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ZIRKON

Silicone-based Vitrimers for High-Voltage Applications

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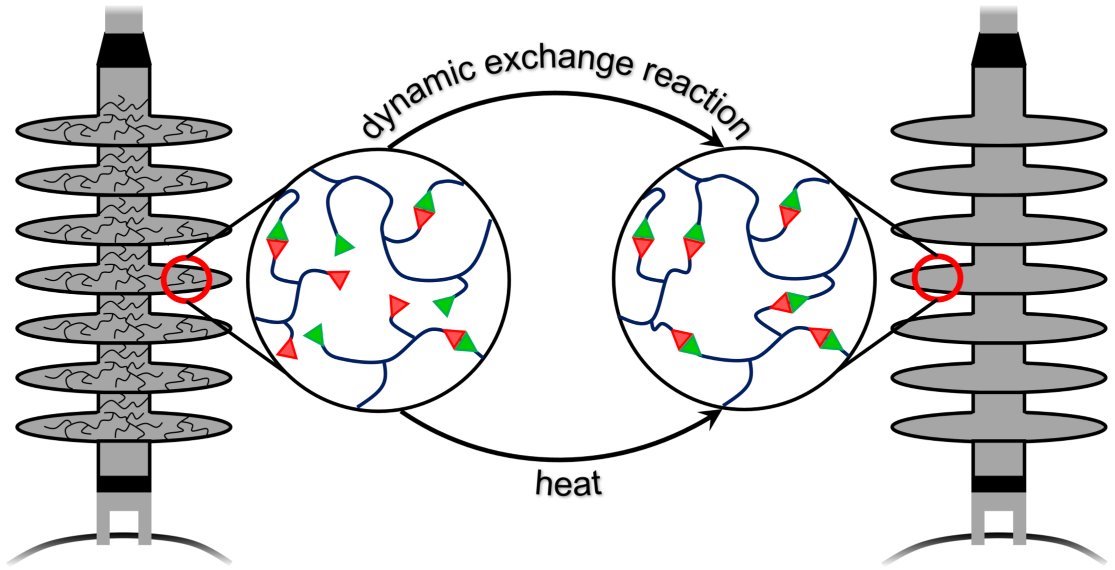
Introduction



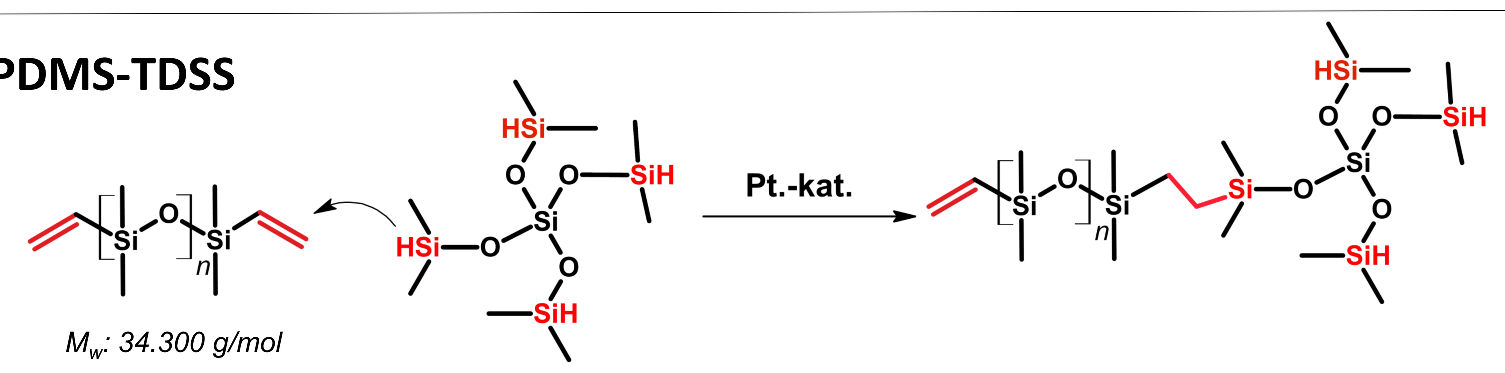
Polydimethylsiloxane (PDMS)-based elastomers, better known as silicones, are one of the most important materials for electrical insulation. However, a major drawback is their challenging recyclability. In recent years, numerous PDMS-based vitrimer systems have been developed in an effort to integrate silicones into a sustainable circular economy. Vitrimers are polymeric materials with thermoset-like behavior that can be reshaped and repaired under the influence of heat due to reversible covalent bond exchange reactions. These dynamic cross-links enable restructuring of the network architecture without permanently affecting the mechanical properties. The aim of our project is to identify vitrimer syntheses that are potentially of interest from an industrial viewpoint, implement them on a larger scale, and evaluate the suitability of the received materials as electrical insulators materials. First results are based on vinylogous urethane-based systems.

Material Preparation

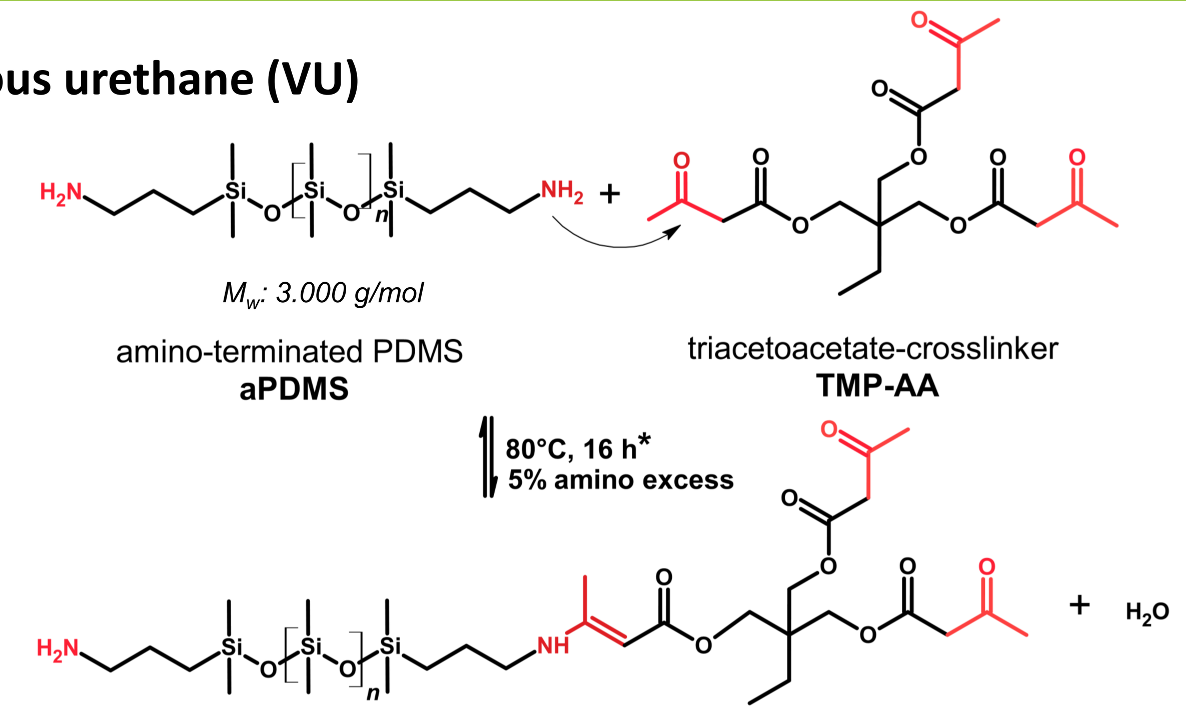
Systems under investigation:

A) vPDMS-TDSS: Pt-catalyzed silicone elastomer – a *pure* PDMS network [1]
B) Sylgard 184: commercial PDMS-rubber with unknown amount of silica fillers, crosslinking chemistry similiar to A (Pt-catalyzed hydrosilylation)
C) VU: vinylogous urethane PDMS, synthesis is based on reference [2]

vPDMS-TDSS



Vinylogous urethane (VU)

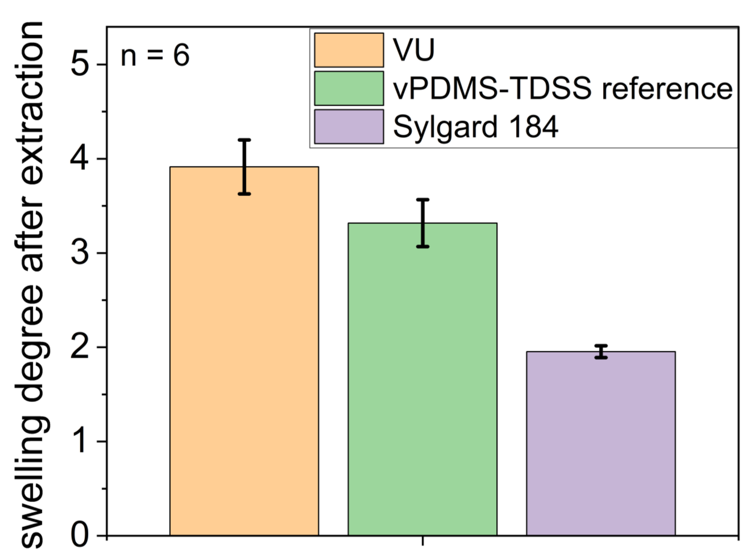


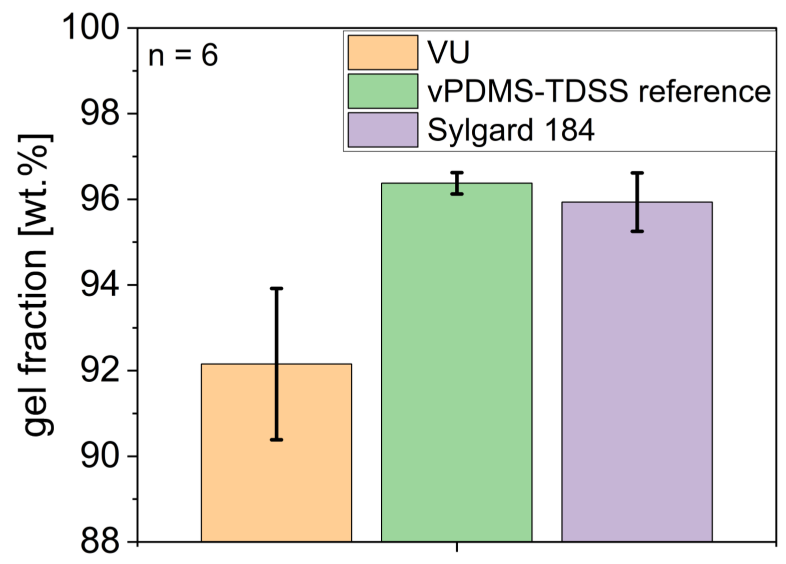
* for larger batches (65 g or more), 15 min pre-mixing with an overhead stirrer is essential to prevent water incorporation into the network

Characterization

Sol-gel analysis

- 6 hours continuous Soxhlet extraction with toluene

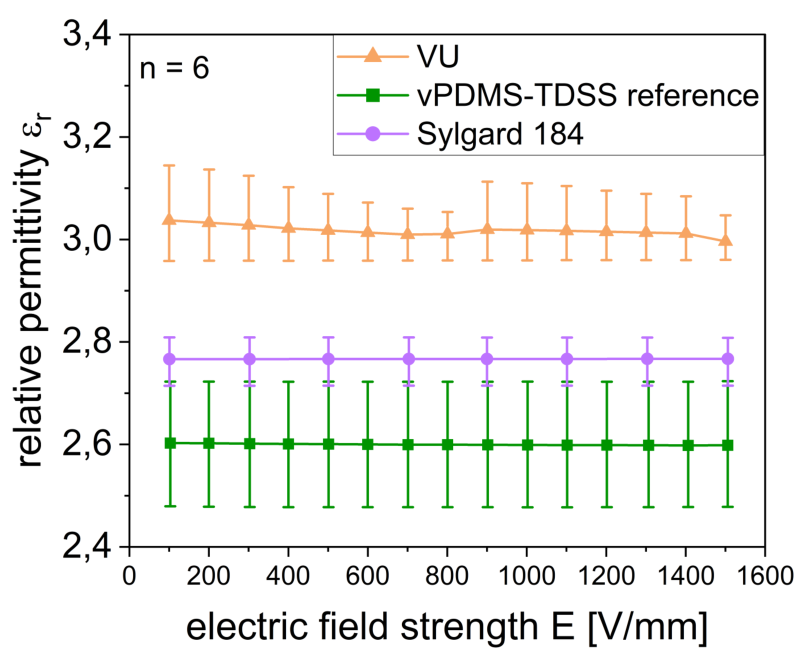


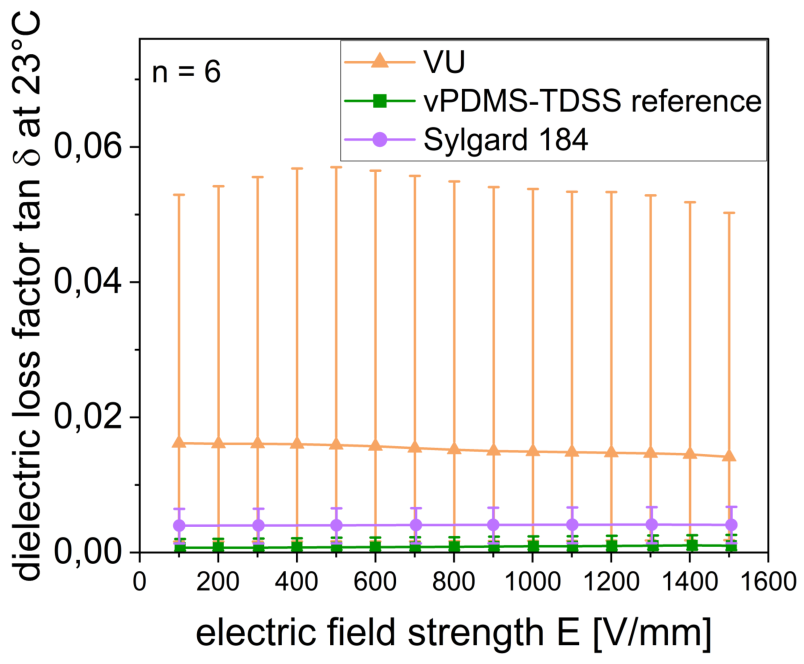


- VU network is insoluble in organic solvents, successful crosslinking
- Slightly higher amount of non-reacted soluble contents than reference materials

Dielectric properties at room climate

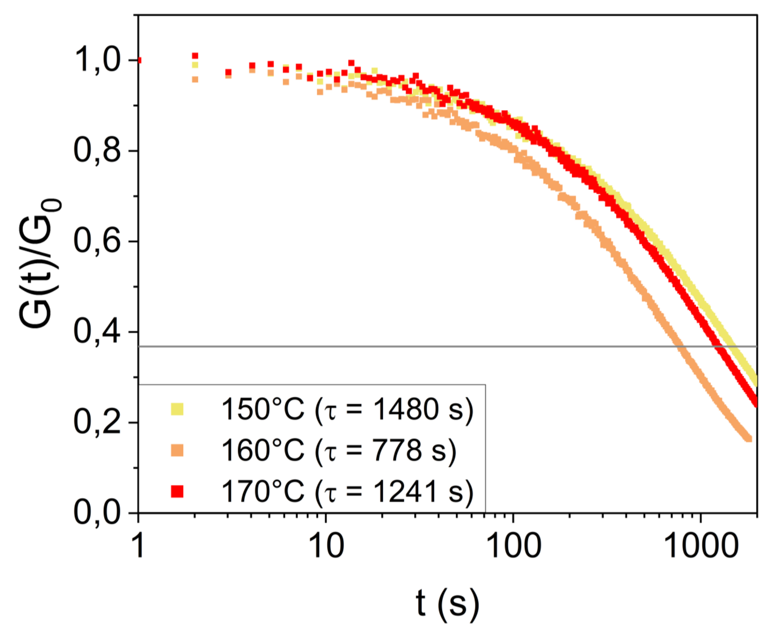
- **Loss factor tan δ**: comparable with filled silicones (low values desirable: 1e-4)
- **Relative permittivity ε_r** (polarisability): normal range for silicone rubber – no field dependence





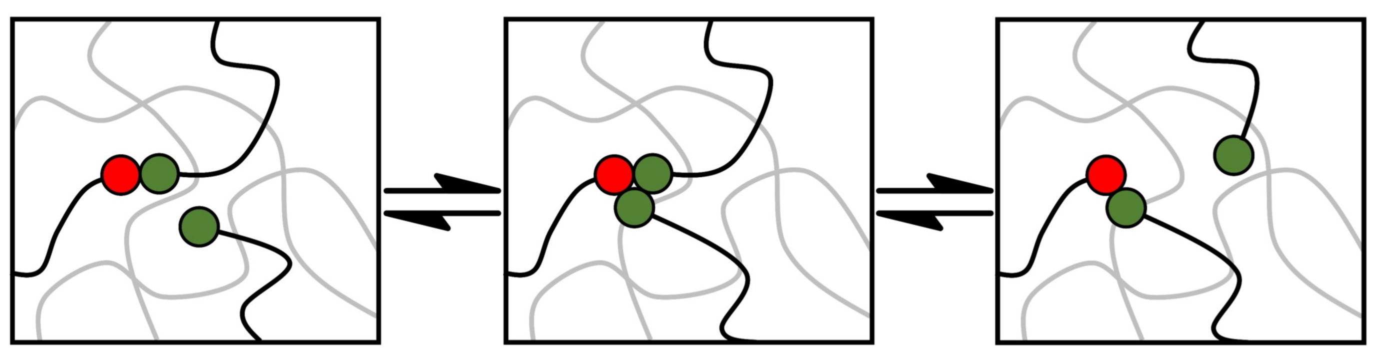
Stress-strain behavior for VU

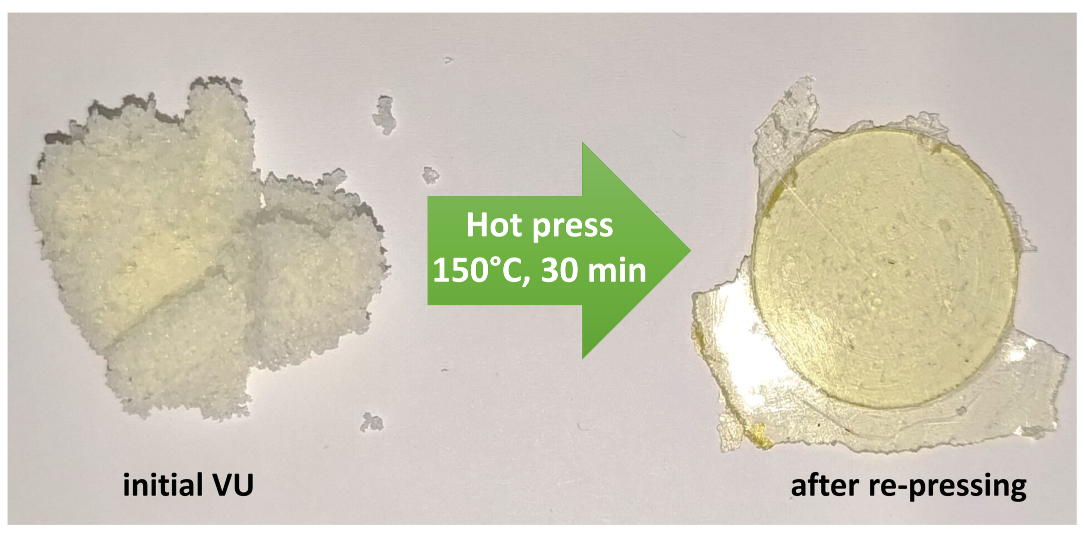
- Prolonged relaxation time at 170 °C → possible thermal decomposition of the product?



Preliminary tests for recycling

- Assumed associative mechanism for bond exchange





- Re-pressed silicones do not exhibit any cracking

Summary and Outlook

- PDMS vitrimers bases on vinylogous urethane chemistry were successfully reproduced, they exhibit vitrimer-like behavior (insoluble in organic solvents, relaxation behavior, and recyclability).
- dielectric measurements show no field dependence, loss factor is slightly higher for unfilled silicone rubber

Outlook:

- Optimization of synthesis procedure and incorporation of mechanical fillers
- Further high-voltage testing according to IEC TR 62039 [3] (electrical breakdown strength, tracking resistance, etc.)

References

[1] F.Praße, S.Kornhuber, B.Voit, J.Weber: Impact of the Network Density of Rough Poly(dimethylsiloxane)-Model Systems on the Hydrophobicity Assessment and Dynamic Wetting behavior, *ACS Applied Polymer Materials* **2022**, *4*, 4109
[2] Y. Spiesschaert, M. Guerre, L. Imbernon, J. M. Winne, und F. Du Prez, „Filler reinforced polydimethylsiloxane-based vitrimers", *Polymer* **2019**, *172*
[3] IEC TR 62039:2021, „Selection Guide for polymeric materials for use under HV stress". *International Electrotechnical Commission*, **2021**

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