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A study on the mechanical behavior of silicone-organically modified montmorillonite composite under human body simulated environment

Ji-Hun Bae [a], Seung-Hwan Chang [a,*]

[a] School of Mechanical Engineering, Chung-Ang University 221, Huksuk-Dong, Dongjak-Gu, Seoul 156-756, Republic of Korea

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ABSTRACT

Among electroactive polymers (EAPs), dielectric elastomers such as silicone are strong candidate materials for biomimetic actuators. They have several unique abilities such as high deformation and fast response time under applied electric energy. In this study, the basic mechanical properties of the silicone elastomer were measured in a simulated human body environment to investigate the applicability of this material as an artificial muscle in *in vivo* environment. To enhance the mechanical properties of the silicone elastomer, organically modified montmorillonite (OMMT) particles were added. The silicone–OMMT composite was submerged in 30 °C water for a few weeks to evaluate the influence of moisture absorption on the mechanical properties of the composite. Tensile and creep tests were carried out according to the moisture absorption rate and OMMT particle content. To observe the change of crystalline structure and the level of dispersion of the OMMT in the silicone elastomer matrix, X-ray diffraction (XRD) analysis and scanning electron microscopy (SEM) were employed.

1. Introduction

Electroactive polymers (EAPs) are materials that respond to electrical stimulation by showing significantly large strains (a few hundred %). Since EAPs behave very similarly to muscles, EAPs have acquired the moniker 'artificial muscles' [1]. EAPs have emerged as promising alternative materials for various applications in advanced technologies. Especially, dielectric elastomers, such as silicone have been actively studied as materials for various actuators [2–7]. Even though acrylic elastomers exhibit a very large electrical-activation strain (max. 380%), they have some critical drawbacks; high viscosity, hysteresis, and frequency-dependent electromechanical response. The silicone elastomer, however, shows temperature stability, short response time, reproducible response to an electrical stimulation, even though its activation strain is relatively low (max. 63%). Such performances have made the silicone elastomer a suitable material for sensors and actuators [1,8]. Polymers added with fillers show improved mechanical and electrical properties; therefore, polymer–filler composites have been widely studied [9–11]. Since a clay particle has an especially large aspect ratio, its filling effect in a polymer matrix can be maximized. Moreover, by adding even a small amount of clay particles, the mechanical behaviors of a polymer, such as tensile strength, can be enhanced without degradation of the polymer's intrinsic toughness [12,13].

In this paper, a silicone-organically modified montmorillonite (OMMT) composite was fabricated with various OMMT contents (0%, 2%, and 5%) to investigate its medical applicability as a material for sensors or actuators in *in vivo* environment with no degradation of its innate characteristics. A tensile test and a creep test were performed to evaluate the basic changes in its mechanical property such as Young's modulus after water absorption in 30 °C water for a maximum of 4 weeks.

2. Experiments

2.1. Materials and specimen preparation

To fabricate the silicone–OMMT composite film a silicone elastomer (DC 3481, Dow corning, USA) (Fig. 1a), a hardener (81-F NW, Dow corning, USA) and OMMT particles (Closite 15A, Southern Clay Inc., USA) (Fig. 1b) were prepared. Test specimens were fabricated using a solution intercalation method according to OMMT content (0, 2, 5%). The fabrication process was as follows: (1) The liquid silicone was dissolved in tetrahydrofuran and OMMT particles were mixed by a magnetic stirrer at 1000 rpm at room temperature for 2 h. (2) The hardener was added and the mixture was stirred for 10 min. (3) The well-mixed solution was poured into a specially fabricated mold used for making a specimen (Fig. 1c) based on ASTM D412-06a [14], and cured in room temperature for 10 h. The fabrication method (solution casting method) of the clay/silicone composite was proved to be safe for medical applications [15]. And the biocompatibility of the specimens fabricated by

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* Corresponding author. Tel.: +82 2 820 5354; fax: +82 2 814 9476.
E-mail address: phigs4@cau.ac.kr (S.-H. Chang).

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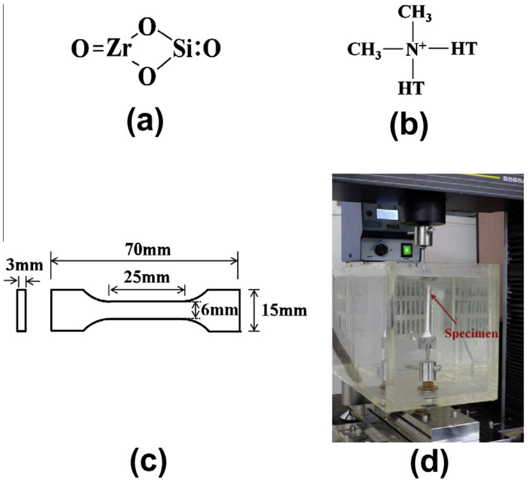

Fig. 1. Chemical compositions of (a) silicone elastomer and (b) OMMT particle. (c) Shape and dimensions of the silicone–OMMT composite specimen. (d) Experimental setup for silicone -OMMT composite in human-body simulated environment.

tetrahydrofuran as a solvent was also examined and verified [16,17]. To prevent void generation in the specimen during the curing and to fabricate a high-quality specimen, absolute pressure of 0.3 MPa was applied in the autoclave. The electro-mechanical behavior of the clay/silicone composite (5% of OMMT contents) was verified by an appropriate pre-test and a reference [18]. We checked the maximum strain of the material was 13% under 7 kV electric stimulation. And biocompatibility of the silicone elastomer and the clay particle was proved by other researchers [19–21].

2.2. Characterization

2.2.1. Moisture absorption

To indirectly investigate the characteristics of the silicone-OMMT composite in *in vivo* environment, the specimens were submerged in 30 °C water in a digital water bath (WiseCircu, WCB-22, Germany) for a maximum of 4 weeks. For the measurement of moisture absorption, the submerged specimens were wiped to remove the surface moisture, and then the wiped specimens were weighed using a digital precision balance (Ohaus, AR2130, Korea) to monitor the change in mass change according to water exposure time. The moisture content, M, absorbed by each specimen was calculated by the following equation:

$$M(\%) = \frac{W_w - W_d}{W_d} \times 100 \tag{1}$$

where W_d and W_w represent the weights of the dry and the wet specimen, respectively. And the equilibrium moisture content, M_m, representing a fully saturated state was also measured. The diffusion coefficient, D, was calculated using Eq. (2) based on the modified Fick's 2nd law [22].

$$D = \pi\left(\frac{h}{4M_m}\right)^2 \left(\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}}\right)^2 \tag{2}$$

where h is the specimen thickness, M_1 and M_2 stand for the moisture contents at times t_1 and t_2, respectively. These time periods were chosen at the early stage of the moisture absorption process, where the weight change can still be assumed to vary linearly with the square root of time.

2.2.2. Mechanical properties

Five specimens were used for the tensile tests at each test condition according to water exposure time (0, 2, 4 weeks). The test speed was 500 mm/min. Tensile and creep tests were performed using a universal testing machine (Instron, 5565A, USA) and an environmental chamber (Fig. 1d) based on the standard test procedures (tensile test: ASTM D412-06a [14], creep test: ASTM D2990-09 [23]). And three kinds of the loads of 1 MPa, 1.5 MPa, and 2 MPa at 2.5%, 3.2%, and 3.7% strains, respectively, from the elastic region of the stress–strain curve of the dry silicone specimen were used to carry out the creep test for 10,000 s [24].

2.2.3. Microscopic observation

An X-ray diffractometer (Bruker-AXS, New D8-Advance, Germany) was employed to observe the change in crystalline structure of the OMMT particle in the polymer matrix due to moisture absorption according to water exposure time. Scan speed and range were 2.4°/min and $2\theta = 10–30°$, respectively. And scanning electron microscopy (SEM) (Hitachi, S3400, Japan) was used to acquire information about the level of dispersion of OMMT particles in the polymer matrix and to observe the fracture surface.

3. Results and discussion

3.1. Moisture absorption

Fig. 2a shows the moisture absorption rate of the silicone–OMMT composites according to OMMT content and water exposure time. All specimens absorbed water rapidly in the initial stage of water exposure and then they showed slightly reduced absorption rates, finally reaching the saturate state. The equilibrium moisture content (M_m) read from Fig. 2a and the measured values were used to calculate the diffusion coefficient (D) by Eq. (2), and the corresponding values including the saturation time (t_s) are listed in Table 1. The kinetics of water diffusion in the silicone–OMMT composites followed the one dimensional Fick's 2nd law, and both moisture absorption rate and diffusion coefficient (D) were proportional to OMMT content [25,26]. This diffusive behavior was due to the bonding between water molecules and the hydrophilic octadecylamine groups, 30% of which are contained in the OMMT particles [27], and this trend was verified by other researchers [28,29]. Furthermore, all specimens expanded with the water exposure time and OMMT content, because OMMT particle is member of a smectite family, which has a large aspect ratio and high swelling coefficient [30]. And the lengths of the specimens recovered to their original dimension after re-drying (Fig. 2b–d).

3.2. Mechanical properties

3.2.1. Mechanical behavior of silicone–OMMT composites by moisture absorption

The tensile test results of the silicone–OMMT composites according to water exposure time (moisture contents) are shown in Fig. 3a–f.

Adding OMMT as reinforcement enhanced in the Young's modulus and tensile strength of the silicone. The silicone–OMMT composites became more brittle as the OMMT content increased. When intercalated or exfoliated structure [31,32] is assumed for the silicone–OMMT composite, the filling effect of the silicone–OMMT composite becomes greater than that of any other composite [12,13,33]. OMMT layers were intercalated by the polymer chains (Fig. 4a) and the bonding between the oxygen atom of liquid silicone having a large electronegativity and the methyl groups of OMMT led to the enhancement of Young's moduli and strengths of the composites [34]. However, the failure strains of the composites decreased with OMMT content. According to Kojima et al. [35] and Manias et al. [36], a region where the polymer chains are