## **Supporting Information**

## Large Scale Monolithic Methyltrimethoxysilane Aerogels Formed by Self-Reinforcement

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Figure S1. An unmodified MTMS aerogel sample showing cracking



Figure S2. An unmodified MTMS aerogel sample showing cracking



Figure S3. An unmodified MTMS aerogel sample showing cracking



Figure S4. An unmodified MTMS aerogel sample showing cracking



Figure S5. An unmodified MTMS aerogel sample showing cracking

## **Synthesis of MTMS Aerogels with Surfactants**

The MTMS-based silica aerogels were fabricated by a two-step base catalysed sol-gel process, followed by ambient pressure drying. MTMS, CTAB or SDS, Ethanol and H<sub>2</sub>O were used as precursor, surfactant and solvent respectively. Initially, the MTMS (5 mL) was dissolved in a mixture of ethanol/water (15 mL/10 mL). CTAB or SDS (amounts between 0.1 and 0.5 g) was then added to the MTMS solution followed by the addition of acetic acid (1 mL, 0.1 M). The MTMS solution was then left to stir for a minimum of 30 minutes and kept in a 45 °C water bath. After sufficient hydrolysis, NH<sub>3</sub>OH (2 mL, 1M) was added to the mixture with stirring. During this stage, the temperature of the water bath was increased to 60 °C. The formed wet gel was then left in the water bath for an hour whilst keeping the container sealed. Finally, the wet gel was dried at 80 °C overnight in an oven to obtain monolithic MTMS based silica aerogels.

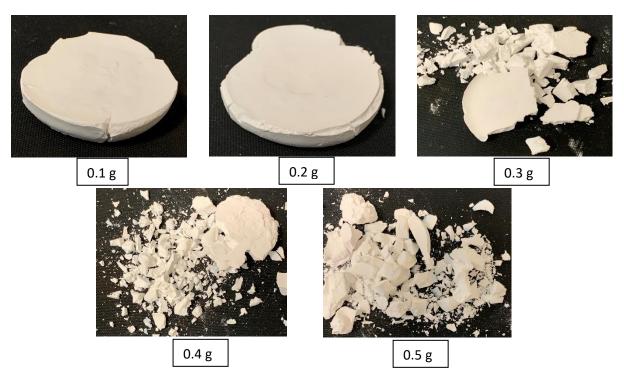


Figure S6. MTMS aerogel pieces formed using various amounts of CTAB (masses shown). Discs prepared have a c.a. 5 cm diameter.



Figure S7. SRA sample showing a moulded central cylindrical section



Figure S8. An SRA formed using three repeated recycles of the aerogel material



Figure S9. Sample of in tact hydrophilized aerogel sitting on top of a burning hydrophobic aerogel after treatment with a blowtorch