

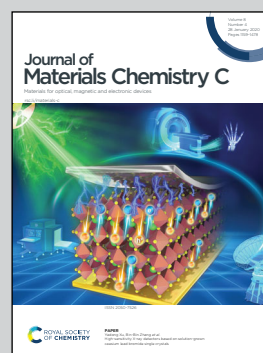


**Showcasing a study on blending commercial silicone formulations with each other to make reliable soft networks with mechanical integrity, by the Danish Polymer Centre in Technical University of Denmark, Kgs. Lyngby 2800, Denmark.**

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

On a research level, commercial silicone elastomers are widely used for fabricating stretchable devices but these commercial silicone elastomer formulations often do not encompass all of the properties necessary to function effectively as stretchable transducers. Commercial silicones are blended with each other in various proportions, to make the blends most suitable for fabricating stretchable transducers. The mechanical and electrical properties, and ageing processes of these blends are thoroughly studied.

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See Anne Ladegaard Skov *et al.*,  
*J. Mater. Chem. C*, 2020, **8**, 1273.



Cite this: *J. Mater. Chem. C*, 2020, **8**, 1273

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

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Elastomers for fabricating soft and stretchable transducers primarily require high elongation at break, high dielectric permittivity, high breakdown strength and low leakage current. Commercial silicone elastomer formulations often do not encompass all of the properties necessary to function effectively as stretchable transducers, but they are nevertheless used out of familiarity. On a research level, Sylgard 184, Sylgard 186, Ecoflex 00-10, Ecoflex 00-30 and Ecoflex 00-50 are widely used for fabricating stretchable devices. We blend these commercial silicones with each other in various proportions, to make the blends most suitable for fabricating specific types of transducers. Furthermore, the properties of these blends, such as ultimate stress and strain, Young's modulus, dielectric permittivity, breakdown strength, viscosity, leakage current and optical transmittance, are investigated and mapped to identify those exhibiting the best-suited properties for fabricating soft and stretchable transducers. The elastomers obtained using the blending methods illustrated herein could act as a starting point for conceptualizing the feasibility of a product on a research level.

Received 14th September 2019,  
Accepted 13th November 2019

DOI: 10.1039/c9tc05072h

rsc.li/materials-c

## 1. Introduction

The evolution of flexible and stretchable transducers as viable alternatives to conventional electromagnetic transducers has sparked unprecedented interest in transitioning from hard to soft equipment.<sup>1,2</sup> Soft, stretchable and lightweight transducers that can generate motion and withstand wear and tear<sup>3,4</sup> are beneficial for developing futuristic applications, for example soft robots,<sup>5</sup> lifelike prosthetics,<sup>6,7</sup> stretchable pressure sensors<sup>8</sup> and intrinsic wearable biomedical devices.<sup>9</sup> Sylgard 184, supplied by Dow, is a very popular elastomer for fabricating such devices, mainly due to its low viscosity and ease of processing. Sylgard 184 has been used to develop a wide range of products, such as shape memory composites,<sup>10</sup> compliant graphite electrodes,<sup>11</sup> micro-fluidic devices,<sup>12</sup> super-stretch strain sensors,<sup>13</sup> micro contact pressure sensors<sup>14</sup> and tactile sensors.<sup>15</sup> Though Sylgard 184 sets the benchmark for commercial silicone elastomers, our measurements show that it has limited extensibility (135%), a relatively high Young's modulus as for most commercial silicone elastomer compositions (2.4 MPa), relatively high leakage current ( $\approx 10^{-10}$  A) and a dielectric permittivity of  $\sim 3.1$  (1 Hz), which is not optimal for

developing stretchable transducers, especially for high-voltage applications. Hence, in pursuit of better alternatives to Sylgard 184, we analyse the properties of Sylgard 186 and Ecoflex by Smooth-On, both of which have higher extensibility and a lower Young's modulus compared to Sylgard 184.<sup>16,17</sup> Lately, Ecoflex has been used widely for fabricating soft, elastic, wearable pressure sensors/electronics.<sup>16,18</sup> Exploiting the synergy between two formulations with contrasting properties is a simple way to formulate a silicone elastomer with desired properties when formulation expertise is not available.<sup>17,19</sup> In previous studies by Russo *et al.*<sup>19</sup> and Park *et al.*,<sup>17</sup> an elastomer blend (Dragon Skin 00 20 + Sylgard 184<sup>19</sup> and Dragon Skin 10 Slow + Sylgard 184<sup>17</sup>) was found to possess higher extensibility, better tear resistance and plasma bonding ability compared to pure Sylgard 184. In this work, five commercial silicone formulations (Sylgard 184 and 186, and Ecoflex 00-50, 00-30 and 00-10) are blended with each other. Some formulations (Sylgard 184 and 186) are also mixed at different stoichiometric ratios, in order to optimise their mechanical and electrical properties. Optically transparent formulations have been chosen, since optical transparency is extensively sought, especially for optics and “invisible” wearable sensors.<sup>20</sup> The blends' properties are mapped and compared to Sylgard 184, which is the current benchmark for stretchable electronics, as well as to Elastosil Film 2030 250/X, which recently has become a new benchmark for dielectric elastomer films, due to their commercial availability. Figures of merit for a dielectric elastomer

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† Electronic supplementary information (ESI) available. See DOI: 10.1039/c9tc05072h

actuator ( $F_{\text{om}}(\text{DEA})$ )<sup>20</sup> and a dielectric elastomer generator ( $F_{\text{om}}(\text{DEG})$ ),<sup>21</sup> respectively, are calculated to assess elastomer performance in the two respective operational modes.

## 2. Materials and sample preparation

Five different two-part (A and B) commercial Pt-curing silicone elastomers are used herein. Sylgard elastomers are purchased from Dow, Germany, Ecoflex elastomers purchased from FormX, Netherlands and Elastosil Films 2030 250/X, where, “X” denotes the thickness of the film (100  $\mu\text{m}$  and 200  $\mu\text{m}$  in thickness), provided by Wacker Chemie, Germany. From now on Elastosil Film 2030 250/X, will be denoted as Elastosil.

Throughout the manuscript the term ‘blend’ will refer to formulations obtained by combining two different commercial silicone systems. Sample compositions are shown in Table 1. Parts A and B of every silicone formulation are weighed according to the mixing ratio and mixed in a SpeedMixer DAC 150 FVZ-K for 2 minutes at 2000 rpm. Then, this well mixed formulation is blended with another well mixed formulation in the ratios specified in Table 1 for another 2 minutes at 2000 rpm, after which the blends are used to prepare samples. The manufacturer's recommended mixing ratio for Sylgard formulations is 10:1 (part A:part B) and for Ecoflex formulations the ratio is 1:1 (part A:part B). Films of different thicknesses are made for various measurements. To make these film samples, the formulations are doctor-bladed on polyester support film, provided by Pütz GmbH + Co. Folien KG, Germany. The blade gaps are 600  $\mu\text{m}$ , 400  $\mu\text{m}$  and 200  $\mu\text{m}$ , respectively. After curing, film thicknesses are  $450 \pm 45 \mu\text{m}$ ,  $300 \pm 30 \mu\text{m}$  and  $100 \pm 20 \mu\text{m}$ , respectively.<sup>22</sup> The precise sample thickness is evaluated using optical microscope. Moreover, each composition

is poured into a metal mold with 1 mm thick spacer and a metal cup in order to obtain samples with thicknesses of  $1 \pm 0.2 \text{ mm}$  and 6.4 mm (or larger), respectively. All formulations are cured at 80 °C for 2 hours, and measurements are carried out in ambient conditions. The resulting elastomers do not adhere to any of the used supports, allowing for an easy peeling off. Swelling tests of prepared samples are performed and gel fraction is calculated as described in ESI.† Gel fractions (Table 1) show that all prepared samples are well cross-linked. For measurements on aged samples, the specimens are stored for 6 months at room temperature, with no exposure to sunlight. The properties of the stored samples are then re-measured and compared to the previous results. The methods for measuring the mechanical and electrical properties of the compositions are described in the ESI.†

## 3. Results and discussion

The properties of the different elastomers, *i.e.* elongation at break ( $\epsilon_{\text{max}}$  (%)), ultimate tensile strength ( $\sigma_{\text{max}}$  (MPa)), viscosity ( $\eta$  (Pa s)), optical transmittance ( $T$  (%)), breakdown strength ( $E_{\text{BD}}$  (V  $\mu\text{m}^{-1}$ )), leakage current ( $i_{\text{leak}}$  (A)) and  $i_{\text{leak}}$  sensitivity to temperature ( $k$  (°C<sup>-1</sup>)), are plotted as functions of the Young's modulus ( $Y$  (MPa)) and shown in Fig. 1. The properties are plotted as functions of  $Y$ , since usually it is the primary specification for applications. All of the properties are also tabulated (Table 2). The  $F_{\text{om}}(\text{DEA})$  and  $F_{\text{om}}(\text{DEG})$  are plotted in Fig. 2. Mechanical and electrical properties are discussed further below. Plots for other properties, namely Shore hardness ( $S_{\text{hard}}$  [A] and  $S_{\text{hard}}$  [00]), dielectric permittivity ( $\epsilon'$ ) and  $T$  over the entire visible spectrum, are provided in the ESI.†

### 3.1. Mechanical properties

Sylgard 184 has the highest  $Y = 2.4 \text{ MPa}$  and Ecoflex 00-10 the lowest  $Y = 0.05 \text{ MPa}$ . Generally, for silicone elastomers, as the  $Y$

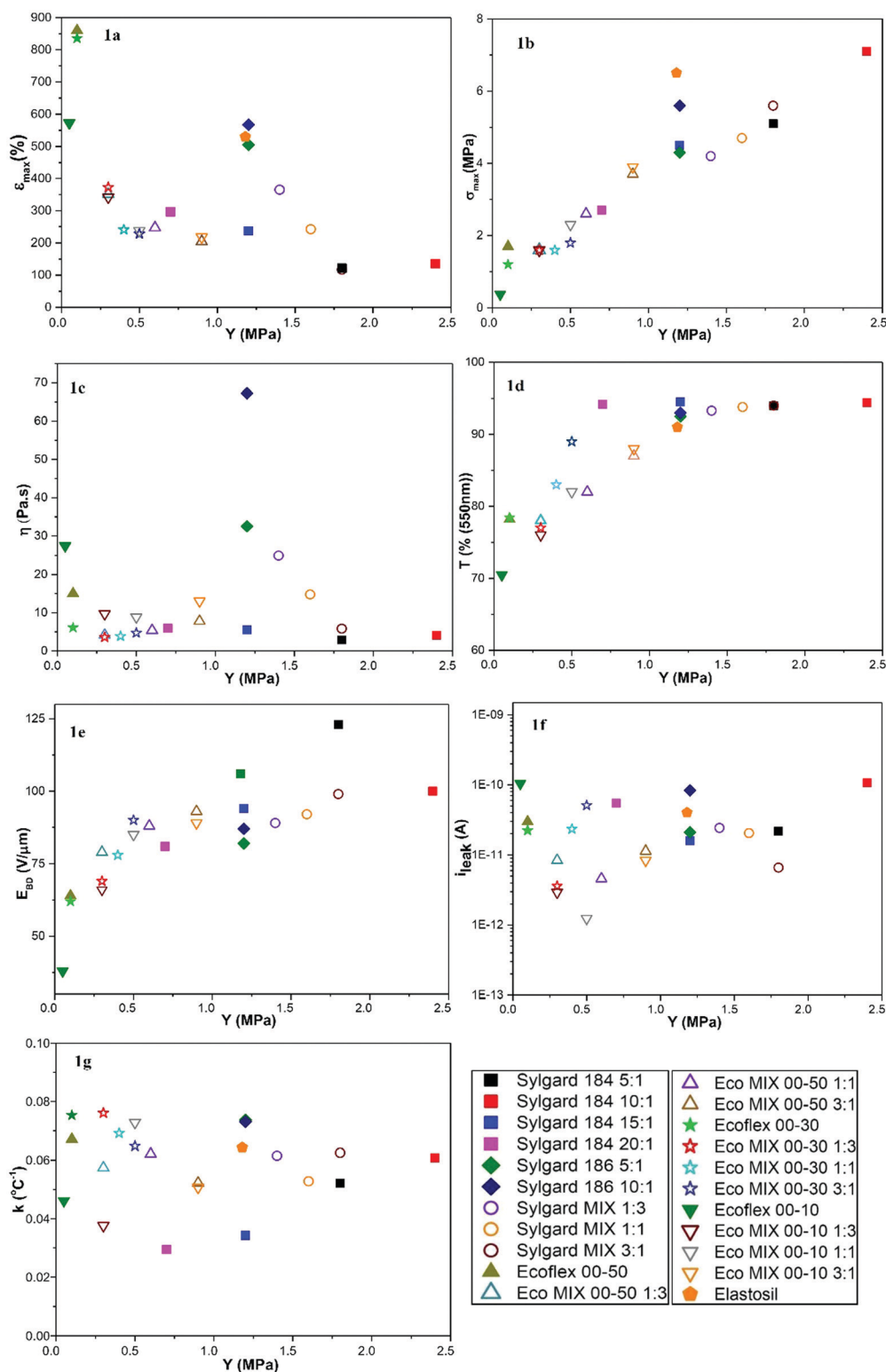
**Table 1** Sample names with their corresponding mixing ratios (part A and part B), blending ratios with other formulations and gel fraction of all formulations

| No. | Sample name              | Part I <sup>a</sup> | Ratio | Part II <sup>a</sup> | Ratio | Gel fraction, % |
|-----|--------------------------|---------------------|-------|----------------------|-------|-----------------|
| 1   | Sylgard 184 5:1          | Sylgard 184 (5:1)   | —     | —                    | —     | 95.1            |
| 2   | Sylgard 184              | Sylgard 184 (10:1)  | —     | —                    | —     | 95.4            |
| 3   | Sylgard 184 15:1         | Sylgard 184 (15:1)  | —     | —                    | —     | 93.2            |
| 4   | Sylgard 184 20:1         | Sylgard 184 (20:1)  | —     | —                    | —     | 91.1            |
| 5   | Sylgard 186 5:1          | Sylgard 186 (5:1)   | —     | —                    | —     | 95.8            |
| 6   | Sylgard 186 <sup>b</sup> | Sylgard 186 (10:1)  | —     | —                    | —     | 95.3            |
| 7   | Sylgard MIX 1:3          | Sylgard 184 (10:1)  | 1     | Sylgard 186 (10:1)   | 3     | 96.1            |
| 8   | Sylgard MIX 1:1          | Sylgard 184 (10:1)  | 1     | Sylgard 186 (10:1)   | 1     | 95.8            |
| 9   | Sylgard MIX 3:1          | Sylgard 184 (10:1)  | 3     | Sylgard 186 (10:1)   | 1     | 95.7            |
| 10  | Ecoflex 00-50            | Ecoflex 00-50 (1:1) | —     | —                    | —     | 54.3            |
| 11  | Eco MIX 00-50 1:3        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-50 (1:1)  | 3     | 66.5            |
| 12  | Eco MIX 00-50 1:1        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-50 (1:1)  | 1     | 76.0            |
| 13  | Eco MIX 00-50 3:1        | Sylgard 184 (10:1)  | 3     | Ecoflex 00-50 (1:1)  | 1     | 85.6            |
| 14  | Ecoflex 00-30            | Ecoflex 00-30 (1:1) | —     | —                    | —     | 43.4            |
| 15  | Eco MIX 00-30 1:3        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-30 (1:1)  | 3     | 57.2            |
| 16  | Eco MIX 00-30 1:1        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-30 (1:1)  | 1     | 69.6            |
| 17  | Eco MIX 00-30 3:1        | Sylgard 184 (10:1)  | 3     | Ecoflex 00-30 (1:1)  | 1     | 82.1            |
| 18  | Ecoflex 00-10            | Ecoflex 00-10 (1:1) | —     | —                    | —     | —               |
| 19  | Eco MIX 00-10 1:3        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-10 (1:1)  | 3     | 60.7            |
| 20  | Eco MIX 00-10 1:1        | Sylgard 184 (10:1)  | 1     | Ecoflex 00-10 (1:1)  | 1     | 72.7            |
| 21  | Eco MIX 00-10 3:1        | Sylgard 184 (10:1)  | 3     | Ecoflex 00-10 (1:1)  | 1     | 83.1            |

<sup>a</sup> Mixing weight ratios for part A and part B (A:B) of the respective silicone kits (in bracket). <sup>b</sup> The formulations of Sylgard 186 15:1 and 20:1 are not tested as these mixtures did not produce free-standing elastomers.

increases,  $\sigma_{\max}$  increases and  $\varepsilon_{\max}$  decreases. The low  $Y$  of Ecoflex 00-50, 00-30 and 00-10 (0.05 to 0.1 MPa) can be increased by blending with Sylgard 184, as shown by the  $Y$

values of Eco MIX 00-50, Eco MIX 00-30 and Eco MIX 00-10 (0.3 to 0.9 MPa). For soft and stretchable devices, extensibility (Fig. 1a) is a prominent parameter.<sup>23</sup> Of all the formulations,



**Fig. 1** Various properties of the investigated silicone elastomers, plotted against the Young's modulus ( $Y$ ). (a) Maximum strain. (b) Ultimate stress. (c) Viscosity@shear rate  $0.1 \text{ s}^{-1}$ . (d) Transmittance at 0% pre-stretch at 550 nm. (e) Electrical breakdown strength. (f)  $i_{\text{leak}}$  (applied electric field of  $5 \text{ V } \mu\text{m}^{-1}$ ) @  $25^{\circ}\text{C}$  and (g) sensitivity of  $i_{\text{leak}}$  to temperature change.

Table 2 Properties of the different silicone elastomer formulations

| Sample name          | Mechanical properties |                       |                       |                        |                                               |                       |                        | Electrical properties |                     |                                         |                                                    |                                                  |
|----------------------|-----------------------|-----------------------|-----------------------|------------------------|-----------------------------------------------|-----------------------|------------------------|-----------------------|---------------------|-----------------------------------------|----------------------------------------------------|--------------------------------------------------|
|                      | Y (MPa)               | $\epsilon_{\max}$ (%) | $\sigma_{\max}$ (MPa) | $\sigma_{100\%}$ (MPa) | $\eta$ @shear rate 0.1 s <sup>-1</sup> (Pa s) | $S_{\text{hard}}$ [A] | $S_{\text{hard}}$ [00] | $\epsilon'$ @1 Hz     | $\tan \delta$ @1 Hz | $E_{\text{BD}}$ (V $\mu\text{m}^{-1}$ ) | $i_{\text{leak}}$ @ 25 °C ( $\times 10^{-11}$ , A) | $\sigma_{\text{AC}}$ @1 Hz (S cm <sup>-1</sup> ) |
| Sylgard 184 5:1      | 1.8                   | 122                   | 5.1                   | 3.42                   | 2.9                                           | 39                    | 75                     | 3.22                  | 0.012               | 123                                     | 2.2                                                | $1.9 \times 10^{-14}$                            |
| <b>Sylgard 184</b>   | <b>2.4</b>            | <b>135</b>            | <b>7.1</b>            | <b>3.86</b>            | <b>4</b>                                      | <b>38</b>             | <b>80</b>              | <b>3.1</b>            | <b>0.002</b>        | <b>100</b>                              | <b>10.6</b>                                        | <b><math>3.5 \times 10^{-15}</math></b>          |
| Sylgard 184 15:1     | 1.2                   | 237                   | 4.5                   | 0.77                   | 5.5                                           | 28                    | 72                     | 3.2                   | 0.008               | 94                                      | 1.6                                                | $1.3 \times 10^{-14}$                            |
| Sylgard 184 20:1     | 0.7                   | 296                   | 2.7                   | 0.39                   | 6                                             | 18                    | 68                     | 3.1                   | 0.013               | 81                                      | 5.5                                                | $2.2 \times 10^{-14}$                            |
| Sylgard 186 5:1      | 1.2                   | 505                   | 4.3                   | 0.49                   | 32.5 <sup>a</sup>                             | 26                    | 75                     | 3.5                   | 0.002               | 82                                      | 2.1                                                | $4 \times 10^{-15}$                              |
| <b>Sylgard 186</b>   | <b>1.2</b>            | <b>567</b>            | <b>5.6</b>            | <b>0.48</b>            | <b>67.3<sup>a</sup></b>                       | <b>24</b>             | <b>73</b>              | <b>3.4</b>            | <b>0.001</b>        | <b>87</b>                               | <b>8.3</b>                                         | <b><math>1.6 \times 10^{-15}</math></b>          |
| Sylgard MIX 1:3      | 1.4                   | 365                   | 4.2                   | 0.60                   | 24.9 <sup>a</sup>                             | 38                    | 75                     | 3.5                   | 0.002               | 89                                      | 2.4                                                | $2.3 \times 10^{-15}$                            |
| Sylgard MIX 1:1      | 1.6                   | 242                   | 4.7                   | 1.12                   | 14.8 <sup>a</sup>                             | 36                    | 74                     | 3.3                   | 0.002               | 92                                      | 2                                                  | $3.1 \times 10^{-15}$                            |
| Sylgard MIX 3:1      | 1.8                   | 117                   | 5.6                   | 2.45                   | 5.8 <sup>a</sup>                              | 34                    | 73                     | 3.3                   | 0.003               | 99                                      | 6.6                                                | $4.4 \times 10^{-15}$                            |
| <b>Ecoflex 00-50</b> | <b>0.1</b>            | <b>860</b>            | <b>1.7</b>            | <b>0.07</b>            | <b>15</b>                                     | — <sup>b</sup>        | <b>35</b>              | <b>3.5</b>            | <b>0.056</b>        | <b>62</b>                               | <b>3</b>                                           | <b><math>1 \times 10^{-13}</math></b>            |
| Eco MIX 00-50 1:3    | 0.3                   | 351                   | 1.6                   | 0.26                   | 4.2                                           | 14                    | 62                     | 3.3                   | 0.035               | 72                                      | 8.4                                                | $6.1 \times 10^{-14}$                            |
| Eco MIX 00-50 1:1    | 0.6                   | 247                   | 2.6                   | 0.62                   | 5.4                                           | 23                    | 67                     | 3.3                   | 0.027               | 88                                      | 4.6                                                | $4.5 \times 10^{-14}$                            |
| Eco MIX 00-50 3:1    | 0.9                   | 204                   | 3.7                   | 0.98                   | 7.8                                           | 28                    | 73                     | 3.2                   | 0.024               | 92                                      | 1.2                                                | $4 \times 10^{-14}$                              |
| <b>Ecoflex 00-30</b> | <b>0.1</b>            | <b>835</b>            | <b>1.2</b>            | <b>0.05</b>            | <b>6.1</b>                                    | — <sup>b</sup>        | <b>23</b>              | <b>3.4</b>            | <b>0.040</b>        | <b>59</b>                               | <b>2.2</b>                                         | <b><math>8 \times 10^{-14}</math></b>            |
| Eco MIX 00-30 1:3    | 0.3                   | 373                   | 1.6                   | 0.23                   | 3.6                                           | 10                    | 53                     | 3.4                   | 0.035               | 66                                      | 0.4                                                | $6 \times 10^{-14}$                              |
| Eco MIX 00-30 1:1    | 0.4                   | 241                   | 1.6                   | 0.36                   | 3.9                                           | 18                    | 64                     | 3.3                   | 0.022               | 78                                      | 2.4                                                | $4.2 \times 10^{-14}$                            |
| Eco MIX 00-30 3:1    | 0.5                   | 228                   | 1.8                   | 0.43                   | 4.7                                           | 28                    | 73                     | 3.2                   | 0.016               | 87                                      | 5.1                                                | $2.7 \times 10^{-14}$                            |
| <b>Ecoflex 00-10</b> | <b>0.05</b>           | <b>573</b>            | <b>0.37</b>           | <b>0.02</b>            | <b>27.5</b>                                   | — <sup>b</sup>        | <b>11</b>              | <b>3.4</b>            | <b>0.090</b>        | <b>38</b>                               | <b>10.5</b>                                        | <b><math>1.6 \times 10^{-13}</math></b>          |
| Eco MIX 00-10 1:3    | 0.3                   | 342                   | 1.6                   | 0.28                   | 9.7                                           | 10                    | 54                     | 3.7                   | 0.049               | 69                                      | 0.3                                                | $1 \times 10^{-13}$                              |
| Eco MIX 00-10 1:1    | 0.5                   | 238                   | 2.3                   | 0.63                   | 8.9                                           | 24                    | 66                     | 3.5                   | 0.061               | 94                                      | 0.12                                               | $1 \times 10^{-13}$                              |
| Eco MIX 00-10 3:1    | 0.9                   | 218                   | 3.9                   | 1.01                   | 13.1                                          | 32                    | 73                     | 3.4                   | 0.027               | 93                                      | 0.8                                                | $5 \times 10^{-14}$                              |
| Elastosil            | 1.18                  | 530                   | 6.5                   | 0.68                   | —                                             | —                     | —                      | 3.26                  | 0.001               | 106                                     | 4                                                  | $1.1 \times 10^{-14}$                            |

<sup>a</sup> Measured at a shear rate of 0.524 s<sup>-1</sup>. <sup>b</sup> Too soft to be measured by this technique.

Sylgard 184 5:1 has the lowest extensibility. By varying the mixing ratio of Sylgard 184, from 5:1 to 20:1, the  $\epsilon_{\max}$  is doubled (122% to 296%). Sylgard 186 shows a high  $\epsilon_{\max}$  of  $\sim 568\%$ . A blend of Sylgard 184 and 186 (*i.e.* Sylgard MIX 1:3) produces a higher  $\epsilon_{\max}$  of 365%, which is a significant improvement compared to Sylgard 184. Ecoflex 00-50 and Ecoflex 00-30 have a high  $\epsilon_{\max}$  of  $\sim 800\%$ , and their blends with Sylgard 184 (Eco MIX 00-50 and Eco MIX 00-30) show a  $\epsilon_{\max}$  of  $\sim 360\%$ . The addition of a soft formulation to a hard one results in a composite with intermediate properties as expected. Sylgard 184 has the highest  $S_{\text{hard}}$  [00] compared to the formulations tested (ESI†). The Sylgard 186 has a lower  $S_{\text{hard}}$  [00] compared to Sylgard 184, whereas their blends (Sylgard MIX) show intermediate  $S_{\text{hard}}$  [00]. Similarly, when the softer Ecoflex 00-10 with low  $S_{\text{hard}}$  [00] is mixed with Sylgard 184, the resulting Eco MIX formulations have a much higher  $S_{\text{hard}}$  [00] compared to the pure Ecoflex 00-10.

### 3.2. Viscosity and pot life

Viscosities are plotted in Fig. 1c and tabulated in Table 2. Formulations with low viscosity are preferred, as they result in easier processing. Generally, Ecoflex 00-50, Ecoflex 00-30, Ecoflex 00-10, Eco MIX 00-50, Eco MIX 00-30 and Eco MIX 00-10 possess low  $\eta$  compared to Sylgard 186 and Sylgard MIX, whilst Sylgard 186 has a higher premix  $\eta$  compared to Sylgard 184. Formulations with a  $\eta \sim 5\text{--}10$  Pa s are relatively easy to process, while higher  $\eta$  formulations typically require extra solvent or flow control agents, which in turn might compromise dielectric and/or mechanical properties.<sup>22</sup> The pot life of Sylgard 184 and 186 is generally more than 1 h at room temperature. Ecoflex 00-50, 00-30 and 00-10 have a pot life  $\sim 20$  min only, which leaves very little time for processing. Interestingly, the blends of

Ecoflex and Sylgard 184 have a pot life of  $\sim 1$  h, which is a significant improvement over the original Ecoflex formulations. The pot life data for all formulations are available in the ESI.†

### 3.3. Optical transmittance

$T$  values (at 550 nm<sup>24</sup> and 0% pre-stretch) are plotted in Fig. 1d. The data indicate that Sylgard 184, Sylgard 186, Sylgard MIX and Elastosil have better optical transparency compared to all of the Ecoflex and Eco MIX samples. It is noteworthy that Ecoflex shows a higher  $\epsilon'$  and a low  $T$  compared to Sylgard 184, which indicates the presence of substances other than organosilicon compounds. Sylgard 186 has a slightly lower  $T$  compared to Sylgard 184. If one needs transparent elastomers, then Ecoflex should be avoided or used in minimal amounts in blends. The  $T$  of the film samples varies over the entire visible spectrum (380–740 nm) and is also affected by pre-stretching (data in ESI†): as the pre-stretch increases, the  $T$  of the films decreases (data in ESI†).

### 3.4. Electrical properties

Of all the tested commercial formulations, Sylgard 184 shows the highest  $E_{\text{BD}}$  of 100 V  $\mu\text{m}^{-1}$  (Fig. 1e). By varying the mixing ratio of Sylgard 184 from 10:1 to 5:1, the  $E_{\text{BD}}$  increases to 123 V  $\mu\text{m}^{-1}$ . Generally, silicone formulations with a higher  $Y$  show a higher  $E_{\text{BD}}$  as well. By changing the mixing ratio of Sylgard 186 from 10:1 to 5:1, a higher  $\epsilon'$  of 3.5 can be achieved (ESI†). Ecoflex 00-50, 00-30 and 00-10 have a lower  $E_{\text{BD}}$  compared to Sylgard 184 and 186, and the blends resulting from the amalgamation of these formulations, namely Eco MIX 00-50, Eco MIX 00-30 and Eco MIX 00-10, benefit from being moderately reinforced by the relatively high  $\epsilon'$  of Ecoflex 00-50, Ecoflex 00-30 and Ecoflex 00-10, and the high  $E_{\text{BD}}$  of Sylgard 184.

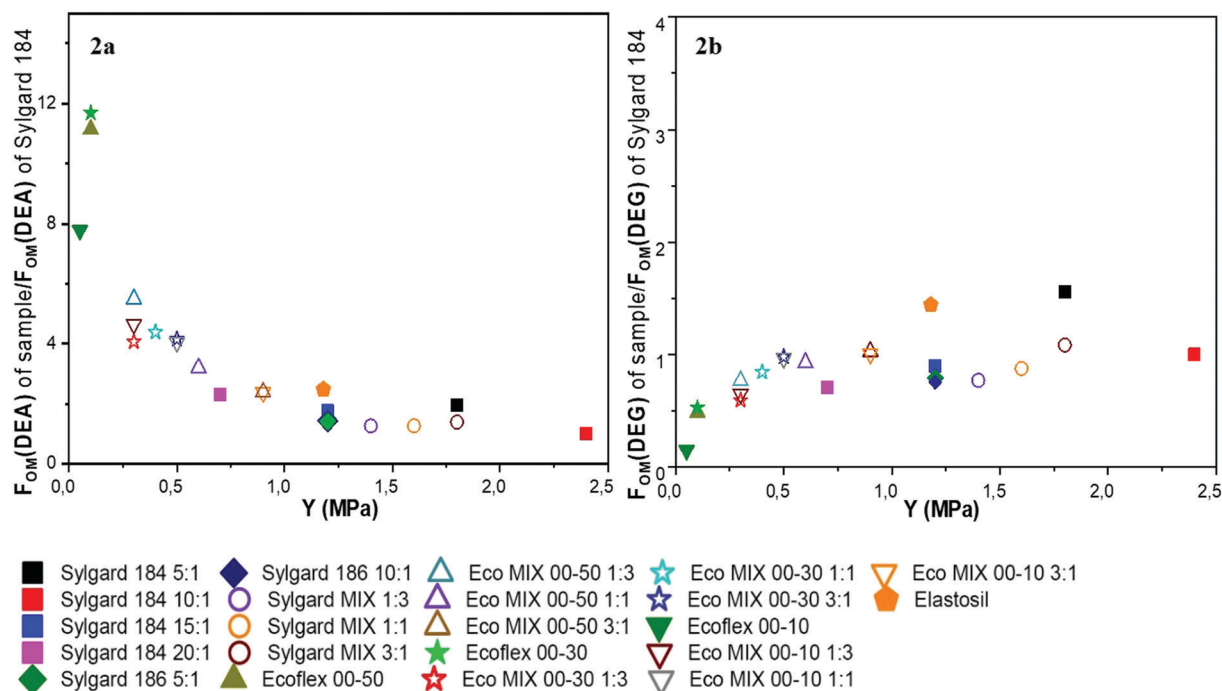


Fig. 2 Figures of merit (a) for actuator ( $F_{om}(DEA)$ ) and (b) for generator ( $F_{om}(DEG)$ ), both normalized with Sylgard 184. For the calculations,  $\epsilon'$  is measured at 1 Hz and  $\varphi$  calculated from hysteresis curves.

An efficient and quick way to include a higher  $\epsilon'$  and  $E_{BD}$  in silicone formulations is by fabricating such blends instead of using high dielectric constant fillers (e.g. titanium dioxide), which require tedious processing.<sup>25,26</sup> Furthermore, it is observed that all of the investigated elastomers break down electrically in similar ways by creating silica as a solid residue.<sup>27,28</sup> The dielectric loss tangent for all elastomers at 1 Hz is much lower than 1. Ecoflex 00-50 and Eco MIX 00-10 1:1 show the highest loss of approximately 0.06, due to their capacitive nature, which is still in the range suitable for performing as dielectric transducers.<sup>29</sup> Conductivity at 1 Hz (Table 2) for the elastomers is very low, and this is ideal for their performance as dielectric transducers.

### 3.5. Leakage current

The  $i_{leak}$  should be ideally lower than  $\sim 10^{-15}$  A for an elastomer to function as a reliable transducer,<sup>30</sup> and it affects the actuation efficiency and the lifetime of the transducer, especially in high-voltage operations.<sup>31,32</sup> In this work,  $i_{leak}$  is measured at an applied electric field of  $5 \text{ V } \mu\text{m}^{-1}$ . Measurements show that Sylgard 184 exhibits a relatively high  $i_{leak}$  of  $\sim 10^{-10}$  A at  $25^\circ\text{C}$ . Sylgard 186 shows the highest  $i_{leak}$  of  $\approx 10^{-9}$  A at  $60^\circ\text{C}$ . The  $i_{leak}$  of the elastomers at  $25^\circ\text{C}$  is plotted and shown in Fig. 1f, and all the  $i_{leak}$  data at  $25^\circ\text{C}$ ,  $40^\circ\text{C}$  and  $60^\circ\text{C}$  are tabulated in the ESI.†  $i_{leak}$  becomes significant with an increasing electric field and generates undesirable heat in the transducer.<sup>33</sup> Also, as the operating temperature increases,  $i_{leak}$  increases – as shown by our measurements. The sensitivity of  $i_{leak}$  ( $k^\circ\text{C}^{-1}$ ) to operating temperatures is plotted in Fig. 1g, which shows that Ecoflex 00-30, EcoMIX 00-10 1:3, Sylgard 186 5:1 and Sylgard 186 have higher sensitivity compared to the remaining elastomers. Apart from Sylgard 184

and 186, and Ecoflex 00-10, all samples show  $i_{leak} \leq 10^{-11}$  A at all three operating temperatures.

### 3.6. Figure of merit

The  $F_{om}(DEA)$ <sup>20</sup> and  $F_{om}(DEG)$ ,<sup>21</sup> which take into account the most important relevant parameters ( $\epsilon'$ ,  $E_{BD}$ ,  $Y$  and strain energy ( $\varphi$ ))<sup>34,35</sup> of dielectric elastomers, help in reviewing their theoretical performance as transducers. However, the  $F_{om}$  does not take into account current leakage or dielectric and mechanical losses.<sup>36</sup>

The  $F_{om}(DEA)$  is derived by Sommer-Larsen and Larsen,<sup>20</sup> and  $F_{om}(DEG)$  is derived by McKay *et al.*<sup>21</sup> as:

$$F_{om}(DEA) = \frac{3\epsilon' \epsilon_0 E_{BD}^2}{Y} \quad (1)$$

$$F_{om}(DEG) = \frac{\epsilon' \epsilon_0 E_{BD}^2}{2\varphi} \quad (2)$$

Here,  $\epsilon_0$  is the permittivity of free space ( $8.854 \times 10^{-12} \text{ F m}^{-1}$ ), and  $\varphi$  is the strain energy function of the elastomer.  $F_{om}(DEG)$  is comprehensive only when the strain energy function of the elastomer is considered, though data on this function are usually scarce. The  $\varphi$  of the elastomers is calculated from their hysteresis curves, the details of which can be found in ESI.†<sup>37</sup> The  $F_{om}(DEA)$  and  $F_{om}(DEG)$  are normalised with the Sylgard 184 benchmark and plotted in Fig. 2 as a function of  $Y$ . Upon analysing the  $F_{om}(DEA)$  and  $F_{om}(DEG)$ , it can be concluded that pure Ecoflex formulations are the best candidates for performing as actuators and by far exceed the Sylgard 184 benchmark, albeit they exhibit lower  $F_{om}(DEG)$  compared to Sylgard 184.

Alternatively, Sylgard 184 5:1 and Elastosil qualify as better materials to perform as generators in comparison to Sylgard 184. From Fig. 2, it is evident that softer elastomers perform well as actuators, and hard elastomers are good candidates for generators. In comparison to Sylgard 184, the following blends, namely Sylgard MIX 3:1, Eco MIX 00-50 3:1, Eco MIX 00-10 3:1 and Elastosil, show a higher  $F_{om}(DEA)$  and  $F_{om}(DEG)$  compared to Sylgard 184.

Hence, for transducer applications, the above blends and Elastosil would perform better than the benchmark. Another issue to consider when transitioning from research to commercialisation is a product's lifetime. Research on this crucial topic is emerging and holds great promise for the transducer technology.<sup>38–43</sup> Of all the properties considered for  $F_{om}$ , the  $E_{BD}$ , though ideally an intrinsic material property, is often compromised as a result of elastomer processing, electrode geometry and transducer operating conditions.<sup>44–46</sup>

### 3.7. Ageing

Properties of the aged samples (6 months) are remeasured to examine how ageing influences the properties of the film samples (tabulated in the ESI†). Sylgard 184 5:1, Sylgard 186, Sylgard MIX 1:3, Sylgard MIX 3:1, EcoMIX 00-50 1:3 and Eco MIX 00-30 3:1 show a higher  $Y$  with age, which is due to post-curing.<sup>47</sup> Post-curing may be an effect of residual hydride groups condensing with water and then subsequently cross-linking,<sup>47</sup> as well as sterically hindered reactive groups being given time enough to react. Generally for silicone elastomers, as  $Y$  increases in post-curing,  $\sigma_{max}$  increases and  $\epsilon_{max}$  decreases. The  $\epsilon_{max}$  of most of the samples depreciated with age. Sylgard 184, Sylgard 184 15:1, Sylgard MIX 3:1 display a higher  $\epsilon_{max}$  with age, due to relaxation processes taking place in the network, which is also manifested by the decreasing  $Y$  with age as the elastomers soften. The  $E_{BD}$  of Sylgard 184 15:1, Sylgard 186, Sylgard MIX, EcoMIX 00-50, EcoMIX 00-30 and EcoMIX 00-10 increased with age, which is commonly observed for silicone elastomers that harden while post-curing.<sup>38</sup>

The  $S_{hard}$  [00] of the samples shows an overall decreasing trend with age except for Sylgard 184 15:1, Sylgard 184 20:1, Sylgard 186, Sylgard MIX 3:1, Ecoflex 00-30, Ecoflex 00-10, Eco MIX 30 1:3 and Eco MIX 10 3:1. With regards to  $T$ , the aged samples of Eco MIX 00-50 1:3, Eco MIX 00-50 1:1 and Eco MIX 00-10 1:3 show higher optical transparency. The rest of the aged samples show a similar or slightly reduced  $T$  compared to the fresh samples. If curing conditions are optimised, the impact of post-curing on elastomer properties can be diminished to a great extent.

## 4. Conclusion

After examining the results, it is notable that the properties of commercial elastomer formulations can be manipulated, by either mixing them at different stoichiometric ratios or by blending two formulations with each other. From the present study, it can also be concluded that blending is a quick method

to make reliable soft networks with mechanical integrity, as opposed to varying the stoichiometry, which, if done inappropriately, often leads to the formation of weak networks.<sup>23</sup> A mixture of two formulations with contrasting properties often results in a blend that is reinforced with the properties that are intermediate to both formulations. The properties of the blends always fall within limits, defined by the parent formulations. Appropriate curing conditions should be employed for these blends, to ensure that their properties remain constant over time. The pure Ecoflex formulations show much higher  $F_{om}(DEA)$  and very poor  $F_{om}(DEG)$ , compared to the Sylgard 184 benchmark. The blends Sylgard MIX 3:1, EcoMIX 00-50 3:1, EcoMIX 00-10 3:1 and Elastosil show a higher  $F_{om}(DEG)$  and  $F_{om}(DEA)$  compared to the Sylgard 184 benchmark, and so they qualify as better candidates compared to Sylgard 184 in terms of fabricating transducers. It is thereby demonstrated that a blend of two formulations with different properties can be advantageous. Moreover, such blends can act as a good starting point for developing advanced hi-tech, versatile and stretchable devices.

## Conflicts of interest

There are no conflicts to declare.

## Acknowledgements

The authors acknowledge funding from the Independent Research Fund Denmark.

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