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Material Behaviour

On the cyclic deformation behavior, fracture properties and cytotoxicity of silicone-based elastomers for biomedical applications

L. Bernardi [a, *], R. Hopf [a], D. Sibilio [a], A. Ferrari [c], A.E. Ehret [a, b], E. Mazza [a, b]

[a] ETH Zurich, Institute for Mechanical Systems, 8092 Zürich, Switzerland
[b] Empa, Swiss Federal Laboratories for Materials Science and Technology, 8600 Dübendorf, Switzerland
[c] ETH Zurich, Laboratory for Thermodynamics in Emerging Technologies, 8092 Zürich, Switzerland

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ABSTRACT

This paper provides results from a comprehensive experimental characterization on five silicone-based elastomers used as substrates for mechanobiological studies or in soft biomedical implants. A previous paper was recently published which focused on the large strain deformation behavior of these materials. This second part analyzes their reliability for biomedical applications in terms of changes of deformation behavior with the history of loading (long term cyclic behavior), ability to resist loads in the presence of defects (fracture properties), and cytotoxicity. For the latter, all materials are confirmed to be non-toxic which is a prerequisite for their use in mechanobiological studies or as part of implants and biomedical devices. The response in long term uniaxial tests over 220′000 cycles was characterized and the results indicate general stability of the mechanical response with, for some conditions, softening mechanisms active mainly in the initial phase of the test (50′000 cycles). A critical aspect of elastomer performance and their suitability for application in biomedical devices concerns their fracture properties. The tearing energy varies in a range from brittle (with approximately 80 J/m^2 for PDMS Sylgard 184) to tough (with approximately 900 J/m^2 for SMI G/G 0.020).

1. Introduction

This article presents the second part of a comprehensive experimental campaign for the characterization of five silicone-based elastomers, i.e. two types of PDMS (Sylgard 184 and Sylgard 186, Dow Corning), and three RTV elastomers vulcanized at room temperature (SMI G/G 0.020", Specialty Manufacturing Inc.; RTV 4528 and RTV 4420, Blue Stars Silicones). These materials were shown to cover a range of mechanical characteristics representative of those of the elastomers commonly used for mechanobiological studies and biomedical devices. The first part of this study [1] provided experimental data and corresponding model equations for the multiaxial monotonic large strain deformation behavior of the elastomers. This paper describes their long term cyclic response, fracture properties and cytotoxicity. Little is reported in previous studies regarding these characteristics, despite their importance for a number of applications. Our companion paper [1] provides a general overview on applications of silicone-based elastomers and the necessity to characterize their deformation response under a variety of mechanical loading conditions. The specific motivation for the analysis of their cyclic response is found in biomedical applications such as for bioreactors mimicking cardiovascular pulsatile conditions [2,3], as part of endovascular grafts and cardiovascular devices [4,5], or other implants exposed to cyclic deformation, e.g. artificial urinary sphincters [6], external shells of breast implants [7] or skin support [8]. In these applications, the mechanical response might change with the history of loading since the long term deformation behavior might differ from that in virgin conditions. This question was addressed here through experiments with >200′000 cycles at nominal strains of up to 30%. Note that the objective of these experiments was not the characterization of the fatigue behavior of the elastomers, which would require a much higher number of cycles. As explained in Ref. [1], the range of deformations considered for the present work were originally motivated by a specific application, viz. as blood propulsion membrane in a pulsatile Ventricular Assist Device (VAD) [9,10].

Another essential feature of materials fulfilling a structural function is their toughness. The ability to resist loads despite the presence of (small) defects is important in particular in view of

* Corresponding author.
E-mail address: bernardi@imes.mavt.ethz.ch (L. Bernardi).

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possible imperfections arising from the manufacturing process. Such defects might affect the mechanical integrity and reliability of biomedical devices or implants. An example is given in the application of silicone elastomers as a joint replacement, e.g. finger joints [11,12], for which fracture initiation and crack growth were analyzed in previous studies [13,14]. The influence of temperature and wet environment (typical for physiological conditions) on the cyclic response and fracture properties of RTV 4420 is also addressed. Finally, in view of the application as implants or substrates for mechanobiological studies, all materials were subjected to cytotoxicity tests.

2. Materials

The characteristics of the elastomers analyzed in this study were described in detail in Ref. [1]. PDMS Sylgard 184, a two component system of base polymer and crosslinker was used in 10:1 ratio; PDMS Sylgard 186 was also used in 10:1 ratio; Silbione® RTV 4528 A&B and Silbione® RTV 4420 A&B are Room-Temperature Vulcanized (RTV) elastomers for which the two components are mixed in 1:1 ratio; SMI G/G 0.020″ also belongs to the RTV family but it is commercially available. All the materials presented in this work are widely used as substrates in mechanobiological studies [15–17], or in soft biomedical implants such as skin-like applications or external maxillofacial prosthetics [18,19].

Samples for long term cyclic and tearing tests were prepared according to the protocol described in Ref. [1]: the elastomer components were mixed by hand for 2–3 min, then degassed and poured into petri dishes of 90 mm diameter. After additional 45 min of degassing, samples were cured in an oven at 60 °C for 4 h, except for RTV 4420 which was cured at room temperature (23 °C). The samples for the long term cyclic tests were rectangular, with a width to length ratio of 1:4, and samples for tearing tests were rectangular with a width to length ratio of 1:6, see Fig. 1.

3. Experimental

3.1. Mechanical testing set-up

The set-up used for both tearing experiments and long term cyclic tests was described in Refs. [1] and [20]. It consists of 2 horizontal hydraulic actuators, 100 N load cells (calibrated for up to 20 N, MTS System, Eden Prairie, USA), a CCD camera (Pike F-100B Allied Vision Technologies GmbH, Stadroda, Germany) with 0.25 × telecentric lens (NT55-349 Edmund Optics GmbH, Karlsruhe, Germany) used for local strain analysis (see Refs. [20,21]).

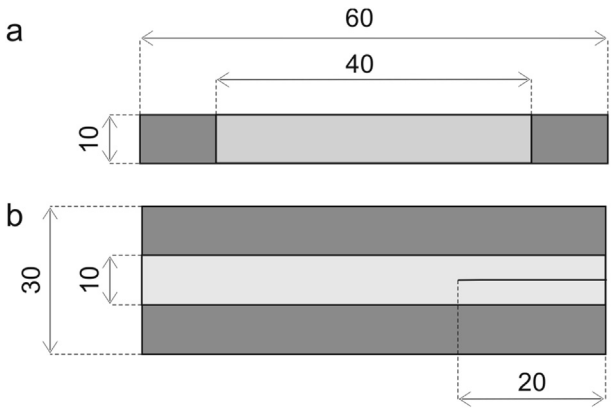

Fig. 1. Sample dimensions in mm for the long term cyclic test (a) and tearing test (b).

Reliable gripping of the sample is provided by gluing a strip of sandpaper onto the surface of the custom-made metal clamps. The clamps are displacement controlled in that a screw is used to apply the initial compression and this remains unchanged during the test. An environmental chamber with a bath, heating coil and a temperature sensor allows testing in wet conditions and at a prescribed temperature (here: 37 °C). Fig. 2 shows the set-up equipped with the environmental bath and the heating coil with a tearing test sample mounted in the clamps.

3.2. Data analysis

Deformation in each sample is reported as engineering strain $\varepsilon = \lambda - 1$, where λ is the stretch in loading direction. Stress is reported as force per unit of initial area (first Piola-Kirchoff stress P). Local strains are calculated from image-based determination of the in-plane deformation. To this end, an optical flow tracker (described in Refs. [1] and [20]) was applied in order to extract the displacement field in the central region of each test piece in order to avoid boundary or clamping effects.

3.3. Effect of the environmental conditions on deformation behavior

Mechanobiological studies as well as many biomedical devices expose the elastomers to a temperature of 37 °C and wet conditions. For example, in cells studies (e.g. Ref. [3]), the silicone membranes are left several days in cell culture medium inside an incubator. In order to mimic these environmental conditions, a few tests were performed in distilled water and saline solution (resembling a cell culture medium) at 37 °C. Saline solutions were prepared with a known concentration of salts in distilled water (0.9 mg/mL NaCl in distilled water [22]). In order to assess the influence of environmental conditions, monotonic uniaxial tests were carried out on RTV 4420 samples left 24 h in distilled water or in saline solution at 37° and compared with results obtained at room temperature in dry conditions.

3.4. Long term cyclic tests

Cyclic tests were performed in uniaxial configuration. Samples were cyclically stretched between 0 and 30% nominal strain at 1 Hz, up to a total of 220′000 cycles. Thus, the total duration of each test was about 60 h and only 1 sample was tested for each material. 200 cycles of initial pre-conditioning at 1 Hz were applied before starting the cyclic experiment in order to stabilize clamping conditions. Forces and images were recorded during the test every 12 h (i.e. every 43′000 cycles) in order to monitor the stress decay associated with the applied local strain for increasing number of cycles. Forces were recorded at a frequency of 512 Hz while images were recorded at a frequency of 20 Hz. A quantitative measure, the reduction in peak stress P_{decay} was calculated normalizing the stress at each time point P_i with respect to the value associated with the applied strain in virgin conditions $P_{initial}$. RTV 4420 was tested also at 37 °C in distilled water; the sample was stored in the corresponding solution and inside an incubator (37 °C) 24 h prior to the test. The incubator was chosen because it is representative of cell cultures.

3.5. Tearing tests

Tearing tests were performed following a protocol based on the original work by Rivlin and Thomas [23]. A cut of 20 mm was created on one of the lateral edges of the test piece (Fig. 1), which was loaded up to the point of crack propagation with a nominal strain rate of 0.3%/s. Fig. 3 shows an example of a sample at the