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Electronic Supplementary information

Voltage-stabilised elastomers with increased relative permittivity and high electrical breakdown strength by means of phase separating binary copolymer blends of silicone elastomers

Aliff Hisyam A Razak^{1,2}, Liyun Yu¹ and Anne Ladegaard Skov¹

¹ Danish Polymer Center, Department of Chemical and Biochemical Engineering, Technical University of Denmark, Building 227, 2800 Kgs. Lyngby, Denmark.

² Faculty of Engineering Technology, University of Tun Hussein Onn Malaysia, 86400 Parit Raja, Batu Pahat, Johor, Malaysia.

1) Calculation of engineering stress and strain

The engineering stress (σ_E) was calculated from the force (F) and the cross-sectional area of the strip (A):

$$\sigma_E = \frac{F}{A} = \frac{F}{t \times w} = \frac{\tau \cdot d}{t \cdot w}$$

Equation 1

where A = film thickness (t) · constant width ($w = 6$ mm) and F = torque (τ) · drum diameter ($d = 10.3$ mm).

The engineering strain (ϵ_E) was calculated as a ratio of a stretched strain ($L - L_0$) to an initial strain (L_0) as:

$$\epsilon_E = \frac{L - L_0}{L_0}$$

Equation 2

where a final strain after stretching (L) was determined from Hencky strain (ϵ_H) as follows:

$$\epsilon_H = \ln \frac{L}{L_0}$$

Equation 3

$$L = L_0 e^{\epsilon_H} = L_0 e^{(r_H \cdot t_s)}$$

Equation 4

where ϵ_H is a product of Hencky rate ($r_H = 1 \times 10^{-3}$ rotation/s) and step time (t_s).

By putting equation (4) in (2), the final expression of engineering strain (ϵ_E) was obtained as below:

$$\epsilon_E = e^{\epsilon_H} - 1$$

Equation 5

Young's moduli were determined from slopes in the linear regime of stress-strain plots at 5 % strain.

2) NMR spectra of synthesised copolymers

The NMR spectra for synthesised PDMS-PPMS and PDMS-PEG copolymers are shown in Figures S1– S5.

a) PDMS-PPMS copolymer (**80DMS_2PMS**, $C_{C_6H_6} = 8.4 \cdot 10^{-4} \text{ mol g}^{-1}$)

$^1\text{H-NMR}$ (CDCl_3 , 300 MHz): δ -0.02 - δ 0.6 (m, 6 H's, $-\text{SiO}(\text{CH}_3)_2-$), δ 4.70 (m, 1 H, $-\text{SiH}-$), δ 7.10 - δ 7.60 (m, 5 H's, $-\text{SiC}_6\text{H}_5-$).

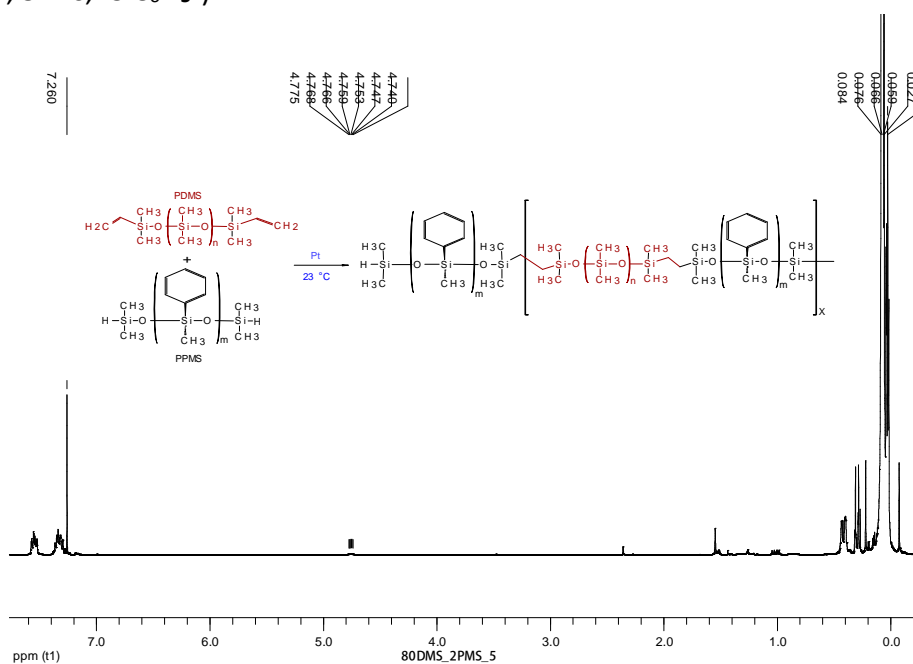


Figure S1 The NMR for 80DMS_2PMS.

b) PDMS-PEG copolymer (**PDMS81-PEG**)

$^1\text{H-NMR}$ (CDCl_3 , 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, $-\text{Si}(\text{CH}_3)_2\text{O}-$), δ 3.50 - δ 3.70 (m, 4 H's, $-\text{C}_2\text{H}_4\text{O}-$), δ 0.98 - δ 1.03 (t, 2 H's, $-\text{SiCH}_2-$), δ 3.53 - δ 3.57 (m, 2 H's, $-\text{CCH}_2\text{O}-$).

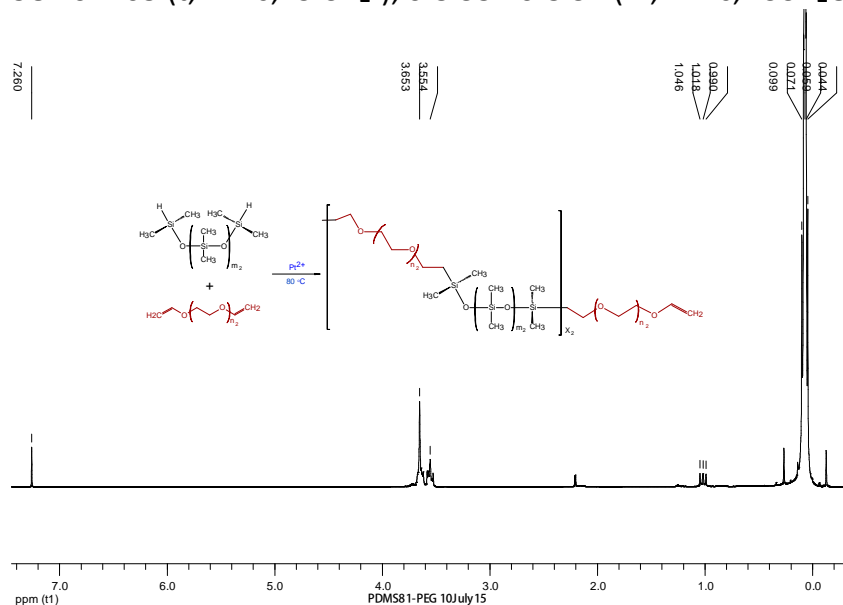


Figure S2 The NMR for PDMS81-PEG.

c) PDMS-PEG copolymer (**PDMS14-PEG**)

$^1\text{H-NMR}$ (CDCl_3 , 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, $-\text{Si}(\text{CH}_3)_2\text{O}-$), δ 3.50 - δ 3.70 (m, 4 H's, $-\text{C}_2\text{H}_4\text{O}-$), δ 0.98 - δ 1.03 (t, 2 H's, $-\text{SiCH}_2-$), δ 3.53 - δ 3.57 (m, 2 H's, $-\text{CCH}_2\text{O}-$).

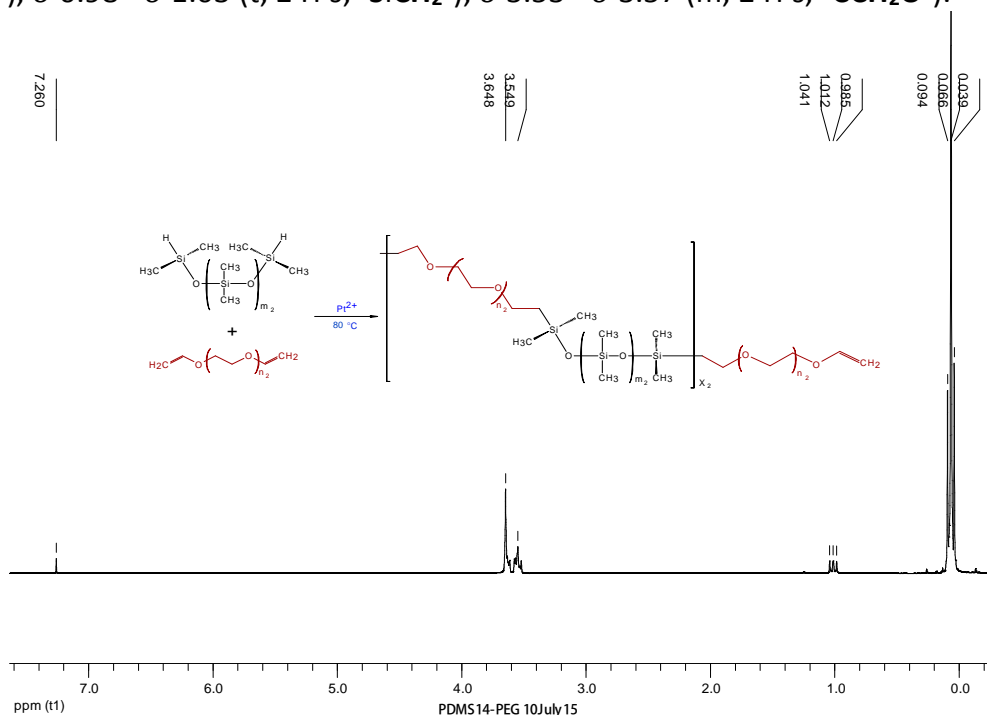


Figure S3 The NMR for PDMS14-PEG.

d) PDMS-PEG copolymer (**PDMS7-PEG**)

$^1\text{H-NMR}$ (CDCl_3 , 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, $-\text{Si}(\text{CH}_3)_2\text{O}-$), δ 3.50 - δ 3.70 (m, 4 H's, $-\text{C}_2\text{H}_4\text{O}-$), δ 0.98 - δ 1.03 (t, 2 H's, $-\text{SiCH}_2-$), δ 3.53 - δ 3.57 (m, 2 H's, $-\text{CCH}_2\text{O}-$).

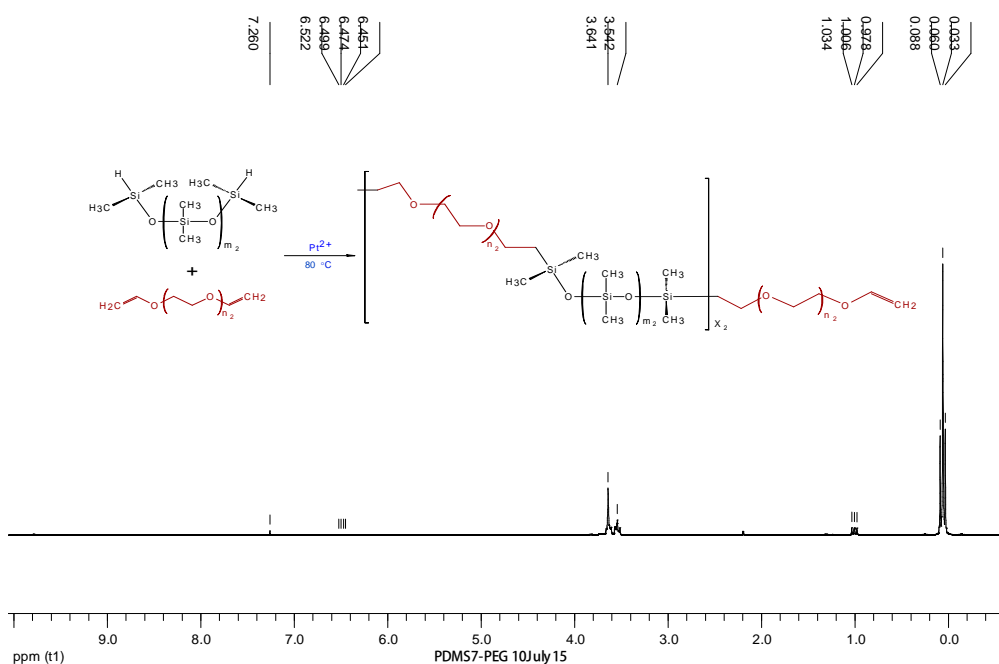


Figure S4 The NMR for PDMS7-PEG.

e) PDMS-PEG copolymer (**PDMS3-PEG**)

$^1\text{H-NMR}$ (CDCl_3 , 300 MHz): δ 0.05 - δ 0.09 (m, 6 H's, $-\text{Si}(\text{CH}_3)_2\text{O}-$), δ 3.50 - δ 3.70 (m, 4 H's, $-\text{C}_2\text{H}_4\text{O}-$), δ 0.98 - δ 1.03 (t, 2 H's, $-\text{SiCH}_2-$), δ 3.53 - δ 3.57 (m, 2 H's, $-\text{CCH}_2\text{O}-$).

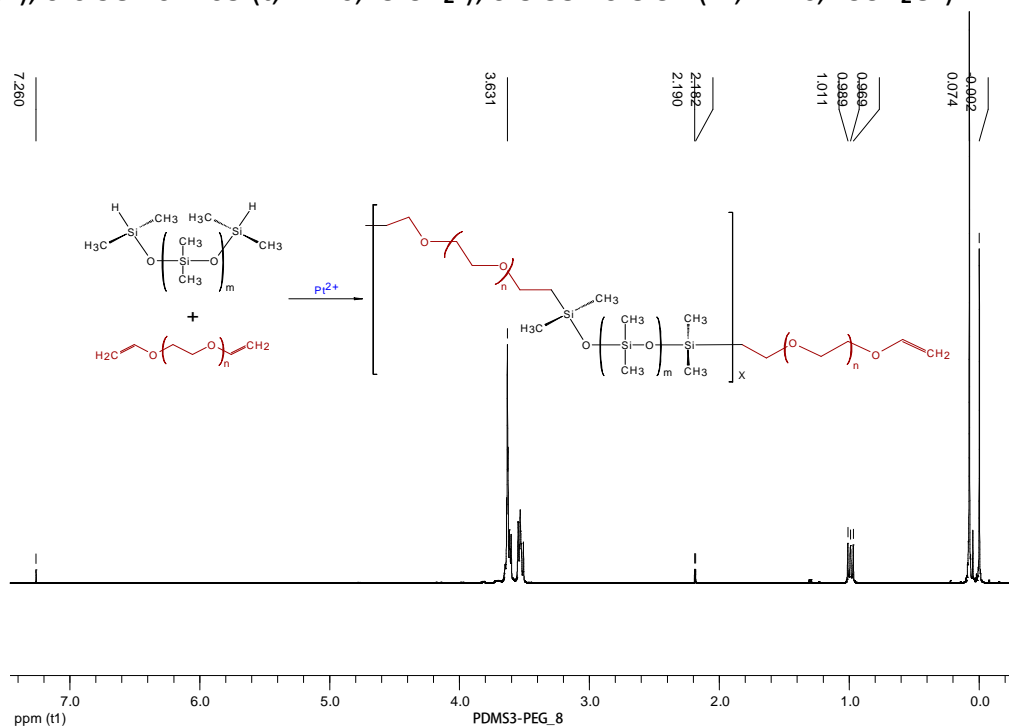
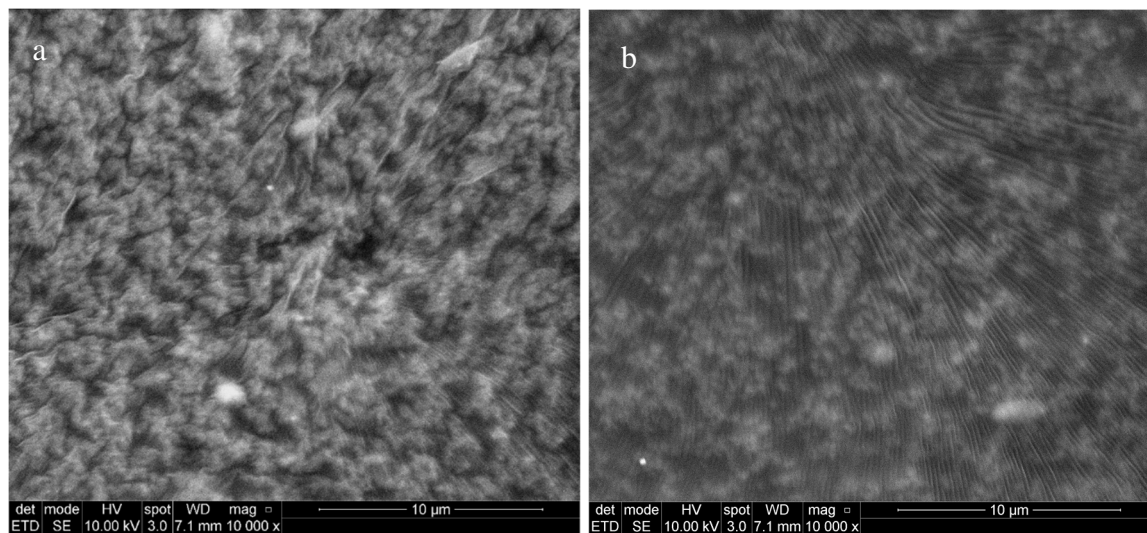


Figure S5 The NMR for PDMS3-PEG.

3) SEM images



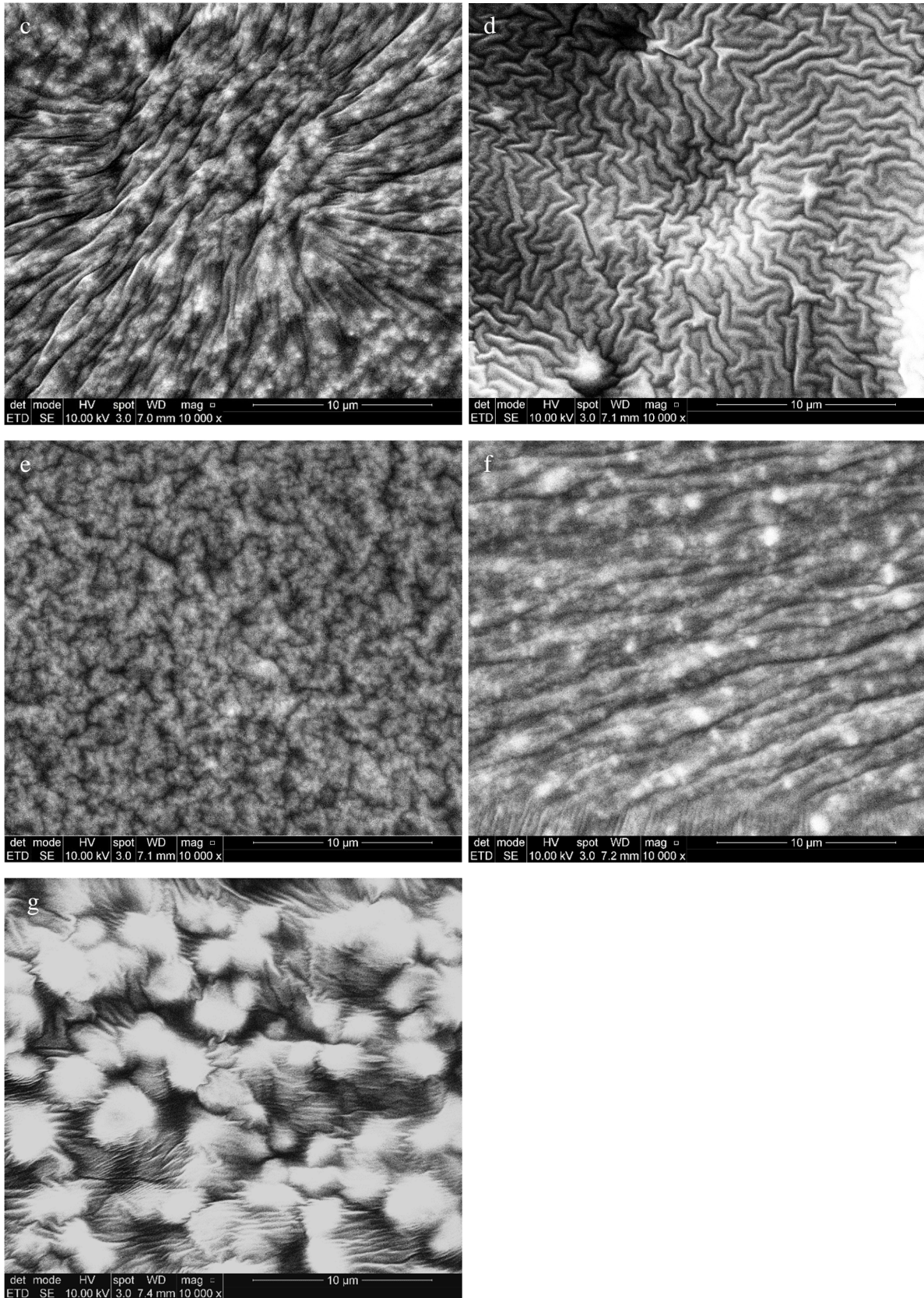


Figure S6 SEM images cross-linked BCBs with: **a)** 10 phr PDMS81-PEG, **b)** 20 phr PDMS81-PEG, **c)** 10 phr PDMS14-PEG, **d)** 20 phr PDMS14-PEG, **e)** 10 phr PDMS7-PEG, **f)** 10 phr PDMS3-PEG, and **g)** 20 phr PDMS3-PEG.