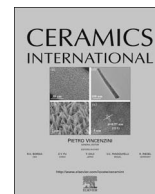




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# Synthesis of refractive index tunable silazane networks for transparent glass fiber reinforced composite

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## ABSTRACT

Organic-inorganic hybrid polymers have recently attracted attention as matrix materials for transparent glass fiber-reinforced composites because of their optical and thermal properties. In this study, a transparent glass fiber-reinforced composite composed of organic-inorganic structures was prepared by hydrosilylation of polysilazanes. To obtain the transparent composite, the refractive index of the silazane matrix was controlled by phenyl groups. Fourier transform infrared spectroscopy was used to identify the chemical structure of the cross-linked polysilazanes. The transmittance of the composite was 89.36%, close to that of the silazane matrix. The 10% weight loss temperature of the matrix resin was determined as 531.9 °C. To observe the mechanical properties of the composite, a three-point bending test was performed. The flexural strength and the flexural modulus of the composite increased by 65% and 52% compared to those of the silazane matrix, respectively. These results suggest that polysilazanes can be utilized for transparent glass fiber-reinforced composites as alternatives to organic polymers.

## 1. Introduction

Composite materials have been widely studied because of their unique properties that cannot be achieved by individual materials [1]. Among composite materials, glass fiber-reinforced composites have been commonly used because of their mechanical, thermal, and electrical properties [2,3]. Recently, glass fiber-reinforced polymer matrix composites have attracted attention for improving the thermal and mechanical properties of organic polymers. They have also been used in various applications such as the covers for electronic devices, transparent substrates, and transparent packages for semiconductors [4–6]. The polymer matrix materials are usually epoxy, vinyl ester, polyester, and thermosetting polymers. However, these organic polymers have thermal stability problems [7,8]. Organic-inorganic hybrid polymers have been proposed to replace the organic polymers because they have both the properties of organic polymers and the high thermal stability of inorganic polymers [4,9–11].

Among various organic-inorganic hybrid polymers, polysilazanes are promising materials because of their low viscosity, excellent adhesion to inorganic materials, and high transparency of their cross-linked form [12,13]. They are silicon-based polymers with Si-N-Si backbone structures and C-H side chains and can be converted to advanced ceramics such as SiC, SiN, and SiCN after thermal curing [14]. In particular, polysilazanes can be utilized as matrix materials for glass fiber-reinforced composites

because they are in the liquid state with low viscosity at room temperature. Therefore, they can easily infiltrate a filler material such as a glass fiber [12]. However, polysilazanes have only been studied as ceramic precursors. For instance, Li et al. reported a conversion process of polysilazanes to SiCN ceramics and Ionescu et al. explored hafnium alkoxide-modified polysilazane-derived ceramics [15,16]. Moreover, polysilazane-derived ceramics with inorganic fillers have been characterized by many researchers [17,18]. However, the optical and mechanical properties of cross-linked polysilazanes as matrix materials have not yet been fully understood.

In this study, we fabricated a transparent glass fiber-reinforced composite using polysilazanes. To obtain a highly transparent composite material, the refractive index of the silazane matrix was controlled by diphenylsilanediol. In addition, the chosen thermal curing temperature was less than 200 °C. The synthesis of polysilazanes and diphenylsilanediol was investigated, and the optical, mechanical, and thermal properties of the composite were discussed. The results indicate that the thermal stability of the composite using polysilazanes is greater than that of the organic polymers used for plastic substrates.

## 2. Experimental procedure

### 2.1. Preparation of matrix resin

To develop a silazane matrix with a tunable refractive index,

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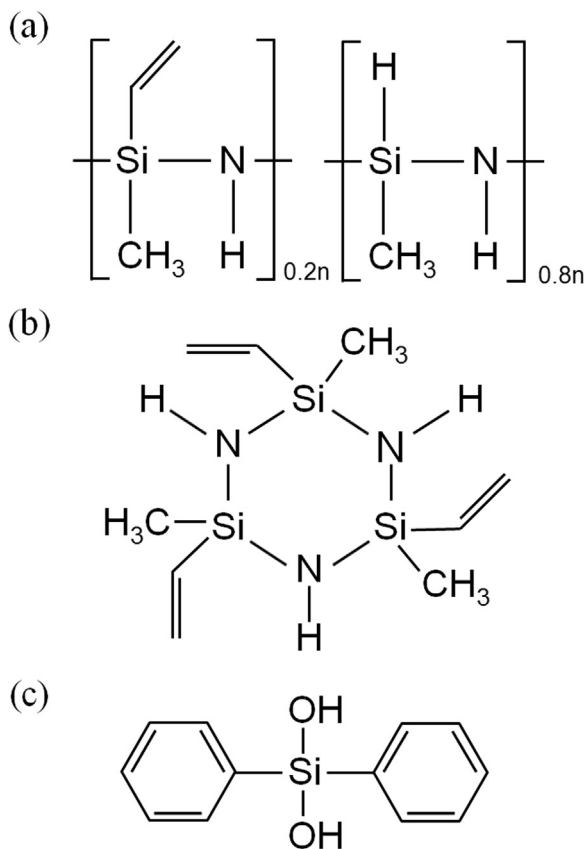


Fig. 1. Chemical structures of (a) OPSZ, (b) CTS, and (c) DPSD.

organopolysilazane (OPSZ, Durazane 1800, AZ electronic materials, Japan) as a precursor, 1,3,5-trimethyl-1,3,5-trivinylcyclotrisilazane (CTS, Tokyo Chemical Industry, Japan) as a cross-linking agent, and diphenylsilanediol (DPSD, Tokyo Chemical Industry, Japan) as a refractive index modifier were used (Fig. 1). They were mixed at various ratios as shown in Table 1. Then, dicumyl peroxide (DCP, Sigma Aldrich, USA) and platinum(0)-1,3-divinyl-1,1,3,3-tetramethyl-disiloxane complex solution (Karstedt's catalyst, Sigma Aldrich, USA) were added into the final mixture as a thermal initiator and a catalyst for hydrosilylation, respectively.

## 2.2. Silane surface treatment of E-glass chopped strand mat

The E-glass fiber chopped strand mat (CSM, Hankuk Fiber Group, Korea), which consists of glass fibers laid randomly across each other, was burned at 550 °C for 3 h. The diameter and relative density of CSM were 9 μm and 2.57, respectively. 3-Aminopropylethoxysilane (Sigma Aldrich, USA) was used for the surface treatment of CSM. A 3 wt% aminosilane solvent (ethanol/water, 95:5) was prepared and the heat-

cleaned CSM was immersed in the aminosilane solvent at 20 °C. After 30 min, the CSM was dried at 110 °C for 10 min and then at room temperature for 24 h.

## 2.3. Fabrication of Glass Fiber Reinforced Silazane Matrix (GFRSM)

The silane-treated E-glass CSM was cut into 50 mm×50 mm sheets (volume fraction: 0.1) and one of the sheets was placed on a Teflon mold. The matrix resin was poured on the CSM sheet. To control the thickness of the GFRSM, the Teflon mold was covered with another Teflon mold and they were placed in a vacuum oven (Ajeon Heating Industrial Co., Ltd., Korea) for further impregnation and thermal curing. The impregnation process was continued until entrapped air and excess matrix resin were removed under vacuum. The sample was cured at 110 °C for 6 h under vacuum conditions and post-cured at 200 °C for 4 h under a nitrogen atmosphere. After the thermal curing process was completed, the GFRSMs were released from the Teflon mold.

## 2.4. Characterization

Fourier transform infrared spectroscopy (FT-IR, VERTEX 80 V, Bruker, Germany) was performed to observe the chemical conversion of GFRSMs after thermal curing. The chemical conversion of GFRSMs was compared to that of the pure silazane matrix (PSM, OPSZ/CTS, 4:1). The refractive index of the silazane matrix containing various amounts of DPSD was measured using an Abbe refractometer (DR-M2, Atago, USA). The transmittance of GFRSMs and PSM was observed by a UV-visible spectrophotometer (UV-2450, Shimadzu, Japan). The thickness of all samples was 1 mm. A three-point bending test was conducted using a micro material tester (load accuracy: 0.5%, strain measurement accuracy: 0.5%) (Instron 5848, Instron Corp., USA) to analyze the flexural stress, strain, and modulus of the GFRSM with the highest transparency and PSM according to ASTM D790-15 with a 25-mm span length and a cross head speed of 1 mm/min. The test was performed five times for each material and the average value was calculated. The flexural stress ( $\sigma_f$ ), strain ( $\epsilon_f$ ), and modulus ( $E_B$ ) of the GFRSM and PSM were calculated as follows [19]:

$$\sigma_f = \frac{3PL}{2bd^2} \quad (1)$$

$$\epsilon_f = \frac{6Dd}{L^2} \quad (2)$$

$$E_B = \frac{L^3m}{4bd^3} \quad (3)$$

where  $P$  is the applied load (N),  $L$  is the span length between the supports (mm),  $b$  is the width of the specimen (mm),  $d$  is the depth of the specimen (mm),  $D$  is the maximum deflection of the center of the specimen, and  $m$  is the slope of the tangent to the initial straight-line portion of the load-deflection curve (N/mm). After the test, the fracture surfaces of the GFRSM and PSM were compared using field emission scanning electron microscopy (SEM, S-4300, Hitachi, Japan). The thermal stability of the GFRSM was determined using thermogravimetric analysis (TGA 209 F3, NETZSCH, Germany) and high temperature differential scanning calorimetry (DSC 404 F1, NETZSCH, Germany) at a temperature increase rate of 10 °C/min under a nitrogen atmosphere.

## 3. Results & discussion

### 3.1. Synthesis and characterization of GFRSMs

The chemical structure of the PSM and GFRSMs was observed after curing at 200 °C (Fig. 2). The FT-IR spectra of the PSM and the

Table 1  
Chemical composition of matrix resin.

Samples	OPSZ: CTS: DPSD Mass ratio
GFRSM1	4.0: 1.0: 3.0
GFRSM2	4.0: 1.0: 3.5
GFRSM3	4.0: 1.0: 4.0
GFRSM4	4.0: 1.0: 4.5
GFRSM5	4.0: 1.0: 5.0
GFRSM6	4.0: 1.0: 5.5

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