Material performance

Effect of urea-containing anti-tracking additive on the tracking and erosion resistance of addition-cure liquid silicone rubber

Weizhen Fang [a], Xuejun Lai [a], Hongqiang Li [a], Wanjuan Chen [a], Xingrong Zeng [a,*], Liping Zhang [b], Siguang Yang [b]

[a] College of Materials Science and Engineering, South China University of Technology, Guangzhou 510640, China
[b] Guangzhou Tinci Silicon Technology Co., Ltd., Guangzhou 510760, China

ABSTRACT

Three novel urea-containing anti-tracking additives were prepared and used to enhance the tracking and erosion resistance of addition-cure liquid silicone rubber (ALSR). The suppression effect and mechanism of the additives on tracking and erosion were investigated by the inclined plane (IP) test, thermogravimetry (TG) and thermogravimetry-Fourier transform infrared spectrometry (TG-FTIR). It was found that the anti-tracking additives not only significantly improved the tracking and erosion resistance but also enhanced the mechanical properties of the ALSR. When using 2 phr of the anti-tracking additive, all the ALSR samples could pass the 4.5 kV level in the IP test. This might be mainly attributed to the arc-quenching capability of the urea group and the formation of a ceramic barrier to protect the underlying materials from further degradation during the dry band arcing. The TG and TG-FTIR results revealed that the urea-containing anti-tracking additives could prevent the radical decomposition of the ALSR below 400 °C. Hence, it considerably improved the initial thermal degradation stability of the ALSR and prolonged the time to track.

© 2014 Elsevier Ltd. All rights reserved.

1. Introduction

Silicone rubber is widely used in polymer insulators, bushings and cable terminations, owing to its light weight, high vandal resistance and capability to maintain and recover surface hydrophobicity in wet and contaminated conditions [1–3]. However, silicone rubber in contaminated applications is susceptible to dry band arcing, partial discharge and corona discharge due to various environmental and electrical stresses. In the long term, these electrical discharges could cause degradation of the silicone rubber in the form of electrical tracking and erosion,

threatening the reliability of the insulation. Of the discharges mentioned above, dry band arcing is considered to be the most damaging, leading to tracking and erosion of a material in a much faster way than corona discharge [4–7]. To enhance the tracking and erosion resistance of silicone rubber, many inorganic fillers have been incorporated into the matrix, including alumina trihydrate (ATH) [8,9], silica (SiO_2) [10,11], alumina (Al_2O_3) [12], titanium dioxide (TiO_2) [13] and barium titanate ($BaTiO_3$) [14]. Typically, ATH is very efficient in improving the tracking and erosion resistance of silicone rubber. When the local hot spot temperature from dry band arcing exceeds 220 °C, ATH begins to release the hydrate, which cools the silicone rubber surface and serves as an arc extinguishing gas, thereby reducing the damage caused by electrical arcs [15,16]. However, to achieve the required electrical properties, a large amount of

ATH (>50 wt.%) is necessary, which severely worsens the processability and mechanical properties of the silicone rubber [17].

To reduce the inorganic filler content, one solution is to incorporate arc-quenching materials into the silicone rubber compound. Arc-quenching materials, such as high nitrogen organic compounds [18,19] and gas-evolving polymers [20,21], have the capability to rapidly evolve gases during arcing, and the gases could change the arcing medium and produce a deionizing action and a cooling effect on the arc, which facilitates arc extinction. This has good potential for introducing arc-quenching properties to silicone rubber, resulting in enhancing the resistance to tracking and erosion. Lars and coworkers [22] combined melamine cyanurate with silica to improve the tracking and erosion resistance of silicone rubber and all the samples passed the IP test at 4.5 kV. This might be because the melamine cyanurate could extinguish the electrical arc. However, in their work, melamine cyanurate was directly added to the silicone rubber, which limited the further improvement of the mechanical properties and the tracking and erosion resistance of the silicone rubber due to the poor compatibility between the melamine cyanurate and polymer matrix. Therefore, it is of great interest to prepare a reactive anti-tracking additive possessing an arc-quenching group, and clarify the relationship between the arc-quenching capability and the tracking and erosion resistance of silicone rubber.

In this work, three types of anti-tracking additives containing vinyl and urea group, (γ-ureidopropyl) allyloxyethoxysilane (UPAS), (γ-isopropylureidopropyl) allyloxyethoxysilane (IPUPAS) and (γ-diisopropylureidopropyl) allyloxyethoxysilane (DIPUPAS) were prepared, as shown in Fig. 1. They were used to enhance the tracking and erosion resistance of addition-cure liquid silicone rubber (ALSR). The effects of the anti-tracking additives on the tracking and erosion resistance, mechanical properties and thermal properties of the ALSR were investigated Moreover, the possible suppression mechanism of the anti-tracking additives on the tracking and erosion of the ALSR was further explored. Through the investigation, we expect to develop a new way to prepare an ALSR with excellent tracking and erosion resistance and good mechanical properties.

2. Experimental

2.1. Materials

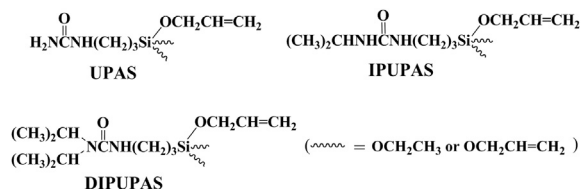Allyl alcohol (AA) was supplied by Xiya Chemical Reagent Co., Ltd., China. (γ-Ureidopropyl) triethoxysilane (UPES) was purchased from Degussa Chemical Co., Ltd., Germany. (γ-*Isopropylureidopropyl*) *triethoxysilane* (IPUPES) and (γ-*diisopropylureidopropyl*) *triethoxysilane* (DIPUPES) were synthesized in our laboratory according to reference literature [23]. The chemical structure of IPUPES was determined by ^{1}H NMR (CDCl$_3$, ppm): 1.1–1.3 (m, –CH$_3$, 15H), 3.8–3.9 (m, Si–OCH$_2$, 6H), 4.3(s, –CONH–, 1H), 4.6 (s, –NHCO–, 1H); and ^{1}H NMR (CDCl$_3$, ppm) of DIPUPES: 1.1–1.3 (m, –CH$_3$, 21H), 3.8–3.9 (m, Si–OCH$_2$, 6H), 4.3 ppm (s, –CONH–, 1H). Toluene was obtained from Guangzhou Chemical Reagent Factory, China. Tetrabutyl titanate (TBT) was purchased from Tianjin Fuchen Chemical Reagent Factory, China. Vinyl-terminated poly(dimethylsiloxanes) (VPDMS, viscosity was 2000 mPa s, vinyl content was 0.23 wt.%), ploy(hydromethylsiloxane) (PHMS, viscosity was 160 mPa s, hydride content was 0.70 wt.%), and platinum(0)-1,3-divinyl-1,1,3,3-tetramethydisiloxane complex (Karstedt's catalyst, 3000 ppm) were supplied by Guangzhou Tinci Silicon Technology Co., Ltd., China. Fumed silica (M-5) was purchased from Cabot Co., Ltd., USA. 1-Ethynylcyclohexanol as inhibitor was offered by Shenzhen Xinzeye Technology Co., Ltd., China.

2.2. Preparation of the urea-containing anti-tracking additives

AA (0.30 mol) and TBT (0.5% of the total mass of reactants) were dissolved in 60 mL toluene, and the mixture was heated to 70 °C. UPES (0.10 mol) was added dropwise to the mixture, and the reaction lasted for 2 h at 70 °C. Then, the reaction was maintained for another 2 h under the reduced pressure and the toluene was removed to obtain the final product, designated as UPAS. The vinyl group content in UPAS was 22.1 wt.%, which was determined by iodometric titration [24]. The chemical structure of UPAS was determined by ^{1}H NMR (CDCl$_3$, ppm): 1.1–1.3 (m, –CH$_3$, 3H), 3.8–3.9 (m, Si–OCH$_2$, 6H), 4.3 (s, –CONH–, 1H), 4.9 (s, NH$_2$CO–, 2H), 5.0–5.4 (m, –C=CH$_2$, 4H), 5.8–6.0 (m, –CH=C–, 2H).

Following a similar procedure but using IPUPES (0.10 mol) or DIUPES (0.10 mol) instead of UPES, the product designated as IPUPAS (vinyl content was 17.7 wt.%) or DIUPAS (vinyl content was 14.7 wt.%) was obtained, respectively. ^{1}H NMR (CDCl$_3$, ppm) of IPUPAS: 1.1–1.3 (m, –CH$_3$, 9H), 3.8–3.9 (m, Si–OCH$_2$, 6H), 4.3 (s, –CONH–, 1H), 4.6 (s, –NHCO–, 1H), 5.0–5.4 (m, –C=CH$_2$, 4H), 5.8–6.0 (m, –CH=C–, 2H). ^{1}H NMR (CDCl$_3$, ppm) of DIPUPAS: 1.1–1.3 (m, –CH$_3$, 15H), 3.8–3.9 (m, Si–OCH$_2$, 6H), 4.3 (s, –NCONH–, 1H), 5.0–5.4 (m, –C=CH$_2$, 4H), 5.8–6.0 (m, –CH=C–, 2H).

2.3. Preparation of the ALSR

VPDMS, fume silica, PHMS, anti-tracking additive and 1-ethynylcyclohexanol were stirred vigorously to mix well, and then the Karstedt's catalyst was added and thoroughly mixed. Finally, the mixture was vulcanized at 120 °C for 10 min under 8 MPa to obtain the ALSR samples. The formula of the ALSR is listed in Table 1.

Fig. 1. Chemical structures of UPAS, IPUPAS and DIPUPAS.