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Influence of Chemical Design of Insulating Silicone Compounds on Hydrophobic and Electrical Behavior

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Abstract

The aim of our investigation is to optimise a two-component liquid silicone rubber (LSR) as a housing and shield material for composite insulators. The investigation focuses on improving erosion resistance, tracking resistance and hydrophobicity transfer. The results show that these properties can be achieved by adding aluminium hydroxide in high concentrations at the expense of flow and mechanical properties of the LSR. A more practical solution is to optimise the network structure of the LSR. This chemical modification means that excellent erosion and tracking resistance and outstanding hydrophobicity transfer can be achieved without reducing flow and mechanical properties.

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Introduction

Composite insulators and surge arresters where the housing and shields are made of silicone elastomers are becoming increasingly important on the insulator market (A, B). Silicone elastomers have excellent insulating properties and low leakage currents of the composite insulators compared to porcelain insulators. Silicone elastomers exhibit regenerative hydrophobicity after influence of moisture and corona discharges and hydrophobicity transfer to external layers or pollution deposits on the surface of insulators. Tests on a 20-year-old silicone insulator in a 420 kV network have shown complete hydrophobicity remains (C). Field investigations on Nordstrand Island have shown hydrophobicity transfer was completely functional (D) after a number of years' use. In addition to hydrophobicity, the erosion and tracking resistance of silicone elastomers is extremely important (E) for use in high-voltage insulators. Today's market requires products that surpass the inclined plane test according to DIN IEC 587 at a test voltage of 4.5 kV.

GE Bayer Silicones has been working with the HTWS Zittau / Görlitz on research aimed at developing two-component liquid silicone rubbers (LSR) with high erosion and tracking resistance under the influence of high voltage while maintaining rapid hydrophobicity transfer to external layers. The mechanical and flow properties of the LSR should also be retained. This report demonstrates how composition of the LSR influences different properties.

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Advantages of LSR

Silicone elastomers can be divided into three groups, with a distinction being made between single component solid rubbers (HCR) and two-component liquid silicone rubbers. The liquid silicone rubbers can be divided into systems cross linking at room temperature (RT2) and those cross linking at higher temperatures (LSR). The differences consist essentially in the chain length and thus the viscosity of the polymers used and the content of reinforcing filler (see [Fig. 1](#)). In contrast to LSR, RT2 does not contain any inhibitor, so the cross linking reaction starts directly after mixing the two components at room temperature. With LSR, the inhibitor prevents cross linking at room temperature. Typical pot life is more than 3 days. However, the cross linking reaction with LSR at temperatures above 90 °C is very fast.

Today, solid rubbers with high filler content are still largely used in composite insulators. In addition to the reinforcing silica, a high amount (by weight) of aluminum hydroxide (ATH) is used as a second filler in solid rubber. The ATH leads to increased erosion and tracking resistance of the HCR (E). However, there are disadvantages to ATH addition. Mechanical properties and in particular, tear resistance, drop sharply. The compounds filled with a high level of ATH tend to produce high mold tack. Adding a mold release agent must compensate this effect.

In addition to HCR with a high filler content, LSR is becoming increasingly important for shielding composite insulators. Predominant reasons for this are (see [Fig. 2](#)): the high degree of automation and short cycle times resulting in high productivity, good mold release properties, high tear resistance at high temperatures and low temperature and pressure stress of inserts (e.g. for the production of surge arresters and long-rod insulators).

LSR can be processed under very clean conditions. A production process, as shown schematically in [Fig. 3](#), can be used to manufacture large volume parts (F). Components A and B are fed by means of two drum pumps into two large-volume cylinders. A third component, such as a pigment paste, can be fed into a smaller cylinder. The movement of the cylinders is controlled by an accurate servo-hydraulic drive that produces a constant mixing ratio of components A and B at 1:1 throughout the entire mold filling process. The volume flow during mold filling can be adapted to a desired profile. The pigment paste is metered in the desired concentration.

The LSR components are then fed into a mixing block and static mixer and blended. The blended LSR passes into a hydraulic valve with a cold runner. This valve is integrated directly into the injection mold. The LSR can be injected into the mold at a high volume flow rate via the hydraulic cylinders. The mold temperature is between 90 °C and 150 °C. As a result of the LSR's low viscosity, long flow paths can be achieved with little pressure loss. The LSR is heated and completely vulcanized by means of the hot mold. Short cycle times of 6 minutes can be achieved. Channels are arranged at suitable points in the mold to minimize the pressure produced in the mold during vulcanization caused by thermal expansion of the LSR. This Channels allows small quantities of the expanded LSR to escape. As a result, clamping units with relatively low clamping forces can be used for large volume parts.

Fig. 1

Type	HCR	LSR	RT2
viscosity at 10 s ⁻¹ (Pas)	150000 - 200000	100 - 1000	10 - 100
polymer viscosity (Pas)	20000	30	1 - 10
type of cross linking	R , A , (S)	A	A , K
content of reinforcing filler *	30	20	10

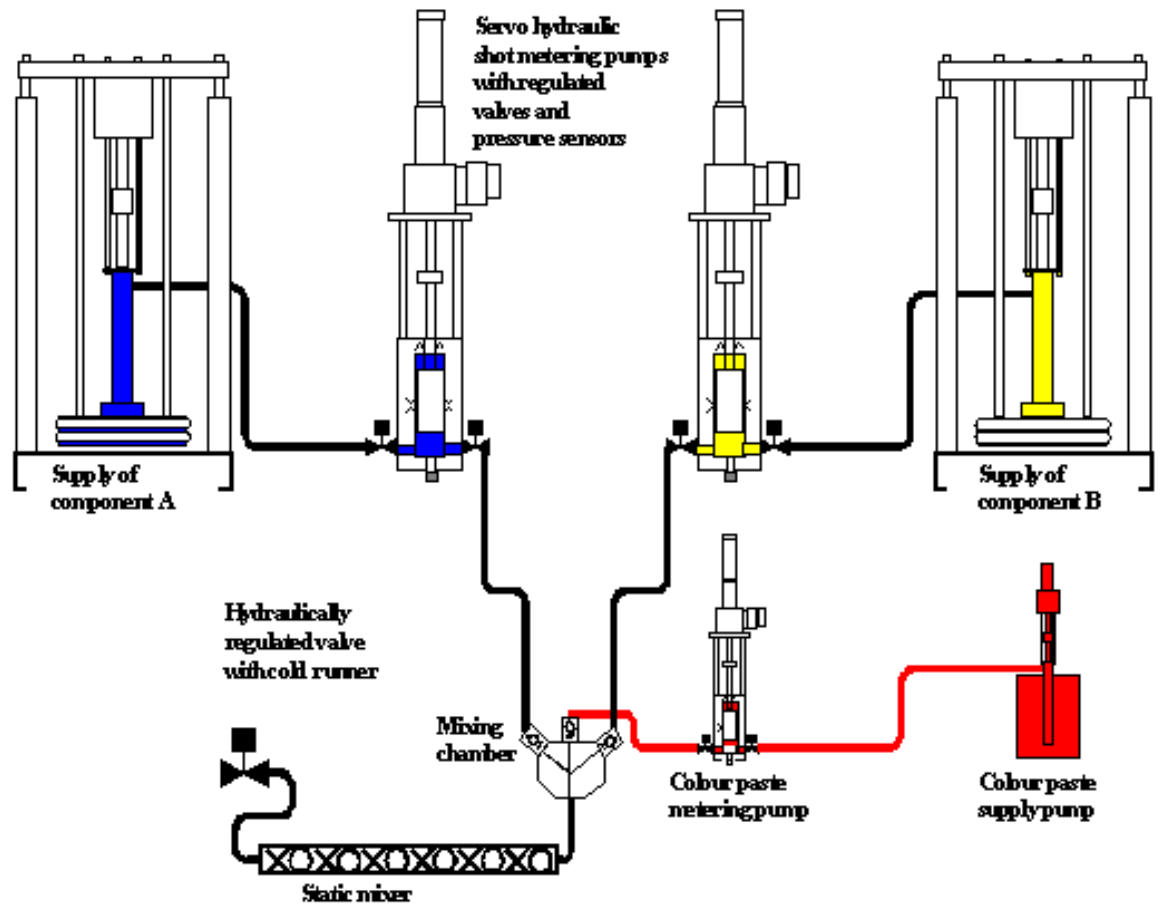
*(50 Shore A) , R = peroxid , A = addition cure , K = condensation , S = rays

Fig. 2

Advantages of LSR

- 2 part addition curing system ready to use
- Long pot life after mixing of components A and B
- Fast vulcanization above 90 °C
- High degree of process automation possible
- No need of heavy duty processing equipment
- Short cycle times
- Low viscosity and excellent flow properties
- Easy demolding at high temperatures
- No post cure necessary
- Low temperature and pressure stress for inserts

Fig. 3



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Test methods

a. Inclined plane test (IPT)

The inclined plane test (DIN IEC 587) is a recognized method for determining erosion and tracking resistance (E&TR) of insulation materials for outdoor high-voltage applications. A continuously flowing solution containing electrolytes and a wetting agent is applied to the surface of the 6 mm thick test plate. Flow effect and discharge caused by high voltage directly erode measurement of hydrophobicity of the silicone elastomers. This method detects a status that in field practice, does not occur in the use of silicone insulators. At 1000 hours in a salt fog test (IEC 61109) this state will be reached.

A number of silicone composite insulator manufacturers demand products which pass the inclined plane test (DIN IEC 587) at a test voltage of 4.5 kV. Apart from the selection criterion of a maximum current flow of $I = 60$ mA over a time period $t \geq 2$ sec., the average mass loss in % w/w and the appearance of holes (depth of erosion > 6 mm) are used as evaluation criteria for the IPT we carried out. As a rule, ten test plates per sample are measured.

In some cases, the arc resistance (AR) according to DIN VDE 0441 Part 1 (or DIN EN 61621) is evaluated as an addition to the IPT. This method permits the rapid measurement of a sufficiently large number of test pieces. The evaluation criterion is the time to disappearance of the arc in the material. At that time the materials start to be electrically conductive. The mean values from 20 tests are determined.

b. Hydrophobicity transfer (HT) to an artificial external layer (pollution simulation)

Initially, the external layer used in this test is a suspension consisting of water, fumed silica and a wetting agent, which is applied to the test plates. The coated test plates are dried for 60 minutes at a temperature of 50 °C. After drying, the external layer is completely hydrophilic. The dynamic receding angle (RA) is used for quantitative evaluation of the HT as a function of the transfer time. Full hydrophobicity, which is the initial state of the silicone rubber surface without an artificial layer, is again produced at an RA of 80 degrees

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Results and discussion

a. Properties of a typical LSR (LSR-A)

Modifications to the properties are illustrated by changing the recipe on the basis of an LSR formulation referred to in the following as LSR-A. LSR-A is a typical LSR formulation with good flow properties (viscosity 60 Pas). LSR-A was not developed for use in composite insulators. The components of the LSR are:

- Divinyl polydimethyl siloxane (polymer)
- Silica for mechanical reinforcement (reinforcing filler)
- Aid for modifying the silica
- Inhibitor
- Cross linking agent
- Platinum catalyst

The IPT results for LSR-A are shown in [Fig. 4](#). The IPT is formally passed at a test voltage of 3.5 kV (current criterion). At this voltage there are some cases of erosion depths of 6 mm (holes). The average mass loss is around 1.3 %. LSR-A fails when a test voltage of 4.5 kV is applied. The IPT did not pass according to the current criterion. The mass loss has rose sharply.

The HT of the synthetic external layer shows a sharp rise ([Fig. 5](#)) after an induction period. The RA reaches a value of 80 degrees after approximately 50 hours. Comparison with literature data (G) shows the HT of the LSR-A can be assessed as "good". As a classification for the starting product LSR-A it can be established that the T&ER is not adequate.

b. Effect of the filler aluminum hydroxide (ATH)

It is known that the E&TR of HCR can be improved by using ATH (E). Even with our LSR developments, the affects of ATH on E&TR and HT should be tested. A concentration range up to a maximum ATH concentration of 60 % w/w was set for this evaluation. A silanised, i.e. surface-modified, grade was used as ATH (ATH1).

The effect of ATH1 on the E&TR at a test voltage of 3.5 kV is shown in [Fig. 6](#). According to the current criterion, all samples in the concentration range formally pass the IPT. However, the mass loss increases as the concentration rises. It passes through a maximum and then drops again sharply for very high concentrations. The filler ATH1 shows a clear effectiveness from 50 % w/w. in regards to the E&TR. The 60 % formulation exhibits excellent low erosion. At lower concentrations, the ATH1 clearly leads to a deterioration of the E&TR compared to the unfilled reference sample.

These results are confirmed by the AR measurements ([Fig. 7](#)). As the ATH1 concentration rises, the average fail time drops, i.e. the AR, like the E&TR, is clearly worse with a low to average concentration. If the ATH1 concentration rises further, the average fail time rises.

For the HT, based on the results of the IPT and AR, only the formulation with a 60% ATH1 concentration is considered. The effect of a second, non-silanised ATH grade (ATH2), again at a concentration of 60 % w/w, is examined for comparison. The results are shown in [Fig. 8](#). The highly filled ATH samples show a rapid HT. Without a recognizable start-up phase, the RA rises very rapidly. No significant difference between the different ATH grades can be identified. In contrast, the unfilled LSR-A shows a clear start-up phase and the RA of 80 degrees is only achieved after clearly higher transfer times.

This result was not anticipated. The diffusion of low molecular weight siloxanes (LMW) from the elastomer to the external layer was discussed as a recognized mechanism (A, B) to the HT of silicone elastomers. The LMW concentration in the filled samples must be lower than the reference sample due to the high ATH content. Therefore, a deceleration of the HT of the filled samples is to be expected. In contrast to this assumption, the HT of the highly filled samples is clearly more rapid and the rate of diffusion of the LMW to the external layer must be increased. A possible cause of the elevated rate of diffusion of the LMWs could be the morphology of the ATH filled samples.

In summary, it can be said that only at high ATH contents of 50 % w/w is a visible improvement in the IPT to be observed. Surprisingly, the highly filled LSR-A samples show a very rapid HT. However, as a result of the sharp rise in viscosity and the sharp drop in mechanical properties, the advantages of the LSR described in Section 2 are largely lost. The addition of ATH thus proves not to be the suitable route to improve the properties of a typical LSR for use in composite insulators.

c. Effect of a chemical variation

On the basis of the results reported, it is obvious that a variation in the filler and in particular, the amount of filler does not produce the desired result for combined property improvement of the LSR. At GE Bayer Silicones, the influence of the network structure and binding of the silica to the polymer with regard to improving the E&TR is being actively investigated. A rapid HT is required with excellent flow and mechanical properties still possible. An LSR (E242) that exhibits the properties described below has been developed on the basis of these investigations.

[Fig. 9](#) shows the results of IPT from E242 at 4.5 kV compared to the samples filled with a high volume of ATH and reference material LSR-A. The chemical variation from LSR-A to E242 leads to an excellent improvement in the E&TR. The average mass loss is very low and the depth of erosion does not achieve the 6 mm limit for any of the test pieces measured. Only the sample filled with the silanised ATH shows any similar low mass loss. However, pitting occurred with several test pieces. E242 also exhibits the same low viscosity as the reference LSR (see [Fig. 9](#)). Again the good mechanical properties are achieved in full.

The HT could likewise be improved by the chemical variation compared with the LSR-A ([Fig. 10](#)). The RA rises without an induction period and is achieved after a transfer time of only 25 hours at 80 degrees.

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Summary

- Research work was carried out with the aim of developing a two-component liquid silicone rubber (LSR) which is especially suitable for use as an insulation material in composite insulators. The good flow and mechanical properties of LSR should be retained. The effect of a change in recipe on electrical properties (erosion and tracking resistance) and on hydrophobicity (hydrophobicity transfer to an external layer) was investigated.
- The basis of the observation is a typical LSR formulation (LSR-A) with good flow properties (viscosity at room temperature of 60 Pas). Comparison with literature data (G) shows that the hydrophobicity transfer to a synthetic external layer can be assessed as "good". However, the erosion and tracking resistance of the required 4.5 kV is not achieved.
- By adding aluminum hydroxide, the erosion and tracking resistance can be significantly improved even at very high concentration (> 50 %w/w). However, the sharp rise in viscosity caused by the high degree of fill and the drop in the mechanical properties means the processing advantages of the LSR are lost.
- Excellent erosion and tracking resistance (4.5 kV) is achieved by suitable chemical modification of the LSR-A to LSR E242 while retaining good flow and mechanical properties. While already good, hydrophobicity transfer to an external layer is improved.

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