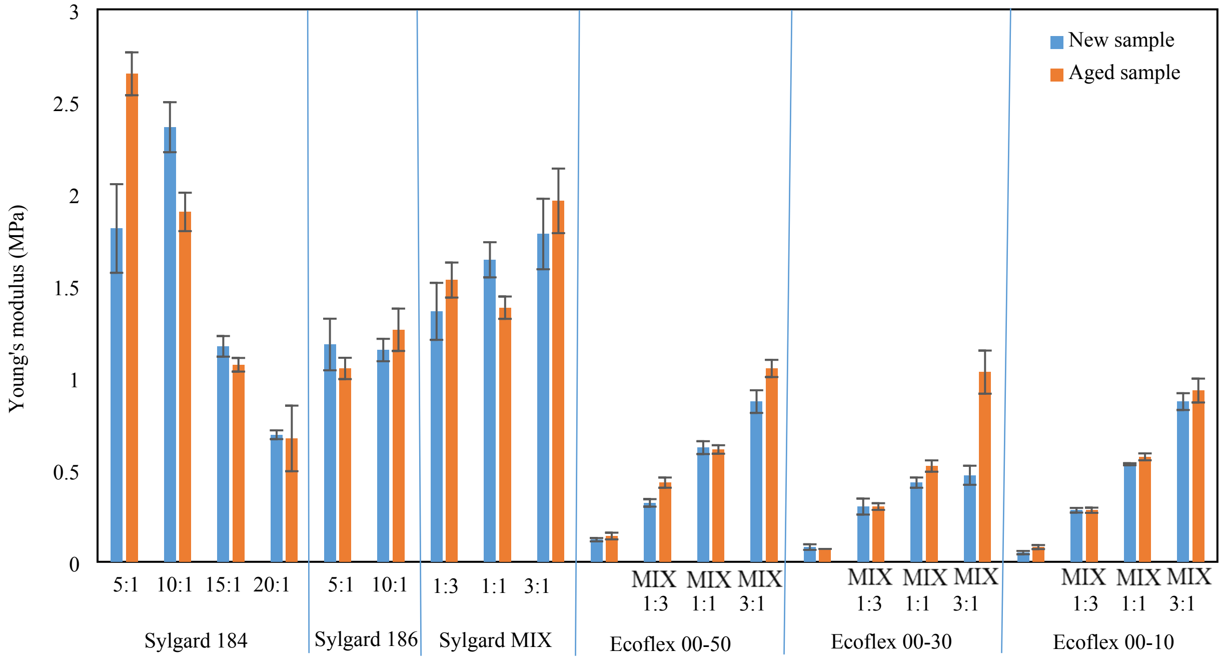

Figure S20: Y of the new and aged samples.

Table S5: Breakdown strength (E_{BD})

Formulation	E_{BD} (V/µm))	E_{BD} (V/µm) after 6 months
Sylgard 184 5:1	123±9	117±8
Sylgard 184	**100±17**	**98±40**
Sylgard 184 15:1	94±3	97±4
Sylgard 184 20:1	81±5.5	79±8
Sylgard 186 5:1	82±12	109±10
Sylgard 186	**87±7**	**112±9**
Sylgard MIX 1:3	99±9	101±14
Sylgard MIX 1:1	92±12	125±13
Sylgard MIX 3:1	89±12	123±9
Ecoflex 00-50	**62±5**	**46±5**
Eco MIX 00-50 1:3	72±3.5	81±5
Eco MIX 00-50 1:1	88±5	91±8
Eco MIX 00-50 3:1	92±7	109±3
Ecoflex 00-30	**59±2**	**35±3**
Eco MIX 00-30 1:3	66±4	76±5.5
Eco MIX 00-30 1:1	78±4	90±8
Eco MIX 00-30 3:1	87±17	114±11
Ecoflex 00-10	**38±14**	**-**
Eco MIX 00-10 1:3	69±5	70±5
Eco MIX 00-10 1:1	94±8.2	92±7
Eco MIX 00-10 3:1	94±5	111±9
Elastosil	**106±8.3**	**-**

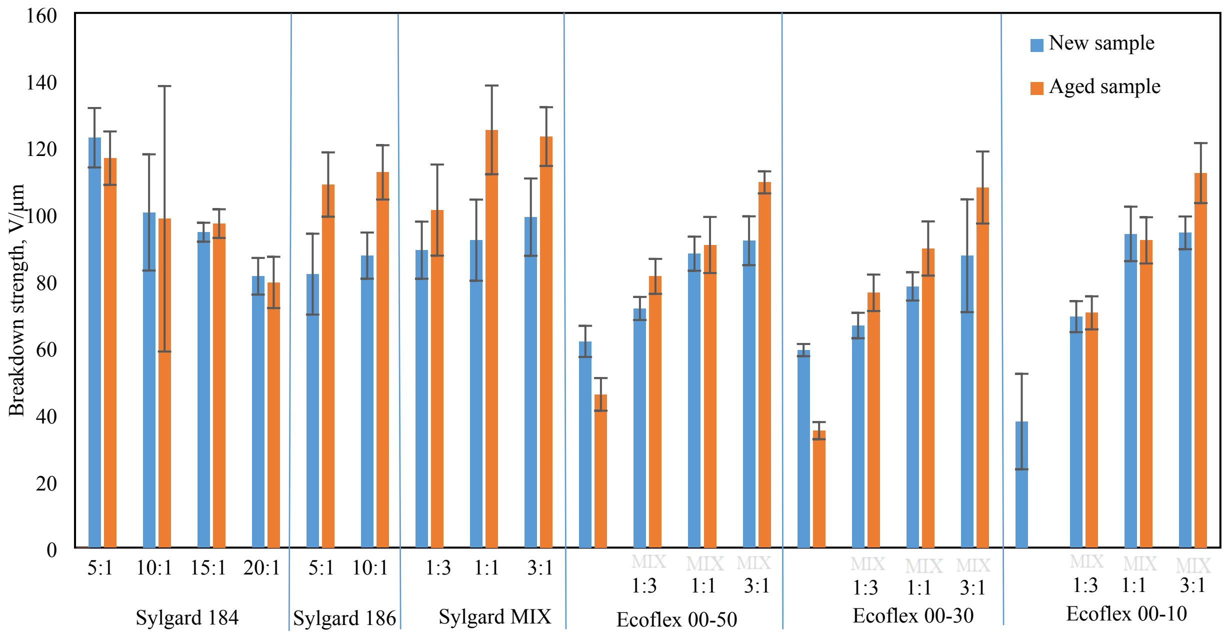

Figure S21: Comparison of the E_{BD} strength of new samples and 6 months aged samples

Table S6: Shore hardness (S_{hard} [A], S_{hard} [00])

Formulation	**S_{hard} [A]**	**S_{hard} [A] after 6 months**	**S_{hard} [00]**	**S_{hard} [00] after 6 months**
Sylgard 184 5:1	39	23	75	73
Sylgard 184	**38**	**21**	**80**	**73**
Sylgard 184 15:1	28	21	72	75
Sylgard 184 20:1	18	20	68	70
Sylgard 186 5:1	26	22	75	76
Sylgard 186	**24**	**21**	**73**	**77**
Sylgard MIX 1:3	38	22	75	76
Sylgard MIX 1:1	36	22	74	72
Sylgard MIX 3:1	34	21	73	76
Ecoflex 00-50	**-**	**-**	**35**	**34**
Eco MIX 00-50 1:3	14	13	62	57
Eco MIX 00-50 1:1	23	17	67	65
Eco MIX 00-50 3:1	28	19	73	70
Ecoflex 00-30	**-**	**-**	**23**	**25**
Eco MIX 00-30 1:3	10	10	53	55
Eco MIX 00-30 1:1	18	17	64	64
Eco MIX 00-30 3:1	28	20	73	70
Ecoflex 00-10	**-**	**-**	**11**	**17**
Eco MIX 00-10 1:3	10	9	54	53
Eco MIX 00-10 1:1	24	19	66	60
Eco MIX 00-10 3:1	32	20	73	75

Table S7: Transmittance (T) of samples @ 550 nm after 6 months

Formulation	**T @ 0% pre-stretch**	**T @ 20% pre-stretch**	**T @ 50% pre-stretch**	**T @ 100% pre-stretch**
Sylgard 184 5:1	93	93	-	
Sylgard 184	**94**	**93**	**92**	**-**
Sylgard 184 15:1	93	91	85	-
Sylgard 184 20:1	94.1	94	89	
Sylgard 186 5:1	92.3	91	90	-
Sylgard 186	**92**	**92**	**93**	**-**
Sylgard MIX 1:3	92	93	89.2	-
Sylgard MIX 1:1	92.5	91	92	-
Sylgard MIX 3:1	93	93	90	-
Ecoflex 00-50	**75**	**67**	**56**	**41.4**
Eco MIX 00-50 1:3	82	73	66	61
Eco MIX 00-50 1:1	84	78	69	-
Eco MIX 00-50 3:1	87.4	85	83	-
Ecoflex 00-30	**79.4**	**69**	**64**	**50.4**
Eco MIX 00-30 1:3	78	72	63.5	62.4
Eco MIX 00-30 1:1	85	79.3	76	76.4
Eco MIX 00-30 3:1	89	86	85.3	-
Ecoflex 00-10	**69**	**48**	**42**	**31**
Eco MIX 00-10 1:3	80.14	67.3	56	-
Eco MIX 00-10 1:1	82.2	73.3	66.4	-
Eco MIX 00-10 3:1	89.3	84	81	-

S4. Strain energy calculations:

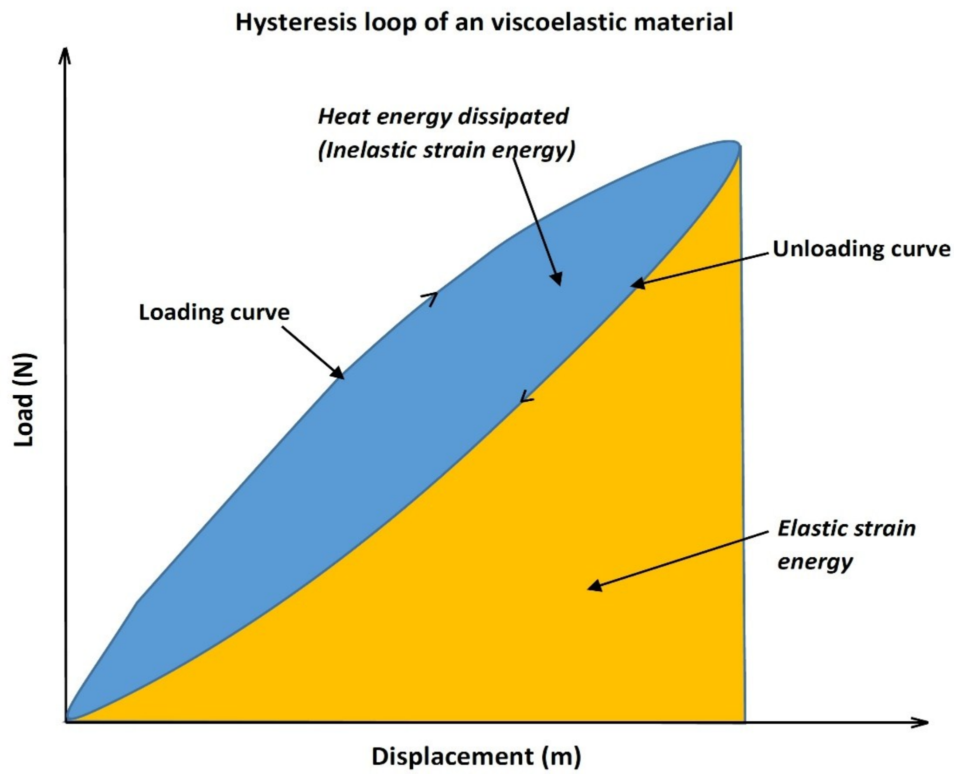

Figure S22: Hysterisis of an elastomer

The strain energy function of the elastomer is obtained by dividing the area under the unloading curve (elastic strain energy) with the area under the loading curve (total energy). For an ideal elastomer this ratio is 1.

Table S8: Strain energy function (φ) calculations for all the formulations from their respective hysterisis curves

Formulation	Total energy (J)	Elastic strain energy (J)	φ = elastic energy/total energy
Sylgard 184 5:1	0.058	0.051	0.88
Sylgard 184	**0.036**	**0.029**	**0.81**
Sylgard 184 15:1	0.022	0.018	0.82
Sylgard 184 20:1	0.013	0.011	0.85
Sylgard 186 5:1	0.022	0.012	0.55
Sylgard 186	**0.021**	**0.018**	**0.86**
Sylgard MIX 1:3	0.026	0.022	0.85
Sylgard MIX 1:1	0.028	0.024	0.86
Sylgard MIX 3:1	0.036	0.031	0.86
Ecoflex 00-50	**0.002**	**0.0017**	**0.85**
Eco MIX 00-50 1:3	0.0123	0.0112	0.91
Eco MIX 00-50 1:1	0.0101	0.0095	0.94
Eco MIX 00-50 3:1	0.0059	0.0056	0.95
Ecoflex 00-30	**0.0012**	**0.001**	**0.83**
Eco MIX 00-30 1:3	0.0041	0.0039	0.95
Eco MIX 00-30 1:1	0.0086	0.0081	0.94
Eco MIX 00-30 3:1	0.0134	0.0124	0.93
Ecoflex 00-10	**0.0014**	**0.001**	**0.71**
Eco MIX 00-10 1:3	0.0046	0.0042	0.91
Eco MIX 00-10 1:1	0.0104	0.0097	0.93
Eco MIX 00-10 3:1	0.016	0.015	0.94
Elastosil	0.0146	0.0141	**0.97**

Table S9: Figure of merit (F_{om}(DEA) and F_{om}(DEG))

Formulation	Strain energy function (φ)	F_{om} (DEA)	F_{om} (DEA) normalized with Sylgard 184	F_{om} (DEG)	F_{om} (DEG) normalized with Sylgard 184
Sylgard 184 5:1	0.88	0.72	2.1	244960.9	1.4

Sylgard 184	**0.81**	**0.34**	1	169351.9	1
Sylgard 184 15:1	0.82	0.63	1.8	152582.6	0.9
Sylgard 184 20:1	0.85	0.77	2.3	105883	0.6
Sylgard 186 5:1	0.55	0.52	1.5	189341.7	1.1
Sylgard 186	**0.86**	**0.57**	1.7	132413.5	0.8
Sylgard MIX 1:3	0.85	0.53	1.5	144325.3	0.9
Sylgard MIX 1:1	0.86	0.46	1.4	143715.8	0.85
Sylgard MIX 3:1	0.86	0.48	1.4	166417.6	0.98
Ecoflex 00-50	**0.85**	**3.81**	11.1	74631.5	0.4
Eco MIX 00-50 1:3	0.91	1.82	5.3	101260	0.6
Eco MIX 00-50 1:1	0.94	1.13	3.3	120299.7	0.7
Eco MIX 00-50 3:1	0.95	0.82	2.4	128915.6	0.8
Ecoflex 00-30	**0.83**	**3.47**	10	69678.3	0.4
Eco MIX 00-30 1:3	0.95	1.43	4.2	75399	0.45
Eco MIX 00-30 1:1	0.94	1.33	3.9	94512.4	0.56
Eco MIX 00-30 3:1	0.93	1.38	4	123329	0.73
Ecoflex 00-10	**0.71**	**2.61**	7.6	30598.6	0.18
Eco MIX 00-10 1:3	0.91	1.43	4.2	78372.1	0.5
Eco MIX 00-10 1:1	0.93	1.34	4	120319.6	0.7
Eco MIX 00-10 3:1	0.94	0.79	2.3	126778	0.75
Elastosil	**0.97**	**0.82**	2.4	167097.9	0.99

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(2) Silau, H.; Stabell, N. B.; Petersen, F. R.; Pham, M.; Yu, L.; Skov, A. L. Weibull Analysis of Electrical Breakdown Strength as an Effective Means of Evaluating Elastomer Thin Film Quality. **2018**, *1800241*, 1–8. https://doi.org/10.1002/adem.201800241.

(3) Technical Data Sheet: SYLGARD 184 Silicone Elastomer https://consumer.dow.com/content/dam/dcc/documents/en-us/productdatasheet/11/11-31/11-3184-sylgard-184-elastomer.pdf?iframe=true.