Supporting Information

Mixing the Immiscible: Blends of Dynamic Polymer Networks

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Table of Contents

Materials

Hydroxyl-terminated polydimethylsiloxane (PDMS, 750 cst) and boric acid (99.8%) were purchased from Sigma-Aldrich and were used as received.

Methods

Fourier transform infrared (FTIR) spectra were registered in a Jasco FT/IR 4100 spectrophotometer, using a Gladi ATR accessory and collecting 32 scans at a resolution of 4 cm^{-1}.

Rheological testing was carried out in a TA instruments AR2000ex rheometer using a 25 mm plate-plate geometry on 1 mm thick samples.

Mechanical testing was performed using an INSTRON 3365 Long travel Elastomeric Extensometer controlled by Bluehill Lite software. Tensile strength measurements were carried out according to UNE-EN-ISO 527 standard, using dump-bell type test specimens and an elongation rate of 500 mm min^{-1}.

Hardness measurements of all the blends and pristine materials were performed using a normalized Shore A durometer.

Synthesis of PUU elastomer

Molecular characteristics and preparation of the PUU elastomer used in this work have been previously described by our group.[1]

Synthesis of Si-Putty

Si-Putty was prepared in a 50 mL Haake Polylab internal mixer previously heated at 200 ºC. PDMS 750 cst (49.5 g) and boric acid (0.5 g) were added into the internal mixer while the rotors were rotating at 30 rpm. In a few seconds, the boric acid was melted and a putty was formed. The material was mixed for further 45 minutes in order to ensure a good homogenization, and the resulting **Si-Putty** was characterized by FTIR (see Figure S1). Yield: 48.3 g, 96.6%. FTIR (ATR, cm^{-1}): ν = 2962 (stretching CH_3), 1257 (CH_3 symmetric bending), 1077 and 1007 (stretching Si-O-Si), 786 (Si-CH_3 rocking).

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[1] a) A. Rekondo, R. Martin, A. Ruiz de Luzuriaga, G. Cabañero, H. J. Grande, I. Odriozola, *Mater. Horiz.,* 2014, **1**, 237-240, b) R. Martin, A. Rekondo, A. Ruiz de Luzuriaga, G. Cabañero, H. J. Grande, I. Odriozola, *J. Mater. Chem. A,* 2014, **2**, 5710-5715.

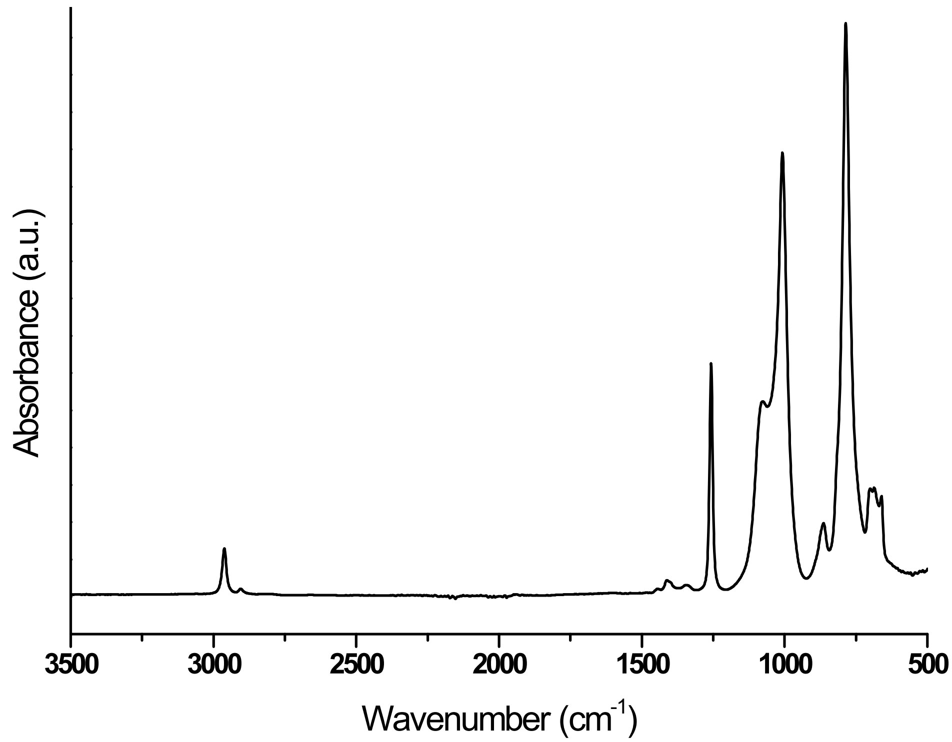

Figure S1. FTIR spectrum of **Si-Putty**.

Synthesis of DNBs

The three different DDNs using different **PUU**/**Si-Putty** ratios (75/25, 50/50 and 25/75 wt%) were prepared in a 50 mL Haake Polylab internal mixer. For the preparation of 50 g of each blend, **PUU** and **Si-Putty** were fed into the internal mixer at room temperature while the rotors were rotating at 30 rpm. Then the internal mixer was heated to 150 ºC and the blends were mixed for 1 hour at 150 ºC. The resulting homogeneous materials were placed in a 2 mm thick square mold and hot-pressed at 150 ºC for 10 minutes, to obtain the materials in the form of sheets. All the blends were characterized by FTIR (see Figures S2-S4).

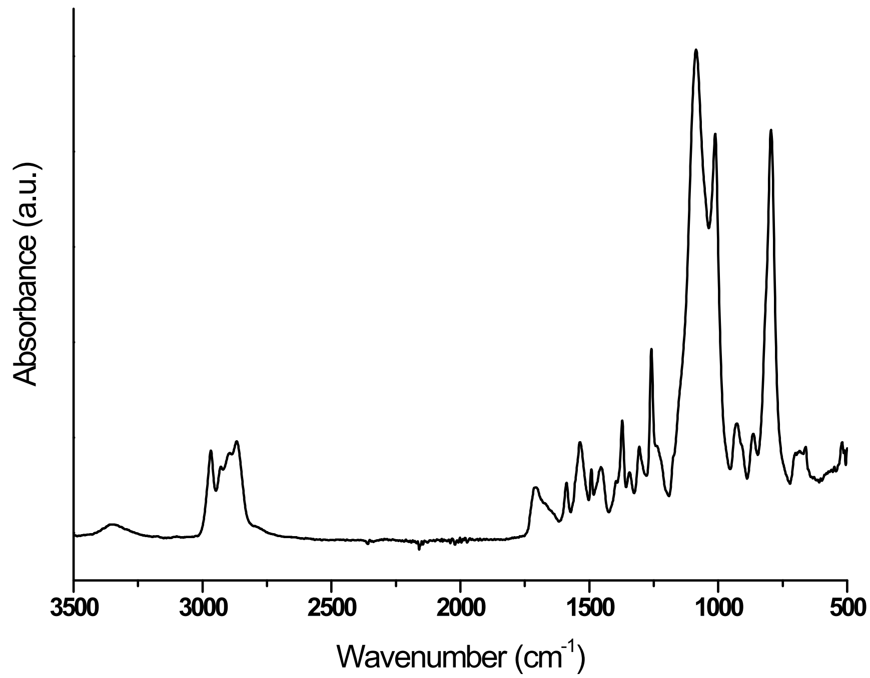

Figure S2. FTIR spectrum of $\mathbf{DNB_{75}}$.

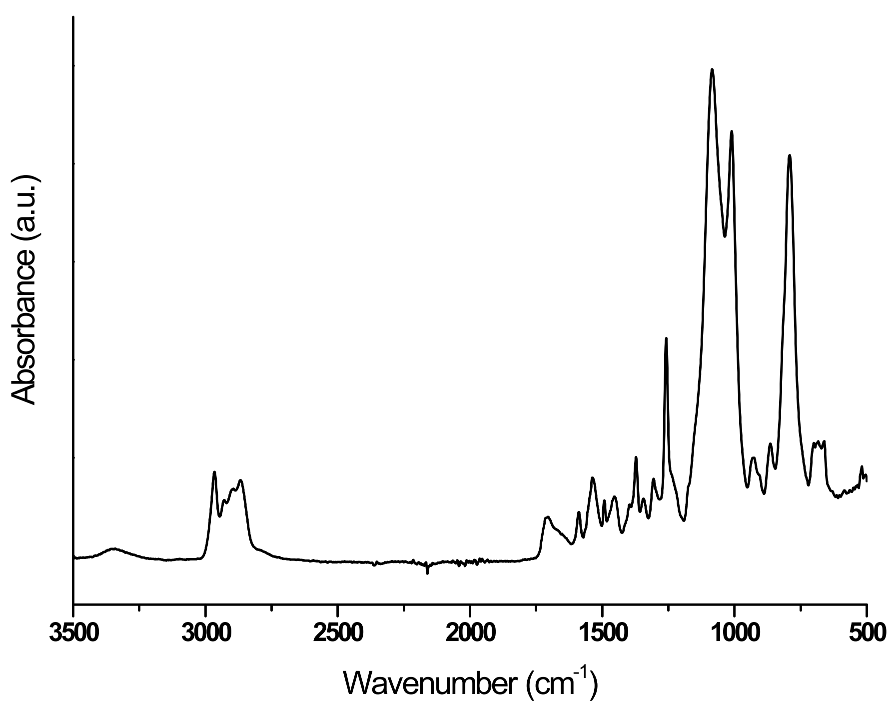

Figure S3. FTIR spectrum of $\mathbf{DNB_{50}}$.

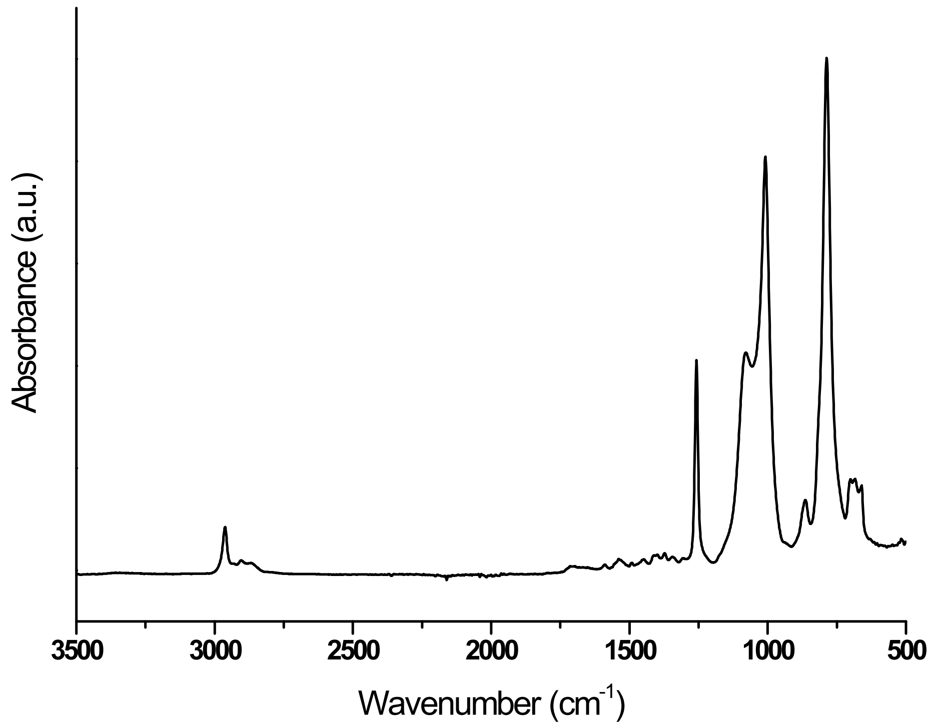

Figure S4. FTIR spectrum of **DNB$_{25}$**.

Rheology of Si-Putty and PUU

Dynamic viscoelastic results at T = 25 ºC. Dependence of both G' and G'' on frequency determined for **Si-Putty**.

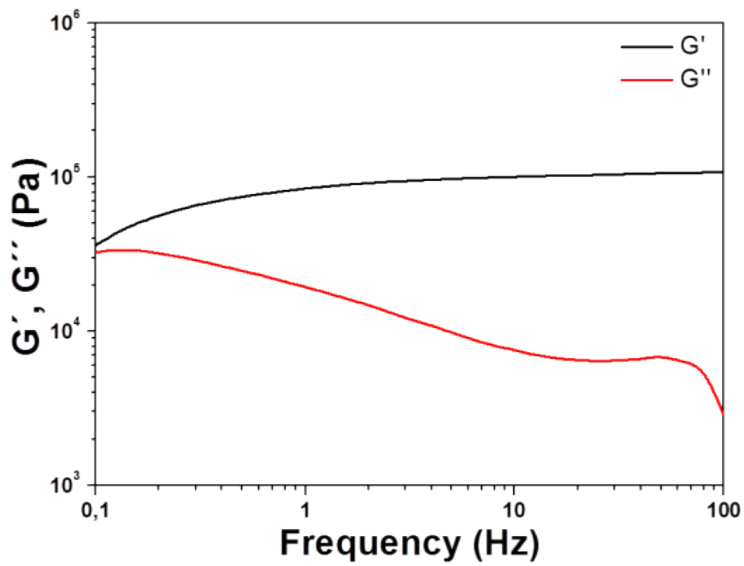

Figure S5. Viscoelastic results at T = 25 ºC. Elastic, G', and viscous, G'', modulus for **Si-Putty**.

Master curve of Si-Putty

Master curves of elastic modulus G' (red) and viscous modulus G'' (black) at Tr = 25 ºC for **Si-Putty**, were built from frequency scans at different temperatures in the range 25–60 ºC, under linear conditions. Viscoelastic models indicate that a relaxation time can be obtained from the frequency ω_x at the crossing point G' = G'': $\tau = 1/\omega_x$

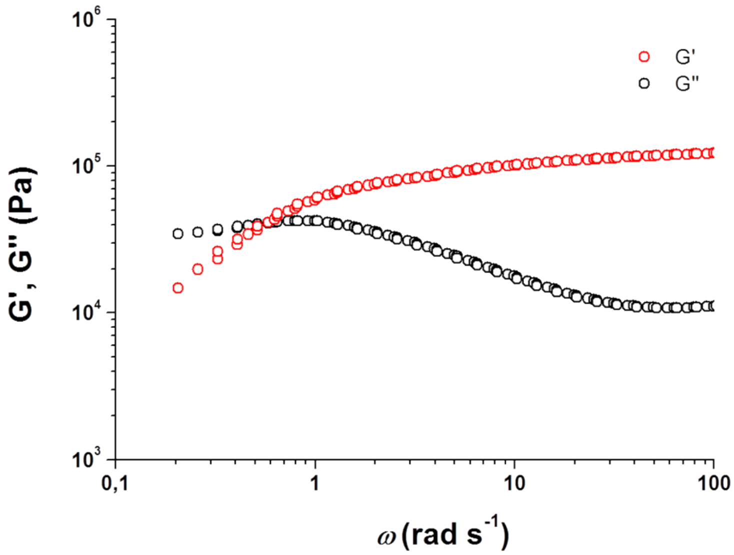

Figure S6. Storage (G´) and loss (G´´) modulus (Pa) vs. frequency extrapolated master curve for **Si-Putty**.

Rheology of PUU

Dynamic viscoelastic results at T = 25 ºC. Dependence of both G' and G'' on frequency determined for **PUU**.

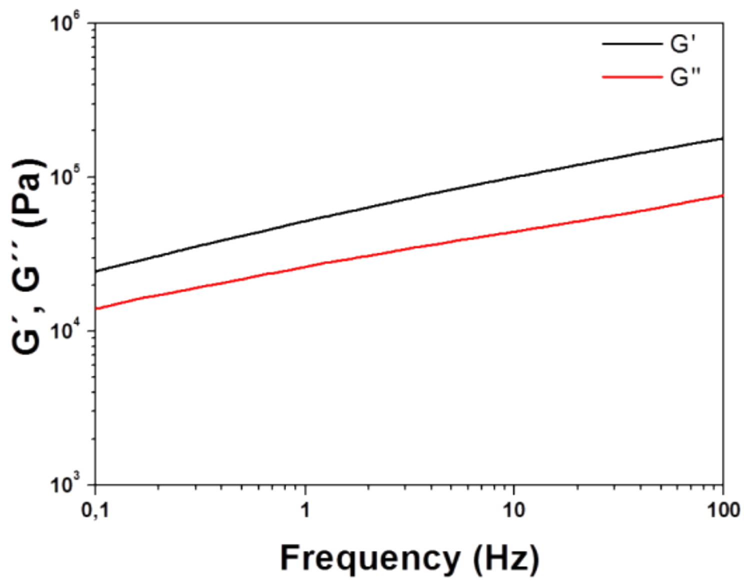

Figure S7. Viscoelastic results at T = 25 ºC. Elastic, G', and viscous, G'', modulus for **PUU**.