The Effect of Absorbed Moisture on Dielectric Behavior of Silica (Micro)-Unsaturated Polyester Composites

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ABSTRACT: It is well known that most of the dielectric failures occur in high voltage equipment due to absorption of moisture by the insulating material from the environment. Hence, the effect of absorbed moisture on electrical and mechanical properties of silica-unsaturated polyester resin (UPR) composites has been evaluated. The absorption of moisture in silica-UPR composites does not show any significant change in electrical and mechanical properties. The effect of acetone and water absorption on silica-UPR composites was determined and it was that found silica-UPR composite shows higher acetone absorption when compared with water. The differential scanning calorimetry and thermal gravimetric analysis studies of silica-UPR composites show no significant change in glass transition temperature using prehumidified (0–95% RH) silica filler. As there is no significant change in thermal and mechanical properties after exposure to humid conditions, it can be concluded that water does not penetrate inside the polymer matrix. Hence, the silica particles are the best choice to use as filler in UPR matrix for UPR composite used in electrical equipment. The developed silica-UPR composite was successfully used in the preparation of medium voltage inductive transformers.
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Key words: unsaturated polyester; polymer composites; dielectric material

INTRODUCTION

Almost every polymeric system absorbs some moisture under normal atmospheric conditions from air. The sensitivity to moisture increases if a filler or fiber is dispersed in the polymer matrix. After the absorption of water the electrical properties of polymer composites, such as volume and surface resistivity, dielectric strength, comparative tracking index and arc resistance decrease while there is a corresponding observed increase in the dissipation factor and dielectric constant. The absorbed moisture also causes the initiation and development of water tree, which finally leads to catastrophic dielectric failure of polymer composites.[1] Thus, the water sensitivity of a polymer composite is an important criterion with respect to electrical properties of polymer. The mechanical properties of polymer composites are also adversely affected. The effects are very similar to the changes observed in electrical properties.[2,3] The effect of moisture on various types of polymer composites[4–9] has been studied by researchers with fillers such as cellular fiber, glass fibers, talc, MMT, and so on. However, there is still a need to determine good filler for a polymeric matrix in humid environmental conditions. Silica- and polymeric materials-[such as epoxy, unsaturated polyester resin (UPR), etc.] based composites are used in several electrical appliances such as cables, insulators, circuit boards, bushings, motors, and generators in there ground wall insulations and cast resin transformers. Although there have been different types of fillers studied in a UPR system, silica has been selected for this study because it has extensive industrial application, good thermal conductivity, excellent thermal shock resistance, and a low specific electrical conductivity. It has extremely good dielectric and insulating properties. It is abundantly available and is a cost effective filler. For these reasons, silica was used as an inert low expansion filler material in the present study.

UPR is used in electrical field for the formation of motor and cable wire enamels, transformer insulation films, capacitors, thermal printing tapes, membrane touch switches (computer and calculator keyboards), and flexible printed circuit films. UPR has advantage over epoxies due to its lower cost, and therefore, UPR widely used as a cost effective alternative in electrical apparatus. As discussed above, absorbed moisture is the main cause for dielectric failure of polymer composite used as electric insulations. Therefore, the effect of absorbed moisture on dielectric and mechanical properties of silica-UPR composites has been evaluated and presented in this article. The water and solvent absorption studies of

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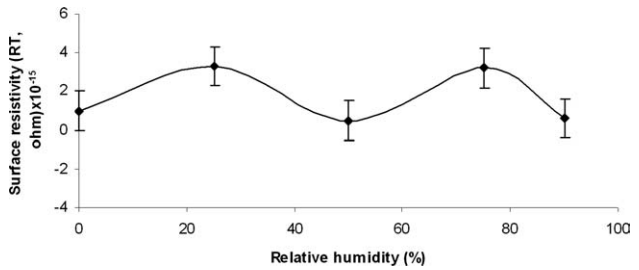

Figure 1 Surface resistivity of silica-micro-UPR composites.

silica-UPR composites were also determined and reported in this article.

EXPERIMENTAL PROCEDURE

Materials

The silica is used as obtained from the Evonik Industries, India. The silica used as filler is micro-sized and has a surface area = 0.05–0.250 m^2/g. The unsaturated polyester used in the study was obtained from M/s Naphtha Resins, Bangalore, India, and was used as such. The resin was preaccelerated with 0.2% cobalt naphthanate (6% Co content) and had a solid content of 55% and used styrene as the reactive diluent. The acid value of the composition was 12 mg KOH/g resin and had a viscosity of 330 mPa s (@ 25°C and 50 rpm). This is an oligomer with unsaturations, which are cured on crosslinking via a free radical initiated reaction. The initiator used for the cure of the UPR was methyl ethyl ketone peroxide (MEKP) obtained from M/s Naptha Resins and used as such at a 1.2% concentration.

Drying and humidification of silica particles

The silica particles were dried at 140°C for 4 h in an air circulating oven. These particles were kept at 25, 50, 75, 90% relative humidity at 37°C for 100 h in a Climatic testing chamber C-1000 made by Weiss-Voetsch Environmental testing Instrument. The moisture equilibrated silica was then used in preparation of silica-UPR composites.

Preparation of samples

Silica-UPR composites were prepared by adding 80 g of appropriately humidified silica in 20 g of UPR resin. The slurry was mechanically agitated to ensure proper dispersion of silica particles in the resin matrix.

The mixing of silica and UPR resin was done using a baffle anchor type agitator, at an rpm of 500 ± 50 at room temperature for a period of 2 h. The composition was then poured into Teflon and metal molds after addition of the initiator stirred for 10 min and allowed to cure at room temperature, that is, 25 ± 1°C for 12 h followed by postcuring at 80 ± 1°C for 4 h. The composites were then allowed to stabilize for 7 days at 25 ± 1°C and 50% relative humidity before any testing was carried out.

Testing

Electrical properties

The electrical testing (surface and volume resistivity, arc resistance, dissipation factor, dielectric constant, and dielectric strength) of samples was performed at Electrical Research and Development Centre, Vadodara, India. The samples used for the electrical study were cast on hard chromed metal molds and the smooth surface used for the test and had dimensions of 100 × 100 × 3 mm^3.

Mechanical properties

The mechanical properties of the silica-UPR composites were evaluated using a Lloyd 50 Universal Testing Machine with a 50 kN load cell in the case of tensile properties and 500 N for flexural properties with a jaw speed of 2 mm/min and a gauge length of 5 cm. The samples used for the evaluation of mechanical properties had dimensions of 100 × 10 × 5 mm^3.

The impact strength of the samples was evaluated on a Denson Avery Impact tester with a striker of 2.7 J with a striking velocity of 3.46 m/s in accordance with ASTM D256.

Thermal properties

The glass transition temperature of the Silica-UPR composites was evaluated on a Mettler Toledo DSC822e machine with a sample weight of 10–20 mg and a heating rate of 10°C/min in a nitrogen atmosphere. The sample was cycled from 25–250°C and 250–25°C and the same was repeated. The glass

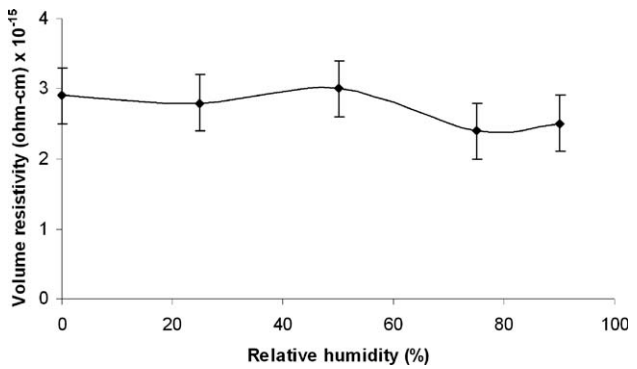

Figure 2 Volume resistivity of silica-micro-UPR composites.

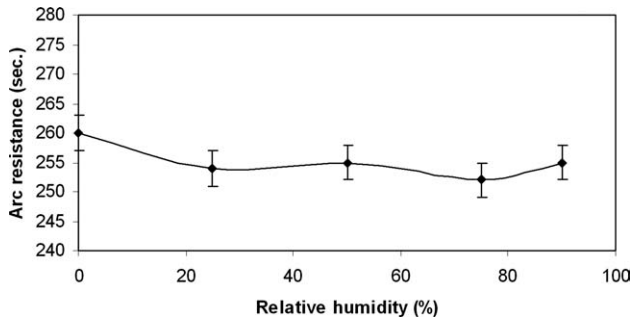

Figure 3 Arc resistance of silica-micro-UPR composites.

transition was evaluated from the second run to eliminate thermal history from the sample.

The thermal degradation study [thermal gravimetric analysis (TGA)] of silica-UPR composites was done using Mettler Toledo TGA/DSC-1 machine in a nitrogen atmosphere with a flow rate 30 mL/min with a sample weight of 15–20 mg and heating rate 10°C/min. The sample was heated from 25–500°C.

Chemical properties

The prepared silica-UPR composites were immersed in acetone and water. Three samples with dimensions of $3 \times 3 \times 0.3$ cm^3 were immersed separately in 100 mL of acetone and water, maintained at 30°C. After the required amount of time, the samples were removed and gently dried using a filter paper to remove acetone and water adhering to its surface and weighed. The dried composite samples were further placed in an air circulating oven, maintained at 100°C, for 2 h and removed, cooled to room temperature in desiccators, and then weighed.

RESULTS AND DISCUSSION

Electrical properties

The electrical properties of dielectric polymeric material have been seen to decrease[10,11] with increasing moisture content in polymeric matrix. The surface and volume resistivity indirectly depends on the chemical composition of the dielectric material. A material with high moisture absorption tendency shows reduction in surface and volume resistivity. The surface and volume resistivity of silica-UPR composite has been determined and presented in Figures 1 and 2. It is clear from Figures 1 and 2 that the surface and volume resistivity of silica-UPR composites were found approximately constant after subjecting the filler to different humidity conditions. The arc resistance of dielectric material measures the localized electrical-breakdown voltage in insulation system. The higher value of arc resistivity for material shows a higher resistance to electrical breakdown. The arc resistance of silica-UPR composites was evaluated, and the results were presented in Figure 3. It is clear from Figure 3 that the arc resistance of the composites was made using dried silica filler is higher than composite made by using humidified silica filler, which then became constant with filler subjected to 25–95% relative humidity conditions. This indicated that the silica initially absorbed some moisture at room temperature and gets saturated after which there is no absorption of further moisture. Hence, the values of arc resistivity were initially higher at 0 phr humidity and then constant with increasing relative humidity. The dielectric constant of silica-UPR composites were measured and shown in Figure 4. It is clear from data shown in Figure 4 that the value of dielectric strength was constant for all relative humidity

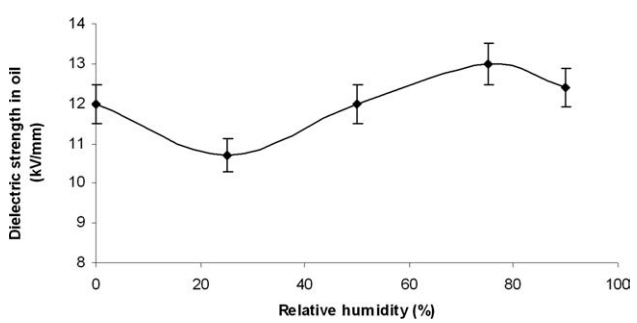

Figure 4 Dielectric constant of silica-micro-UPR composites.

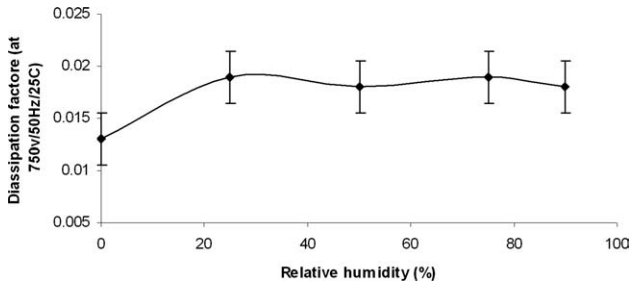

Figure 5 Dissipation factor of silica-micro-UPR composites.

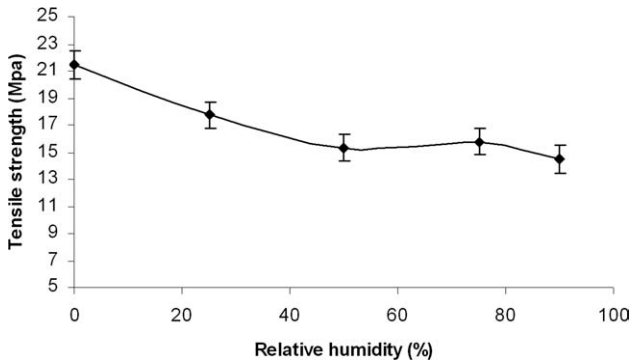

Figure 6 Tensile strength of silica-micro-UPR composites.

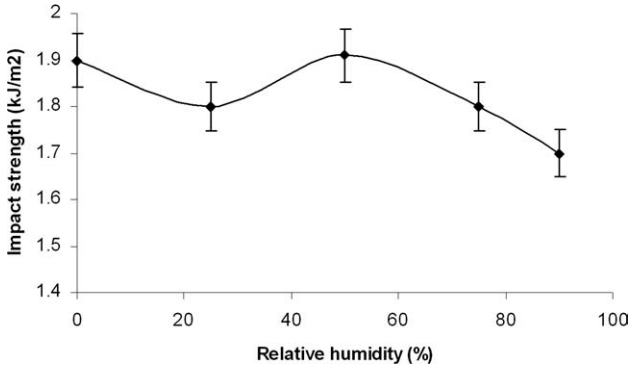

Figure 7 Impact strength of silica-micro-UPR composites.

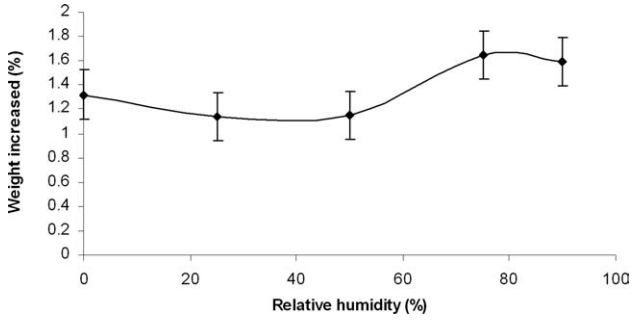

Figure 9 Acetone absorption of silica-micro-UPR composites.

conditions. Dissipation factor (tan δ) is ratio of the power loss in a dielectric material to the total power transmitted through the dielectric system. The values of dissipation factor of Silica-UPR composites were evaluated for fillers subjected to different humidity conditions and presented in Figure 5. Figure 5 indicates that the value of dissipation factor does not change at different relative humidity condition. Hence, it is clear from above studies that the electrical properties of silica-UPR composites were not adversely affected when the silica was subjected to different relative humidity condition. It may be due to low water absorbance tendency and low surface area of silica. Therefore, the silica is the best filler for preparation of polymer insulating material.

Mechanical properties

Earlier it was observed by other researchers that the mechanical properties of various polymeric materials decrease with an increase in the moisture content in polymer matrix.[12–14] Hence, mechanical properties of silica-UPR composites were determined to evaluate the effect of moisture. The tensile strength of silica-UPR composites was determined and depicted in Figure 6. It is clear from Figure 6 that the tensile strength decreased in composites with silica subjected to 25% RH. This may be due low interaction of silica microparticle and UPR in presence of water. The impact strength (Fig. 7) and flexural strength (Fig. 8) were constant at different relative humilities.

Chemical properties

The solvent absorption studies of silica-UPR composites were done with acetone and water, and the obtained results are presented in Figures 9 and 10. The acetone absorption study was done according to the procedure given in "Chemical properties" subsection of section "Testing." It is clear from Figure 9 that silica-UPR composite had acetone absorption within the range of 1.35–1.59%. The water absorption study of silica-UPR composite was done, and the results obtained are presented in Figure 10. It is clear from Figure 10 that the water absorption was within the range of 0.33–0.54%. Hence, it is clear from the above study that the silica-UPR composite shows higher organic solvent (acetone) absorption when compared with water absorption. Therefore, the water penetrates to a lower extent inside the polymer composite matrix, when the products made by silica-UPR composites were placed in open environmental condition. Hence, the silica particles are the best choice to use as filler in UPR matrix for application in electrical equipment.

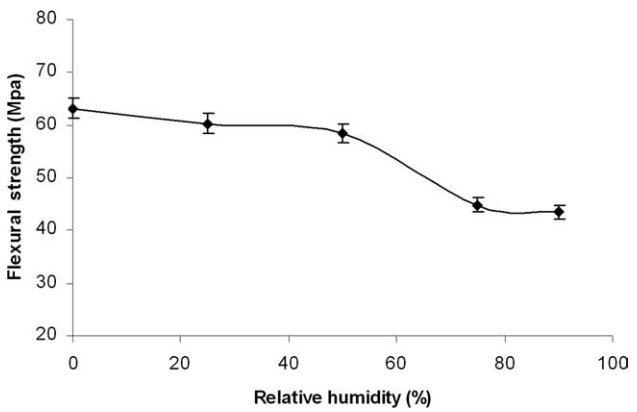

Figure 8 Flexural strength of silica-micro-UPR composites.

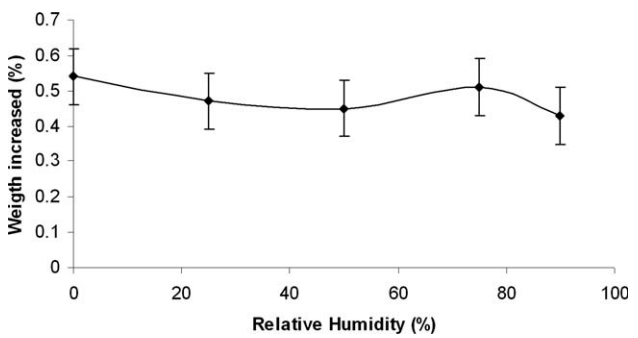

Figure 10 Water absorption of silica-micro-UPR composites.

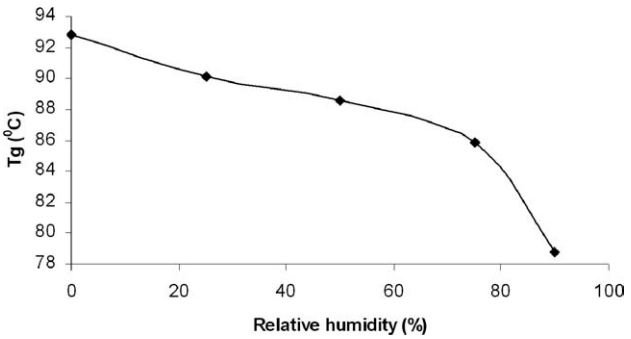

Figure 11 Glass transition temperature

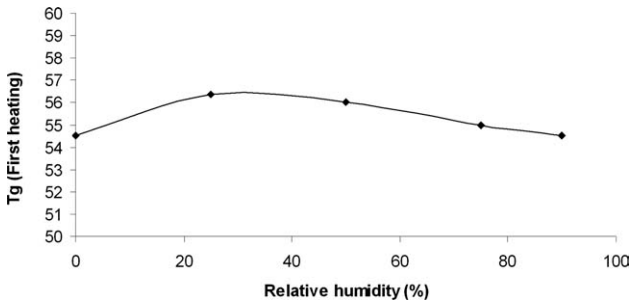

Figure 12 First heating values of silica-micro-UPR composites.

Thermal studies

Differential scanning calorimetry (DSC) measurements were used to measure the glass transition temperature (T_g) of the silica-UPR composites made using humidified silica at different relative humility conditions and are shown in Figure 11. The DSC results show that the glass-transition temperature of silica-UPR composites decreased slightly with increasing relative humidity. As the environmental relative humidity increased from 0 to 75%, the T_g of the silica-UPR composites decreased by $\sim$ 5°C, but a slightly higher variation at 90% RH was observed. This was due to plasticization of the polymer matrix induced by inclusion of water molecules in the polymer matrix. It was seen that this inclusion of water in the matrix did not adversely affect the electrical properties of the composites to a large extent, with the exception of the tan δ, which increased with the RH.

The values of first heating run (Fig. 12) also indicate that no significant effect of moisture was observed.

The TGA of silica-UPR composites was done to evaluate the phase transition study with variation of temperature and results are shown in Figure 13. This is clear from Figure 12 that the TGA patterns of all silica-UPR composites are similar, which indicate that there is no change in material property after using silica as a filler, after being humidified at different relative humidity condition. The degradation of material takes place in temperature range at 330–440°C. This is due to degradation of UPR resin. Hence, humidification did not adversely affect the thermal stability of the composite.

Industrial application

Polymer concrete-based composites are extensively used in electrical industry as a casting composition, and they are also replacing porcelain and epoxy insulation in high voltage electrical application.

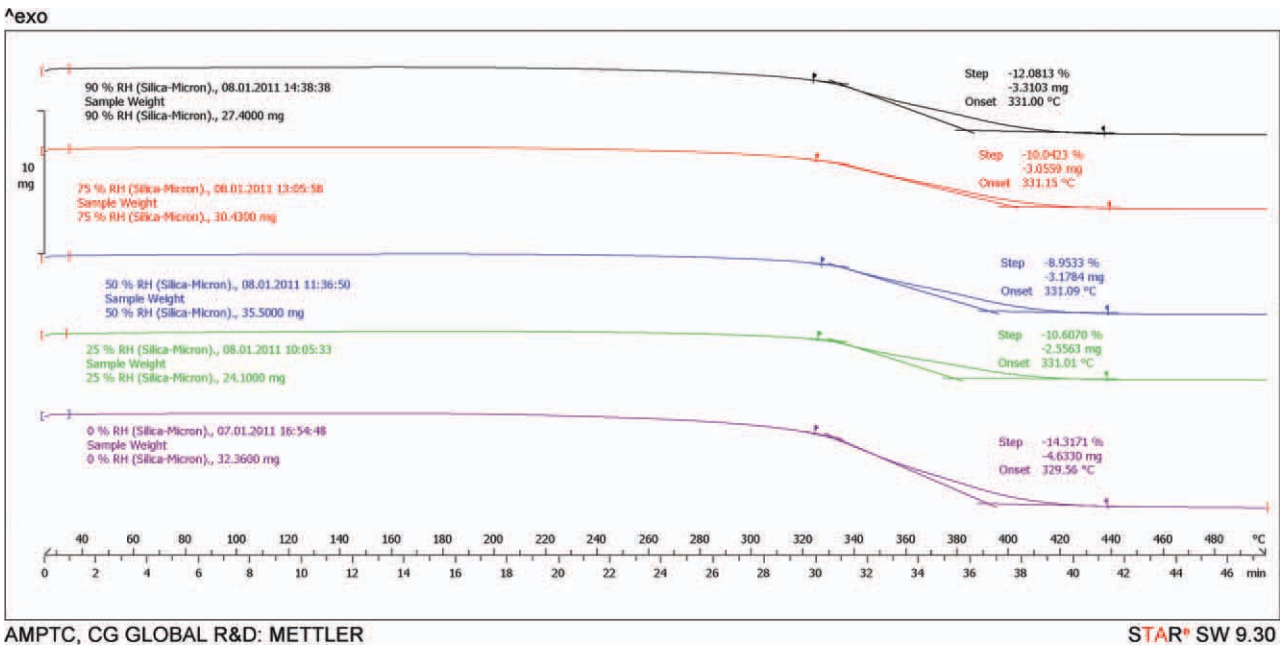

Figure 13 TGA graphs of silica-micro-UPR composites. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

CONCLUSIONS

The effect of humidity on dielectric and mechanical properties of silica-UPR composites was studied. It was observed that the electrical and mechanical properties of silica (micro)-UPR composites were not changed significantly at different humidity condition. This may be due to the fact that silica has a very low surface area (BET surface area, for 80/100 retension on 100 mesh sieve = 0.05–0.250 m^2/g). Therefore, the mixture absorbs very low quantities of water from atmosphere. The organic solvent and water absorption of silica-UPR composites was evaluated, and it was that found the silica-UPR composites showed higher acetone absorption (1.59%) when compared with water (0.55%). The DSC and TGA studies of silica-UPR composites showed no significant change in glass transition temperature and phase change of material. Therefore, water will not permeate inside the polymer composite matrix of products made using silica-UPR when placed in an open environment Hence, silica particles are the best choice as filler in UPR matrix for applications in electrical equipment.

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