High-voltage insulation and dielectric properties of ceramic-glass composites

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\section*{ABSTRACT}
This paper reveals the high-voltage insulation properties – namely the dielectric constant, dissipation factor, breakdown strength, and voltage – of epoxy resin samples of various thicknesses containing the microadditives albite, potassium feldspar, lithium glass, and zinc glass in different concentrations. 0.1 wt\% KAlSi\textsubscript{3}O\textsubscript{8} ceramic powders as fillers in 1 mm epoxy composites designed for insulation is taken as the choice for highest breakdown strength 36.6491 kV/mm. After that, 5 wt\% Li-Bi-B-O glass at 35.4687 kV/mm, 10 wt\% Zn-Bi-B-O glass at 35.0432 kV/mm, and 10 wt\% NaAlSi\textsubscript{3}O\textsubscript{8} at 33.6504 kV/mm. The increase of thickness more than 1 mm is not recommended in the practical application due to the decrease of breakdown strength. Multi-layer thin structure is recommended for insulation purpose.

\section*{1. Introduction}
Additives enhance the applicability of insulation materials by improving their structure or properties. The most important changes in structure are associated with the relationship between the filler and the porosity because this relationship directly affects the electrical properties, especially dissipation factors, breakdown strength, and voltage. Compared to the filler, when high voltage is applied, high porosity means the volume of air in the structure will increase due to the high temperature, which might crack the structure. The filler can be treated as the trimming factor for base insulation materials.

In ceramic–polymer composites, dielectric loss is dominated by the polymer phase, whereas steady-state electrical conduction and low-frequency dielectric loss at high temperatures are mainly determined by the ceramic phase [1]. A multicore model for nano-composites has been developed, and differences in the properties of nano-composites and microcomposites have been reviewed [2]. Metal oxide nanoparticles can mitigate interfacial space charge and interfacial polarization of trapped entanglement through the Maxwell–Wagner effect [3]. The energy of covalent bonding between the matrix and filler decides the high-voltage endurance and the temperature at which the breakdown strength decreases [4].

Glass is the most well-known amorphous solid; its conductivity varies in accordance with the charge carrier mobility and concentration. For example, phosphate glasses are considered strong electrolytes [5], whereas alkali oxide glasses are considered weak electrolytes [6]. Experiments into the carrier concentration and mobility in glasses have shown that the conductivity and dielectric properties vary in different types and strengths of electric fields [7–11].

Bismuth borate glasses are widely used because their glass transition temperature is low (600–800°C); their structure and physical properties have been analyzed and surveyed [12–14]. Because of the advantages of bismuth borate glasses, studies on ZnO-Bi\textsubscript{2}O\textsubscript{3}-B\textsubscript{2}O\textsubscript{3} glass systems have explored their potential in optical, thermal, and electrical applications [15–17]; in addition, their properties under the addition of certain chemicals have been investigated [18–20]. Moreover, alkali metals have been added to lithium bismuth borate glasses so they can be employed as conductors; the addition creates nonbridging oxygen ions that alter the density and electrical conductivity of the glass [21,22].

Studies have focused on the addition of fillers to epoxy to improve its electrical insulation properties under high-voltage environments [23,24]; as such, research on the properties and insulation improvement with the addition of glass as filler in epoxy is lacking. In this paper, we investigate the dielectric and high-voltage insulation properties of glass–ceramic composites containing fillers; these materials have potential applications, especially in cases where various recycled materials are used to manufacture several types of glasses.

The powder of albite and potassium feldspar are the common raw materials in the industry of glass and ceramic manufacturing, and the mineral resource is rich in felsic rocks. The properties and the manufacturing process of ZnO-Bi\textsubscript{2}O\textsubscript{3}-B\textsubscript{2}O\textsubscript{3} and Li\textsubscript{2}CO\textsubscript{3}-Bi\textsubscript{2}O\textsubscript{3}-B\textsubscript{2}O\textsubscript{3} glasses are already developed and certified as stable,
which can reduce the unpredictable and potentially dangerous situation, and the result of measurements could be analyzed more easily.

2. Experimental procedures

The epoxy base should have properties that help reduce the difficulty in sample manufacturing. EpoFix resin and EpoFix hardener from Struers were used in this study. EpoFix samples have excellent adhesion, low viscosity, superior penetration into cracks and pores, low curing temperature, and a long curing time of approximately 12 h. To harden the ceramic and glass EpoFix composite samples, a resin/hardener mixing ratio of 25:3 by weight was employed.

The original materials in the powder are Al₂O₃, SiO₂, Na₂CO₃, K₂CO₃, B₂O₃, Bi₂O₃, ZnO, and Li₂CO₃. Al₂O₃ powder with an average particle size of 40–50 nm (CAS:1344–28-1) from Alfa Aesar and SiO₂ powder with an average particle size of 40 nm (CAS:60676–86-0) from UniRegion Bio-Tech were used for preparing albite and potassium feldspar. Na₂CO₃ powder and K₂CO₃ powder from SHOWA were particular used for preparing albite or potassium feldspar. B₂O₃ powder from NOAH technologies and Bi₂O₃ powder from SHOWA were used to produce the Bi₂O₃–B₂O₃ glass base. ZnO powder from SHOWA and Li₂CO₃ powder from Alfa Aesar were used as modifiers in Bi₂O₃–B₂O₃ glasses.

Na₂CO₃, Al₂O₃, and SiO₂ powders were mixed in proportion and calcined at 650°C to produce albite powder. K₂CO₃, Al₂O₃, and SiO₂ powders were mixed in proportion and calcined at 850°C to produce potassium feldspar powder. ZnO–Bi₂O₃–B₂O₃ and Li₂CO₃–Bi₂O₃–B₂O₃ glasses were melted at 900°C–1000°C and annealed at 350°C–450°C to produce.

Desiccation of the filler powders was performed at 120°C for 1 h. The filler powders were stamp-milled and passed through a 100-mesh screen. Achieving a uniform distribution of glass powder particles is difficult when using an ultrasonic oscillator for direct dispersion; thus, dispersion takes considerable time. Samples of different thickness (1, 2, and 3 mm) were manufactured.

The dissipation factor and capacitance were measured using an LCR meter (GWInstek LCR-821) with an Agilent 14651B dielectric test fixture. Connection setup is shown in Figure 1(a). The relative dielectric constant was determined using the measured capacitance value.

The breakdown voltage of the samples was measured using the Baur DTA-100 with disk electrodes. Schematic diagram of practical environment is shown in Figure 1(b). Furthermore, the breakdown strength and voltage of the samples were determined using the Weibull distribution.

3. Results and discussion

A surface scanning electron microscopy (SEM) image of the composite sample with 5-wt% filler concentration is shown in Figure 2. Because of the small particle size, the NaAlSi₃O₈ and KAlSi₃O₈ powders can hardly be observed in the composite samples. Air bubbles and air gaps were easily detected around the glass powder filler.

The dissipation factors of the samples were measured and are listed in Table 1. The applied frequencies can be considered as 1st, 10th, 100th, and the 1000th harmonic and the corresponding impedance is surveyed. When a voltage field is applied, harmonics can damage the power equipment or cause a fault in the insulation protection. The dissipation factors of the composite samples were less than 1 kHz under most conditions, indicating that the rate of energy loss, taking the form of electrical oscillation, was low. When the high-frequency (e.g. 100-kHz) electrical field was applied, the rate of energy loss was higher and greater electrical and mechanical oscillations were produced. Such electrical and mechanical oscillations should be treated as internal sources of damage, which can affect the heat source and molecular vibrations, subsequently weakening the structure of a solid sample. Furthermore, the rate of energy loss was higher under some manufacturing parameters when a 100-Hz electrical field was applied, implying aggregation of the additives during the molding process proceeding from gel to solid.

![Figure 1](image-url)  
Figure 1. Schematic diagram of setup of test cells (a) LCR meter with dielectric test fixture (b) Setup of high voltage breakdown test.
which have maximum the tron electrons, and the tunnel transporta tion of the material's ability to trap electrons, but too many electron traps may result in small distances between electron traps, increasing the effective length of the electron transportation tunnel due to an increase in the likelihood of trap-to-trap electron transfer. Increasing the thickness of the sample can raise the maximum limit of the effective length of the electron transportation tunnel. Ceramic fillers in epoxy resins have greater ability to trap electrons than glass fillers, which could be verified for fillers with low concentration in epoxy composite samples. Electron transportation tunnels can be easily stuck by applying a high-frequency electrical field; according to the measurement results, tunnel stacking begins to appear between 10 KHz and 100 KHz.

A low dissipation factor is mostly found for composites with the additives 5-wt% NaAlSi3O8, 10-wt% KAlSi3O8, 0.1-wt% Li–Bi–B–O glass, and 5-wt% Zn–Bi–B–O glass; this suggests that the average activity of the metal elements in the filler powder strongly affects the dissipation factor. Glass fillers with a low Li–Bi–B ratio have lower dissipation factors than those with other fillers.

A breakdown voltage test was performed under a voltage increase rate of 500 V/s. The Weibull parameters of the composite samples are presented in Table 2. The alpha parameter indicates the characteristic breakdown strength (kV/mm), which is the value at 63.2% probability of the cumulative distribution.
increasing the condition.

The beta value of the Weibull distribution shows that the failure rate is highly related to the increase of time, which is related to the voltage increasing rate applied. The breakdown strength decreased with increasing thickness, indicating that the electrons accumulated at a fixed increasing rate; however, the electron transportation tunnels stocked easily when the thickness was small. The filler concentration affected the breakdown strength slightly; under a direct electric field, the filler acted more like a trap than a tunnel.

The beta parameter could be treated as the failure mode. All composite samples had a beta value of more than 10, which means that the failure rate increased with time. Glass filler composites with high concentration and thickness aged more easily under the test condition. Increasing the sample thickness changed the failure mode of the composite samples, especially the Zn–Bi–B–O glass composite samples.

The dielectric constant of 1-mm-thick samples was measured under 1 kHz and 25°C, and the values are listed in Table 3. The dielectric constant of the NaAlSi3O8 and KAlSi2O8 composite samples decreased with increasing filler concentration because of the electron traps. The dielectric constant of the Li–Bi–B–O glass composite samples varied over a small range and increased with increasing filler concentration because of the electric transportation tunnel.

Dielectric constant of NaAlSi3O8 and KAlSi2O8 is higher than pure Epoxy Resin, and the dielectric constant of lithium glass and zinc glass is also higher than pure Epoxy Resin. The composites in this work show that the permittivity of non-pure materials is dominated by the surface interaction between different materials, not the inner effect of each material.

4. Conclusions

This study investigated ceramic powders (NaAlSi3O8 and KAlSi2O8) and glass powders (Li–Bi–B–O glass and Zn–Bi–B–O glass), and the results can serve as a reference for tuning the parameters of the insulating parts of electrical equipment. Insulation with thickness greater than 2 mm is not recommended for the failure mode because of its strong time dependence. Insulating composites should be multilayered to prevent the formation of electron transportation tunnels and increase the number of surface electron traps. To conclude, ceramic powders work better than glass powders as fillers in epoxy composites designed for insulation.

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Disclosure statement

No potential conflict of interest was reported by the author(s).

Data availability statement

All data generated or analyzed during this study are included in this published article [and its supplementary information files].

Table 2. Weibull Parameters of the samples.

| Thickness (mm) | Weibull Parameter α | Weibull Parameter β | NaAlSi3O8 | KAlSi2O8 | Li-Bi-B-O glass | Zn-Bi-B-O glass |
|---------------|---------------------|---------------------|-----------|-----------|-----------------|-----------------|
| 1             | 31.8052             | 20.4149             | 35.9412   | 35.9412   | 19.3239         | 19.3174         |
| 2             | 20.1376             | 44.7241             | 44.7925   | 44.7925   | 39.2996         | 39.2996         |
| 3             | 10.1512             | 14.2672             | 14.3426   | 14.3426   | 14.0828         | 14.0828         |

Table 3. Dielectric constant of 1-mm-thick samples.

| Filler                  | Concentration (wt%) | Dielectric Constant (at 1 kHz) |
|-------------------------|---------------------|---------------------------------|
| NaAlSi3O8               | 0.1                 | 4.9624                          |
|                         | 5                   | 4.8237                          |
|                         | 10                  | 4.8096                          |
| KAlSi2O8                | 0.1                 | 5.0438                          |
|                         | 5                   | 5.0030                          |
|                         | 10                  | 4.6276                          |
| Li-Bi-B-O Glass        | 0.1                 | 5.7167                          |
|                         | 5                   | 5.8458                          |
|                         | 10                  | 5.6658                          |
| Zn-Bi-B-O Glass        | 0.1                 | 6.0924                          |
|                         | 5                   | 6.1188                          |
|                         | 10                  | 6.1568                          |
Compliance with Ethical Standards
1. No disclosure of potential conflicts of interest; 2. No research involving Human Participants and/or Animals; 3. All authors informed consent.

Author contributions
All authors contributed to the study conception and design. Material preparation, data collection and analysis were performed by Ming-Yueh Hsieh, Wen-Shiush Chen, Cheng-Hsing Hsu, and Cheng-Hsuan Wu. The first draft of the manuscript was written by Ming-Yueh Hsieh, Cheng-Hsing Hsu and all authors commented on previous versions of the manuscript. All authors read and approved the final manuscript.

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