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## Ultra-Durable, Fluorine-free Superhydrophobic Fabrics by Plasma Exploiting Self-healing Wrinkled Skins on Fibers

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Superhydrophobic (SH) fabrics show great potential for functional in gear, clothing, and other use applications; however, the introduction of toxicity and degraded wearing comfort restrict their applications. However, SH fabrics are not durable against laundering where the surface coatings can be easily destroyed by abrasion[1]. Inspired from earthworms with excellent soil repelling and deformation adaptability, here, we report ultra-durable SH fabrics by exploiting the formation of wrinkled poly(dimethylsiloxane) (PDMS) skins on fabric fibers. Uniform wrinkles are created due to the surface instability of PDMS coating induced by Ar-plasma generated gradient crosslinking. Both the surface topographies of the wrinkles and the viscoelasticity of the underlying compliant layer endowed the treated fabrics with extraordinary superhydrophobicity durability, withstanding 800 standard laundries or 1000 rubbing cycles under 44.8kPa. Additional, SH fabrics are self-healable after heating or plasma treatment. Our approach paves a new way to design ultra-durable and fluorine-free fabrics by engineering soft skins on fabric fibers with periodic sub-micron surface topography and modulus gradient.

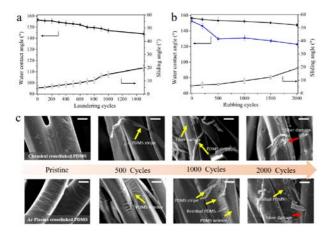


Figure1 a, WCAs and SAs of Ar-plasma PDMS treated fabrics with different laundering cycles. b, WCAs and SAs of fabric treated by different treatment methods under rubbing at a pressure of 44.8 kPa. Black line: Ar-plasma crosslinked PDMS,blue line:thermally crosslinked PDMS. c, Surface morphologies of thermally crosslinked and Ar-plasma crosslinked PDMS on PET fibers under different rubbing cycles (44.8 kPa).

The Ar plasma treated PET fabric exhibited exceptional laundering durability, maintaining superhydrophobicity

after at least 800 cycles according to a standard procedure (AATCC 61-2006-2A); even after 1500 laundering cycles, WCA was still as high as ~145° and the sliding angle (SA) was ~ 20° (Figure 1a). In contrast, SH fabric coated by commercial fluorine-free waterproofing agents such as Ruco®-Dry Ecoplus (Rudolf GmbH) and ZelanTM R3 (Chemours Chemical Co. Ltd.) could only bear 300 laundering cycles. Our PDMS wrinkle coated PET fabric is even superior than fluorine-containing material coated fabrics in terms of washing durability. To elucidate the reason behind the unprecedented laundering performance of our fabrics, we imaged the PDMS coated PET fibers after laundering.

To quantitatively explore the mechanical failure mechanism and self-healing behaviors of wrinkled PDMS coating on fibers, we performed dry mechanical rubbing based on the AATCC 8-2007 colorfastness method with high pressure of 44.8 kPa (ISO 105-X12:2001)4 for accelerating coating damage, while normally 12 kPa was used for abrasion test. Ar-plasma treated fabric exhibits a WCA of ~152° and SA of ~9° after 1000 rubbing cycles, while SH fabric coated by conventional, thermally crosslinked PDMS lost their superhydrophobicity after 200 rubbing cycles (Figure 1c). Clearly, the difference in mechanical failure is originated from the difference in PDMS coating. Based on the evolution of PDMS morphologies after rubbing in Figure 1c, deformation of Ar-plasma crosslinked PDMS coating appeared in two ways, reversible wrinkle relaxation under light load and irreversible surface flattening and fracture of the stiff layer under heavy load.

These results are consistent with the partial damage of the stiff layer and retainment of compliant PDMS layer on gradient PDMS coated fibers. Together, the winkled surface and gradient PDMS coating provide mechanisms to alleviate stress concentration, leading to high durability in laundering and the daily abrasion.

## References

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Super-hydrophobic Surface from a Hierarchically Wrinkled Nanoporous Polymer. ACS Appl Mater & Interfaces. 2019; 11, 40875-40885.