

Supporting Information

Designing recyclable inorganic-organic hybrid polymer coatings through integrating silicone-based molecular scission points

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Supporting Information

This document contains eight pages, including one table and eight supplementary figures.

The supporting information contains contact angles against water as well as CH₂I₂ and surface energy values for every coating system, images of colored coatings on different substrates, ²⁹Si-NMR spectra of the soluble fraction of the depolymerization mixture, a general reaction scheme of anionic ring opening polymerization (ROP), an overview of the reuse of non-silicone matrix shares, light scanning microscopy (LSM) analysis of a delaminated microscope glass slide, images of the delamination of different substrates, an image of the general coating delamination process, SEC data of a neat D₄ sample, the neat vinyl-capping agent and the repolymerized PDMS.

Table S1: Contact angles against diiodomethane and surface energy values for every coating system.

Samples	M_w [g/mol]	Contact angles against diiodomethane [°]	Surface energy [mN/m]
S_4%	550 (short chains, S)	68 (± 2)	26 (± 1)
S_8%		69 (± 1)	25 (± 1)
S_11%		74 (± 1)	23 (± 1)
S_14%		72 (± 3)	23 (± 2)
S_20%		70 (± 1)	25 (± 1)
L_4%	3400 g/mol (long chains, L)	90	15
L_8%		84 (± 2)	18 (± 1)
L_11%		87 (± 4)	17 (± 3)
L_14%		93 (± 6)	15 (± 4)

L_20%		92 (± 3)	15 (± 2)
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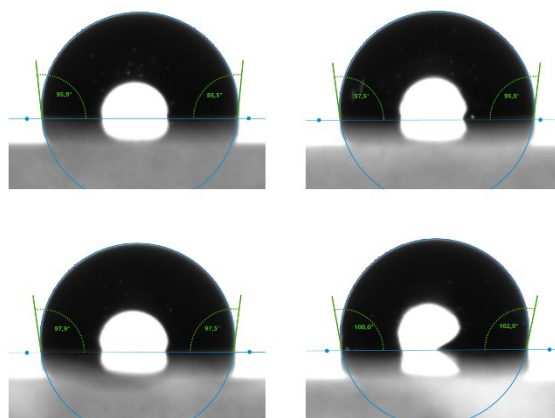


Figure S1: Water contact angle measurements for sample S_4%.

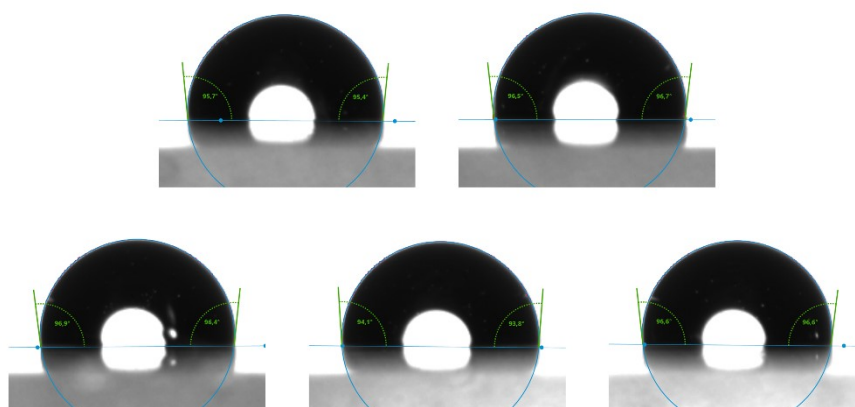


Figure S2: Water contact angle measurements for sample S_8%.

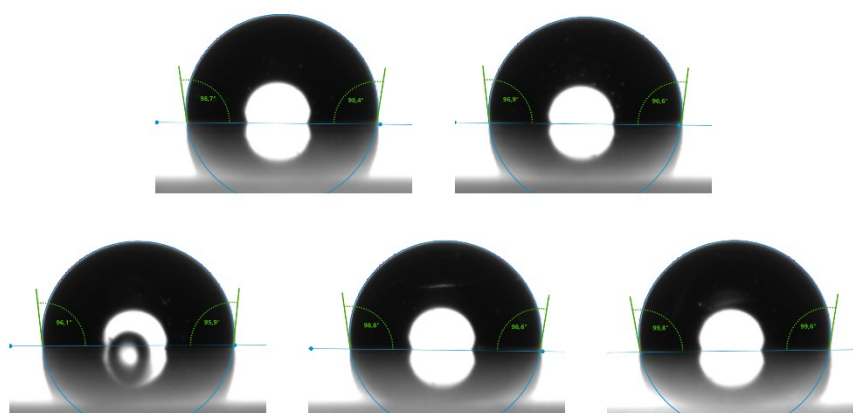


Figure S3: Water contact angle measurements for sample S_11%.

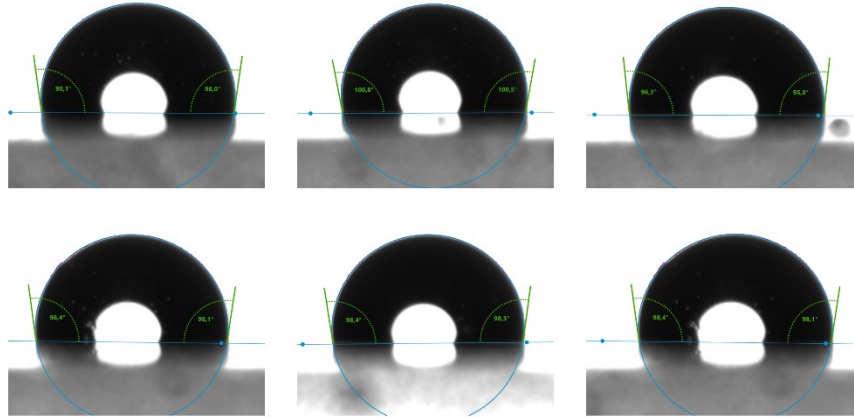


Figure S4: Water contact angle measurements for sample S_14%.

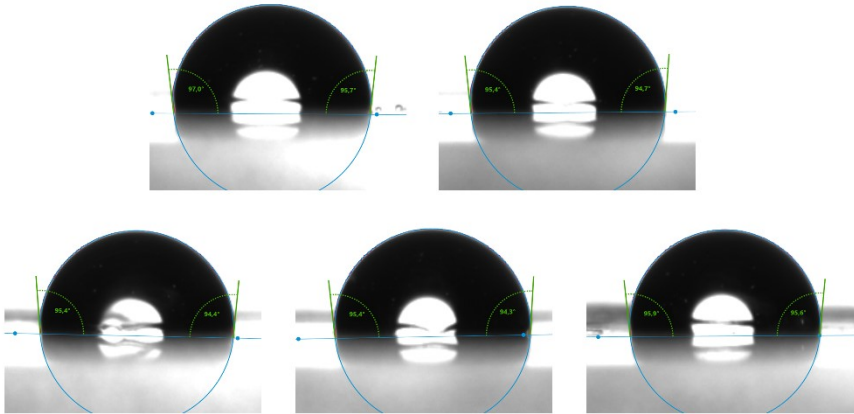


Figure S5: Water contact angle measurements for sample S_20%.

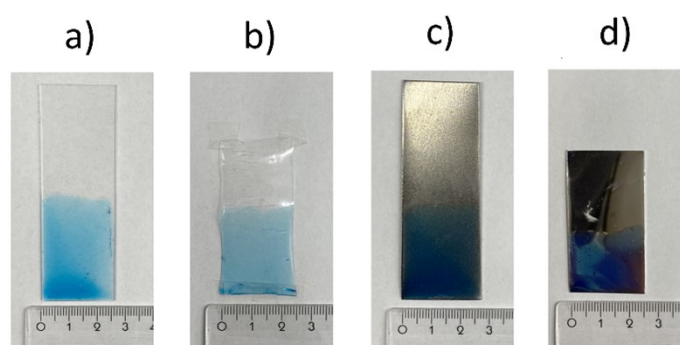


Figure S6: Coatings on a) glass, b) polyolefin, c) stainless steel and d) aluminum substrates. Transparent coatings were colored blue for display purposes using Ciba® ORASOL® Blue GN (Cu-phthalocyanine).

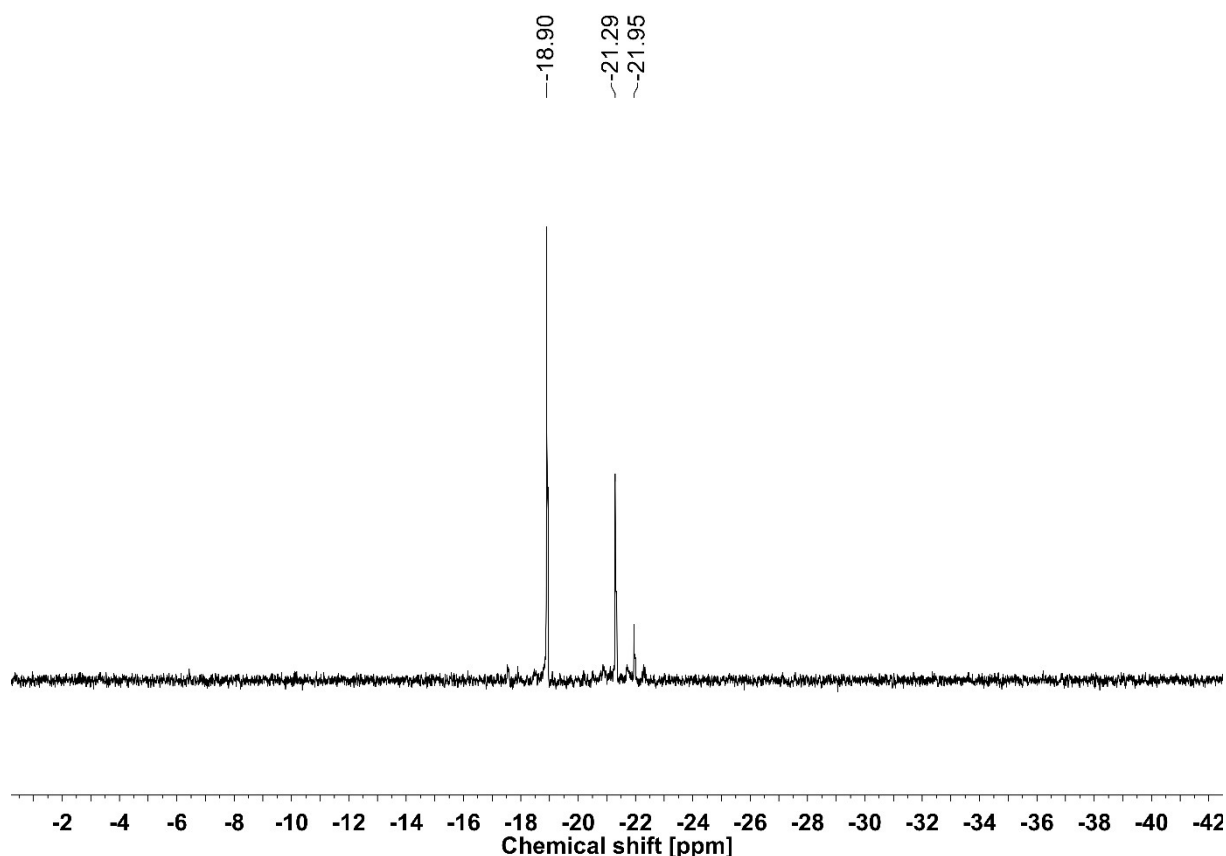


Figure S7: ^{29}Si -NMR spectrum of the soluble fraction of the depolymerization mixture. Signals at - 18.90 ppm, - 21.29 ppm and - 21.95 ppm can be assigned to cyclic siloxanes D_4 , D_5 and to linear polydimethylsiloxane (PDMS).^{1,2} TMS was used as a reference.

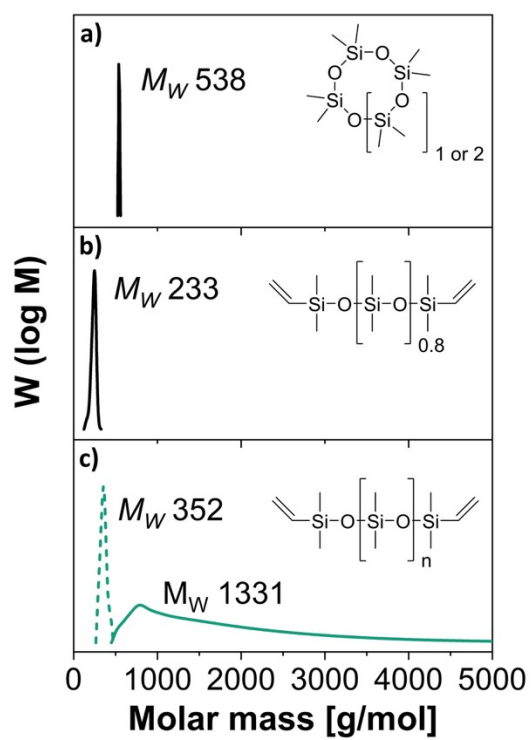


Figure S10: SEC of a D₄ reference (a), vinyl capping agent (b), and the reaction product of the ROP (c).

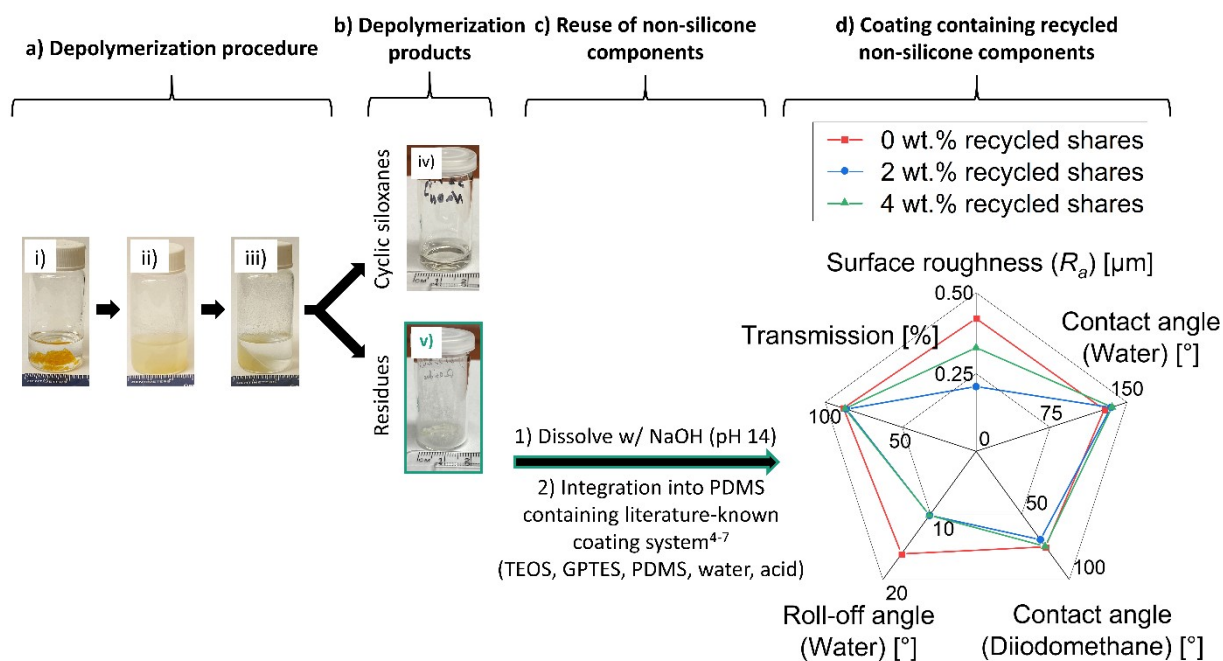


Figure S11: Reuse of inorganic-organic hybrid polymer coating's (IOHPC) non-silicone shares derived from the selective silicone depolymerization procedure: a) The depolymerization procedure (i: Polymer submersion in THF/TBAF, ii: Depolymerization at RT within 24 hours, iii: Separation of hybrid components (solid precipitate) by centrifugation) yields the depolymerization products, namely b) cyclic siloxanes (iv) and the non-silicone residues as a colorless powder (v). c) This powder is reused by dissolution in an alkaline solution of pH 14. The mixture is integrated into a PDMS-containing sol-gel coating, which is reported in literature⁴⁻⁷. d) The coating without recycled shares is characterized and compared with coatings containing 2 and 4 wt.% of recycled content (spider chart). Properties like transmission, surface roughness (R_a), contact angles against water and diiodomethane remain in the same area. The water roll-off angle decreases for coatings containing recycled shares.

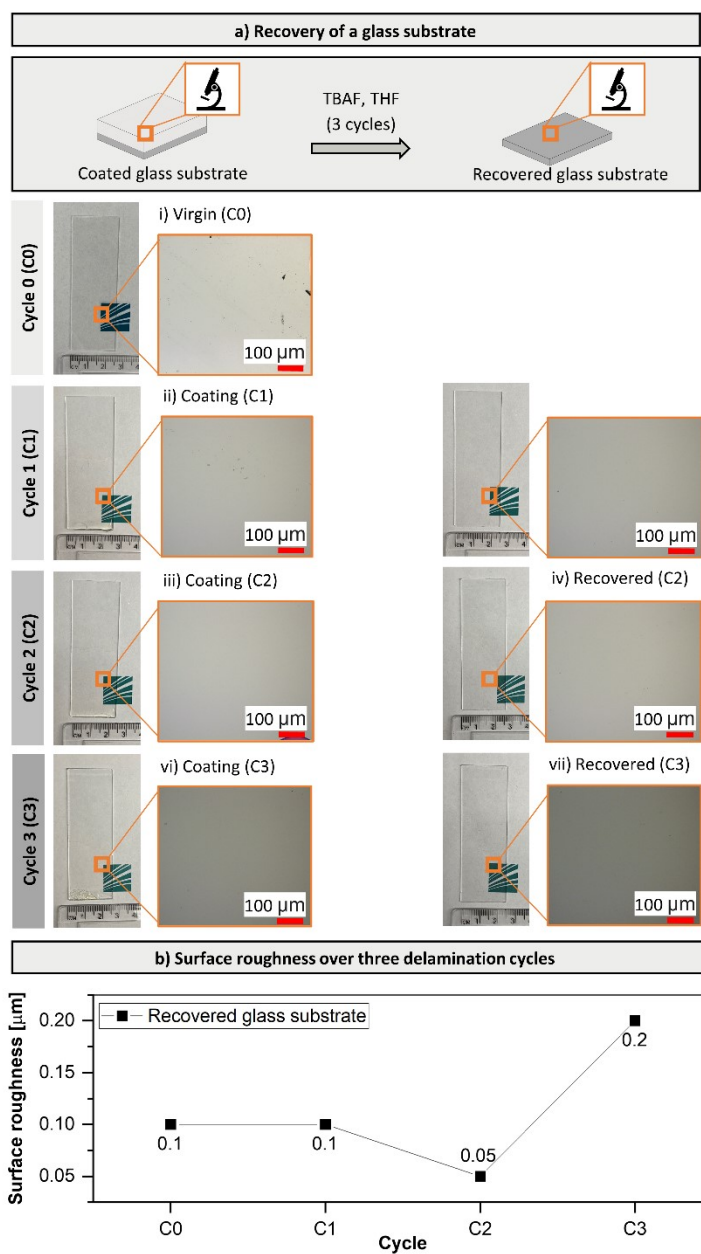


Figure S12: a) Recovering of a coated glass substrate by delamination with a depolymerization solution (tetrabutylammonium fluoride (TBAF) in tetrahydrofuran (THF)). Surface changes were evaluated *via* laser scanning microscopy (LSM). b) The evolution of the surface roughness values (R_a) are displayed for the reference and the recovered substrate.

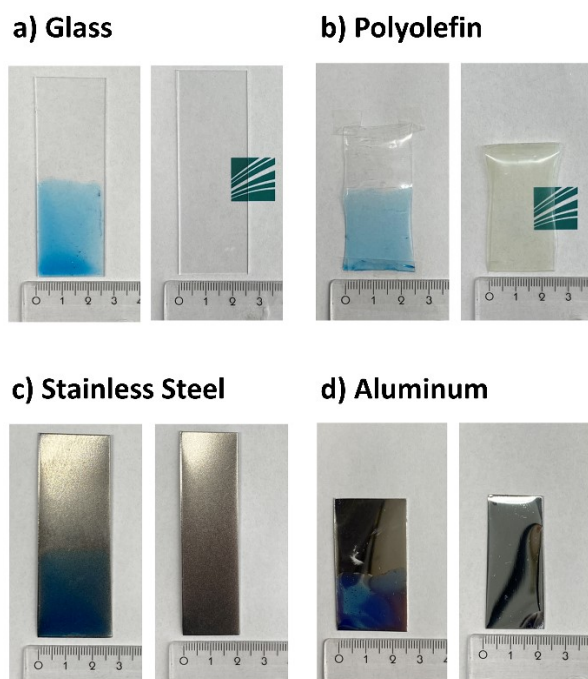


Figure S13: Photographs of delaminated a) glass, b) polyolefin, c) stainless steel and d) aluminum. Transparent coatings were colored blue for display purposes.

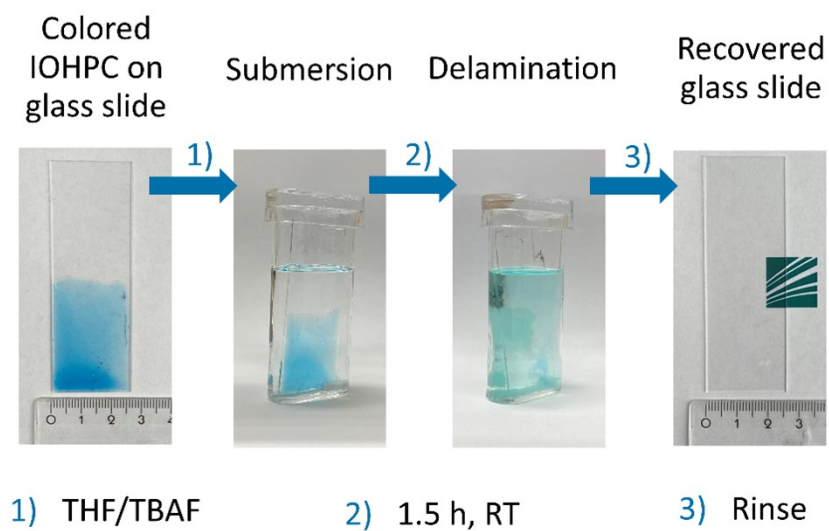


Figure S14: General delamination process of a colored IOHPC on a glass slide. 1) The coated glass slide is submerged in a THF/TBAF solution. 2) After approximately 1.5 hours of stirring at room temperature (RT) the delamination begins. 3) The glass slide is rinsed with THF and dried with a tissue.

References

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