Electronic Supplementary Material (ESI) for Soft Matter.
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Electronic Supplementary Information (ESI)

Observation on chemically protected polydimethylsiloxane: towards crack-free PDMS

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Crack density analysis

To evaluate the crack density on the surface of the samples, images of 8 randomly selected regions of the surface of each sample are analyzed using a custom MatLab® script (MATLAB®, R2016b). This analysis leads us to the total length of the cracks in each image. Dividing this length by the area of the image is defined as the *local* crack density. The crack density is then simply defined as the average of the local values, with the standard deviation of the local values as the error.

In the following, the details of the analysis code is provided through a stepwise example. The code uses Image Processing Toolbox, version 9.5 (R2016b). The following parameters are defined and used in the code. The exact same values have been used to analyze all the data. Their values are manually optimized according to the outputs of each step.

```
gfilt = 20;
bulk = 40;
thresh = 0.07;
threshrange = [0.1, 0.3];
scale = 1 / 370;
```

A region of O_2 plasma treated PDMS with the area of 5.74 mm^2 is shown in Fig. S1 (a). The background is not homogeneous and the cracks do not exhibit a unique thickness or visibility. As the first step, the colored image, `img`, is transformed to grayscale and the range of values are broadened to span all the range. The output is shown in Fig. S1 (b).

```
img = rgb2gray(img);
img = imadjust(img, double([min(img(:)); max(img(:))])/255, []);
```

Now the cracks are expressed stronger. But so does the inhomogeneous background. Moreover, there are some out-of-focus artifacts that need to be eliminated. A Gaussian filter is convenient choice for both purposes as shown in Fig. S1 (c)

```
img = imgaussfilt(img, gfilt) - img;
```

While the crack pattern has remained intact, the elimination of the artifacts and the background is almost completely done. In the following lines a binary mask is defined, the small isolated parts of it are removed and the enhanced mask is applied to the image. The output can be seen in Fig. S1 (d). In this example, the output is just slightly different from the previous step. In fact this step is most effective when multiple isolated bright spots, for instance due to the existence of microscopic dust particles on the surface, need to be removed from the image.

```
bw = imbinarize(img, thresh);
bw2 = bwareaopen(bw, bulk, 8);
img(bw & ~bw2) = 0;
```

Finally, the edges are detected using Canny method, which detects both strong and weak edges based on the local maxima of the gradient of the image. The weaker edges are considered only when they are connected to stronger edges. This feature reveals thin branchings while neglecting most of the background. The second function transforms 8-connectivities to 4-connectivity, which results in a more isotropic relation between the physical length of a crack and the number of edge pixels. The final output of the image processing is shown in Fig. S1 (e).

```
img = edge(img, 'Canny', threshrange);
img = bwmorph(img, 'diag');
```

According to the final image, the crack length is calculated as follows:

```
crack_length = sum(img(:)) * scale / 2;
```

The division by two is due to the two edges detected on both sides of each crack. The same process is performed automatically for all the data.

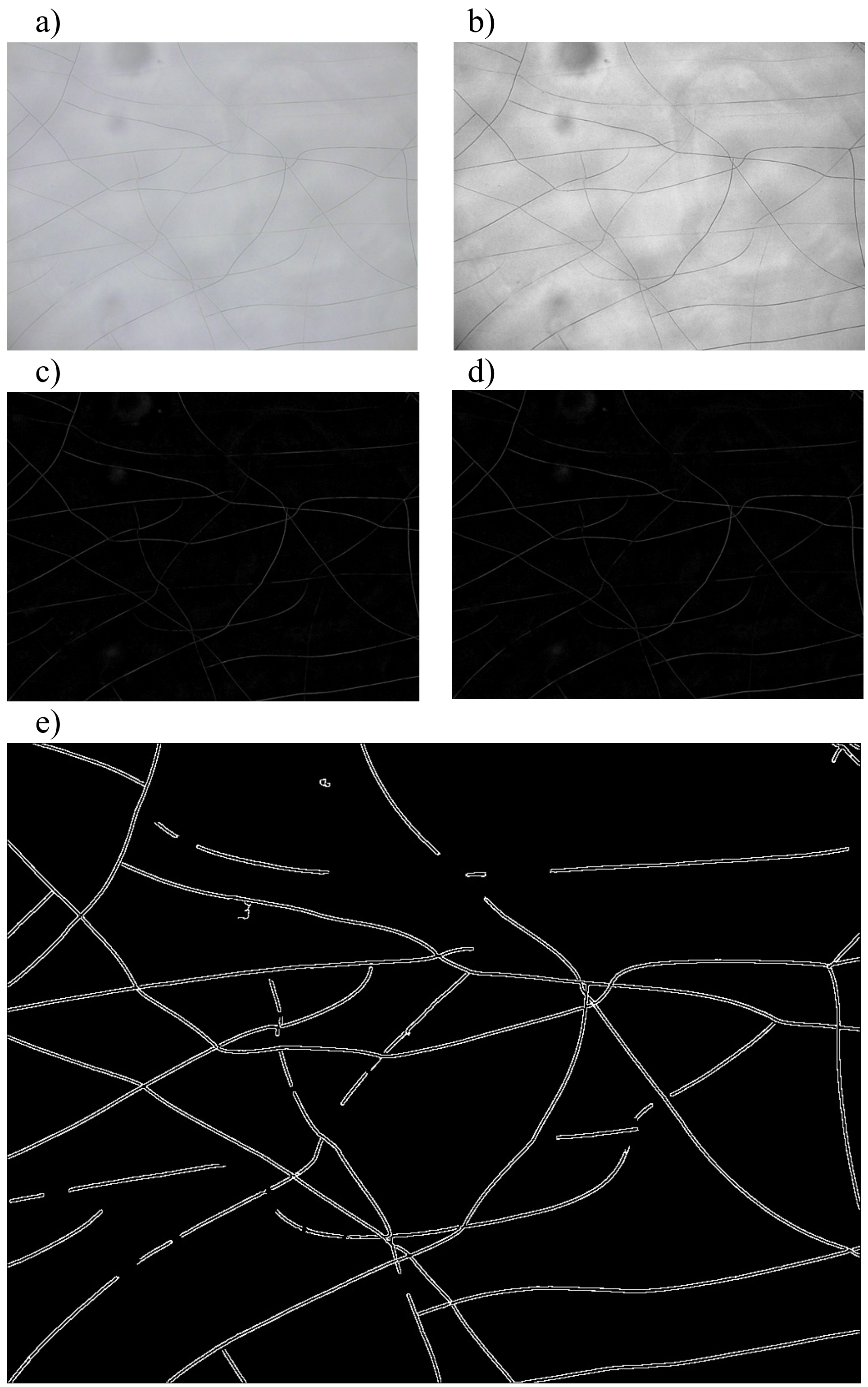

Fig. S1 The initial image (a) is transformed to grayscale and the range is adjusted (b) then a Gaussian filter is applied to remove the background, while keeping the cracks as sharp as they were (c). The possible small islands are eliminated (d) and finally the edge detection is performed (e).

Energy-dispersive X-ray spectroscopy

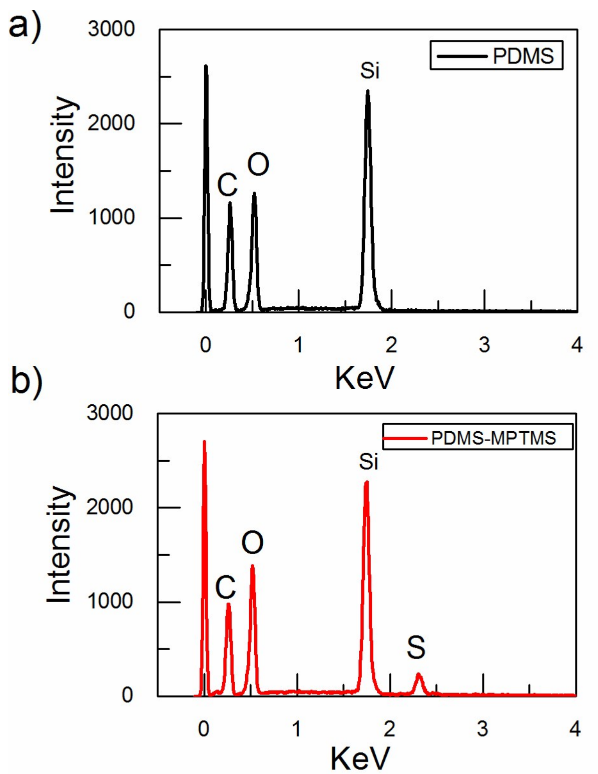

Fig. S2. Elemental Analysis of the (a) pristine PDMS and (b) PDMS–MPTMS. The analysis verify the presence of surface coating of PDMS with MPTMS due to the presence of the sulfur peaks at 2.3 and 2.46 KeV.