Rheological, Electrical and Thermal Properties of Enhanced Epoxy/silica Composites

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Abstract — The subject of nanocomposite materials with reinforced filler has been studied extensively in the recent years. Fillers such as silica is commonly incorporated in epoxy formulations used for transformers bushings, to lend specific characteristics including mechanical, electrical, and thermal properties. But, the polar interactions among very small size particles in the range of micro and/or nano scale through hydrogen bonding and Van der Waals forces cause fillers to aggregate into a flocculated network, which is holding back mass production of optimal epoxy/silica composite formulations and products. In the present study, the problems of unwanted agglomeration and filler structure rebuilding have been achieved using a cycloaliphatic epoxy resin and specific formulations of micro and nano sized silica fillers. Experimental results revealed that new epoxy formulations in combination with electrospinning based mixing technology can enhance the heat distortion temperature (HDT) and electrical breakdown strength of the epoxy composite up to 40% and 20% respectively compared to the existing products. In addition, the viscosity or flowability of epoxy composites was improved five times or greater compared to the conventional formulations enabling new processing capabilities. The results of this work has significant impact on industry of epoxy composites.

I. Introduction

Over the last decade, rapid advances in the area of composite materials have made composites produced from epoxy resins as the matrix material to become one of the most common composites. Epoxy resins exhibit good electrical insulating properties, resistance to moisture, high mechanical strength, good adhesion to metals, and the ability to be cast in complicated shapes around conductors in a simple operation [1]. Epoxy resins are traditionally used either as castings or laminates reinforced with glass-fiber in the manufacture of outdoor insulators and bushings. Unlike porcelain, epoxy insulators do not shatter when damaged by impact and exhibit minimal change in leakage current due to improper assembly [2]. Epoxy insulators are therefore significantly less susceptible to destructive discharges than their porcelain counterparts and can be manufactured using a continuous rather than batch process [2].

Epoxy resins, similar to most of other resins, increase in density or shrink during polymerization or curing. This is due to replacement of many intermolecular spacings with closer inter-atomic bonds or intramolecular spacings, which contracts the structure [3]. This shrinkage initiates internal cracks in the epoxy insulator, which ultimately reduces the life time of epoxy insulators. Internal cracking of epoxy insulators can be avoided by adding a high-volume percentage of non-shrinking filler particles in order to reduce the coefficient of thermal expansion and enhance the thermal conductivity in comparison to the unfilled epoxy resin. The addition of fillers also enhances other mechanical and electrical properties by increasing the mechanical strength and electrical insulation properties of the epoxy composites. However, the epoxy composite becomes increasingly more difficult to process with increasing filler loading level limiting the advancement of material properties.

Currently, industry prepares epoxy composites with loading level approaching 70% filler by weight to achieve desired properties for outdoor insulator applications. However, the viscosity of epoxy composites increases with filler loading level, making composites increasingly difficult to mix and cast. Moreover, as it is difficult to degas highly viscous epoxy composites, internal voids remain within the composite which degrade its electrical insulation properties. It is generally understood that improving filler dispersion in a composite will reduce the increase in viscosity, and thus, it is deemed to be a potential solution to the above stated problem.

II. MATERIALS AND METHODS

In the present study, a unique combination of different size micro and nano particles along with electrospinning based mixing technology [4] are used to achieve the superior performance target while maintaining the epoxy formulation viscosity at a significantly low value compared to those used in commercial insulators. The experiments were conducted using a cycloaliphatic epoxy resin and specific formulations of micro and nano sized silica fillers. Five different silica-epoxy composite formulations were made by incorporating three specific different particle sizes of silica including 1) large micro particles (average particle size ~ 45 μm), 2) small micro particles (average particle size ~ 1.5 μm), and 3) nano particles - fumed silica particles (average primary particle size ~ 7-12 nm) in the epoxy resin. In addition to particle sizes, the effect of surface treatment of nano sized silica was investigated using hexamethyldisilazane aftertreated fumed silica (hydrophobic silica grade - aerosil® R8200) compared to the untreated silica particles (hydrophilic silica grade - aerosil® 300VS).

III. RESULTS AND DISCUSSION

Table 1 shows various formulations that were made during the investigation. The formulated samples were compared with a commercial sample to illustrate the enhancement of properties of new formulations. As illustrated in Figure 1, the viscosity of all the formulations decreases with increasing temperature. The viscosity of all the new formulations (samples 1 to 5) is 5 or more times less than the viscosity of commercial sample at room temperature (25 ^{0}C). This significant decrease in viscosity at room temperature will enable formulation processing at room temperature reducing considerable amount of cost saving to the industry.

Table 2 shows the values of hardness, HDT, and dielectric breakdown voltage of different silica-epoxy composites. The hardness, HDT, and dielectric breakdown voltage were measured according to the standards of ASTM D2240, ASTM D648, and ASTM D149 respectively. As shown in Table 2, the hardness of silica-epoxy composites did not change with the variation of silica type and concentration as the hardness is mainly governed by the specific epoxy resin. However, both HDT and dielectric breakdown voltage significantly improved of majority of new formulations compared to the commercial sample. Experimental results revealed that new epoxy formulations can enhance the heat distortion temperature and dielectric breakdown voltage of the epoxy composites up to 40% and 20% respectively compared to the commercial formulations.

Table 1: Compositions of various silica-epoxy formulations.

Sample No.	Epoxy resin (wt. %)	Silica particle concentration (wt. %)			Other fillers (proprietary) (wt. %)	Plasticizer and surfactant (wt. %)
		large micro	small micro	nano		
Commercial sample	28.5	57.0	-	-	10.7	3.8
Sample 1	28.5	38.0	19.0	0.0	10.7	3.8
Sample 2	28.5	28.5	28.5	0.0	10.7	3.8
Sample 3	28.5	37.0	18.5	1.5[1]	10.7	3.8
Sample 4	28.5	37.0	18.5	1.5[2]	10.7	3.8
Sample 5	28.5	37.0	18.5	1.5[3]	10.7	3.8

[1]*hydrophilic fumed silica grade - Aerosil 300VS*

[2]*Hydrophobic fumed silica grade - Aerosil R8200*

[3]*Aerosil R8200 and Aerosil 300VS in 2:1 ratio*

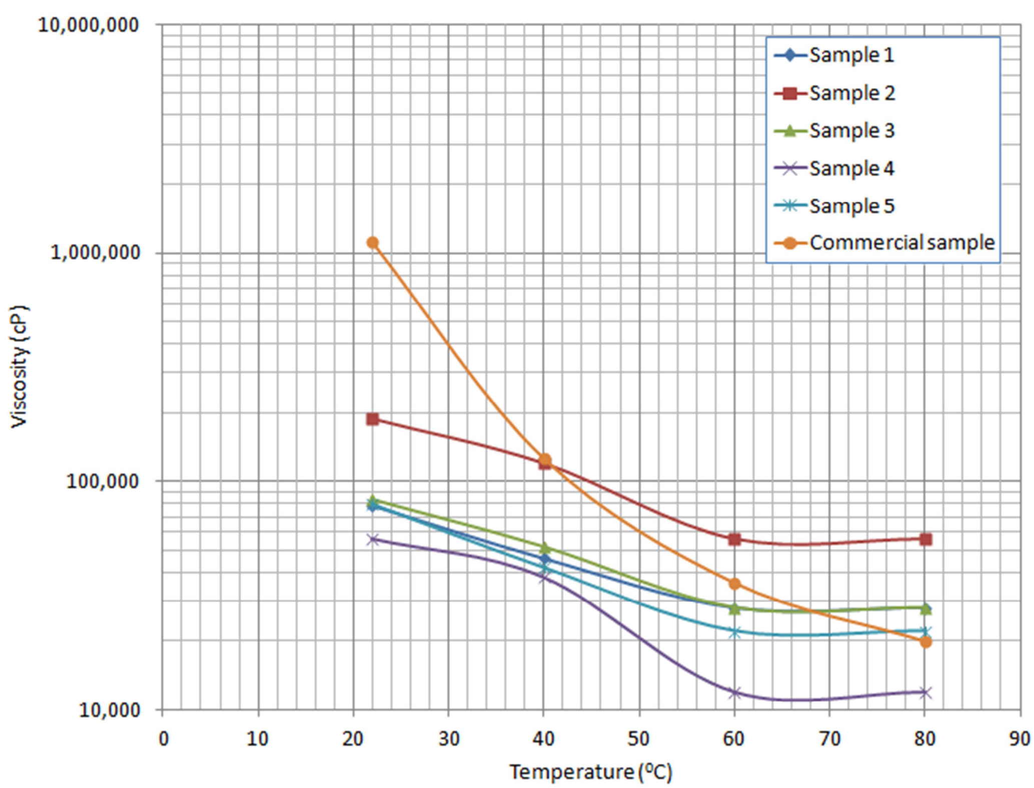

Fig. 1: Variation of viscosity of different formulations with increasing temperature.

Table 2: Hardness, HDT, and Dielectric Breakdown Voltage of various silica-epoxy composites.

Sample No.	Hardness	Heat Distortion Temperature (HDT)	Dielectric Breakdown Voltage (kV)
Commercial sample	95D	70^0C	47.0
Sample 1	95D	77^0C	47.1
Sample 2	95D	81^0C	54.9
Sample 3	95D	71^0C	56.2
Sample 4	95D	60^0C	54.0
Sample 5	95D	98^0C	54.9

IV. CONCLUSION

Present study revealed that new epoxy formulations using different size fillers in combination with electrospinning based mixing technology can significantly enhance the epoxy composite properties compared to the existing products. The viscosity of new formulations is 5 or more times less than the viscosity of commercial sample at room temperature (25 ^{0}C) as well as the heat distortion temperature (HDT) and electrical breakdown strength of epoxy composites improve up to 40% and 20% respectively compared to the commercial sample. These new epoxy formulations enable preferred viscoelastic and flow behavior, with higher filler loading levels and enhance product properties than are achievable with current formulations unlocking new applications.

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