Supporting Information

Highly stretchable dielectric elastomers composites containing high silver nanoparticles volume fraction

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Figure S1. TEM micrographs of the AgNPs prepared with the polyol synthesis at 130 °C. Size distribution histogram is inserted on the right.
Figure 2S. XRD spectrum of the AgNPs prepared by polyol synthesis at 130 °C. The typical cubic silver diffraction was observed.

Figure 3S. TEM micrographs of the Ag@SiO$_2$ produced via upscaling through the addition of 0.6 vol% ethanolic TEOS solution into the reaction mixture.
Figure 4S. TEM micrographs of the AgNPs (left) and Ag@SiO$_2$ core-shell particles (right) and their corresponding EDX spectrum below.

Figure 5S. DLS spectra of AgNPs (□), silica-coated particles Ag@SiO$_2$ (○), and of HMDS surface treated core/shell particles Ag@SiO$_2$@Si(CH$_3$)$_3$ (Δ).
**Figure 6S.** Normalized UV-vis of AgNPs (□), silica-coated particles Ag@SiO$_2$ (○), and of HMDS surface treated core/shell particles Ag@SiO$_2$@Si(CH$_3$)$_3$ (Δ). A slight bathochromic shift is observed as a result of the change in medium from PVP/EtOH to SiO$_2$.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Elemental analysis</th>
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<tbody>
<tr>
<td></td>
<td>C [%]</td>
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<tr>
<td>Ag@SiO$_2$</td>
<td>1.11</td>
</tr>
<tr>
<td>Ag@SiO$_2$@Si(CH$_3$)$_3$</td>
<td>1.75</td>
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**Figure 7S.** Elemental analysis of the core-shell particles before and after surface treatment with HMDS.
Figure 8S. XPS spectra of AgNPs prepared with the polyol synthesis at 130 °C. The peak indicative for the oxidized silver species is missing.

Figure 9S. The mechanical properties of B_{20} aged at room temperature for 10 days and at 150 °C for 5 days: the cyclic (left) and the stress-strain (right) tests.
**Figure 10S.** The strain reloading stress-strain curves after initial strain excursions of 50, 75, 100, all the way up to 450% for $B_{14}$.

**Figure 11S.** Permittivity as function of filler content of the composite of series A and B.
**Figure 12S.** Breakdown field $E_B$ as function of filler content of the composite of series A (black) and B (red). The averaged values are depicted with error bars and were obtained from the measurements using the electrodes with an area of $1 \text{ mm}^2$.

**Figure 13S.** Electrodes used for breakdown strength measurements (left and right). Two conductive plates with a thickness of $1 \text{ mm}$ were embedded in an epoxy matrix. The embedded electrodes were polished to a smooth surface. The thickness of the sample is measured prior the breakdown measurement. The two electrodes are used as shown on the left. The overall set-up of the system is depicted on the right.
The thicknesses of the samples used for the dielectric measurements were below 50 µm and were measured using a Heidenhein high accuracy gage. The samples were measured using Hewlett Packard 16451B dielectric test fixture equipped with round electrodes (left). To ensure a good contact and a uniform “pressure” over the samples (because of the round electrode used), gold plates slightly smaller than the coated electrodes were used. The setup shown above (left), allows a tender touch of the sample by the top electrode that is connected to a micrometer. So, there should be no displacement or pressure in the samples. The sample coated with 21 mm electrode was also measured using the setup with which the Novocontrol Alpha-A Frequency Analyzer is equipped (right) and the dielectric results were similar to those obtained using the other setup. Because rather thin samples were used, which were coated with conductive layers of defined surface, even when the right setup was used and more pressure was applied to the sample by screwing, no change in the permittivity was observed.