

# Investigation on Thermal Aging and Mechanical Properties of the Nano-SiO<sub>2</sub>/Epoxy Adhesive

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*Abstract—Nano-inorganic/Epoxy composite is superior to pure Epoxy resin in the electrical and mechanical properties. In this paper, contents of 1wt% and 2wt% nano-Silica were added into Epoxy Tung-maleic Anhydride Adhesive Resin to study the electrical and mechanical properties of the composite. Firstly, the breakdown strength, flexural strength and impact strength of the composite are measured at room temperature. The experiment results indicate that with the increasing of nano-particle content, the breakdown strength of the composite increases, and 2wt% loading nano-Silica composite shows a better flexural and impact strength. Secondly, a thermal aging process which lasts for 168 hours, 336 hours, 504 hours and 672 hours respectively at 180°C is proceeded to the composite, and the breakdown strength after each thermal aging period is measured subsequently. The experiment results indicate that addition of nano-Silica improves the breakdown strength of adhesive resin before and the early period of thermal aging. However, the breakdown strength of 1% and 2% nano-Silica composite and adhesive resin trend to the same at the end of 28 days thermal aging. The research shows that the flexural and impact strength of Epoxy Tung-maleic Anhydride Adhesive Resin are effectively improved by the addition of nano-SiO<sub>2</sub> while the breakdown strength is improved slightly.*

*Keywords—Epoxy Tung-maleic Anhydride Adhesive Resin; Breakdown Strength; Thermal Aging ; Mechanical Property*

## I. INTRODUCTION

The main insulation of stator is the most important part of high voltage electrical machine insulation system, and it directly affects the technical and economic parameters of the electrical machine. At the same time, it determines the operating reliability and service life of the electrical machine to a large extent[1]. Stator insulation system of high voltage ac generator was asphalt-mica tape in the early stage. In 1964, Epoxy Tung-oil Anhydride Adhesive Resin(TOA) was developed and successfully used on the stator bar of 10.5kV hydraulic turbine generator and 13.8kV steam turbine generator. In the middle age of 1970s, the TOA system has already replaced the asphalt system. On the basis of TOA, Epoxy Tung-maleic Anhydride Adhesive Resin system was developed and the heat level improved from class B to class F. Its mechanical property such as flexural strength has greatly improved. By the mid 1980s, five kinds of insulation system have formed and Epoxy Tung-maleic Anhydride Adhesive

Resin system is the most representative one among them. It has become the main insulation system of electrical machine in China since it first came into use on 15.75kV and 210MW steam turbine generator. Now, it is used on nearly all the hydraulic Turbine generator, including the three gorges generators[2].When Epoxy Tung-maleic Anhydride Adhesive Resin is used in electrical machine insulation, it is important to maintain its long-term stability. Epoxy Adhesive is a kind of thermal-setting resin and its cured products have good heat resistance. However, it has high cross-linking density and presents a three-dimensional reticular formation after the curing, which makes it have a larger internal stress leading to crack. Adhesive resin plays an important role in the electrical apparatus, and is also the most common part to suffer aging. On the one hand, electrical aging and thermal aging occurs during the operating of the apparatus, which produce various forms of degradation such as color change, electrical and mechanical performance decline, etc. The degradation caused

the failure of insulation system, and eventually induce the fault of electrical machine. On the other hand, main insulation will be damaged under the influence of electrical stress due to suffering partial discharge, and thermal aging, also mechanical stress. In order to reduce the thickness of the main insulation and improve mechanical properties, the improvement of electrical breakdown and mechanical strength of Epoxy Tung-maleic Anhydride Adhesive Resin is still required. The development of nanotechnology provides a new way for modifying traditional polymer materials and developing new materials. Nano-Silica is a kind of high quality inorganic filler which has excellent performance such as high heat resistance and low stress concentration[3]. The large surface area of nano-Silica has the special structure of unsaturated bond and dangling bond, and has very large surface activity. It is able to modify the micro zone of adhesive resin by careful controlling the dispersion of nano-Silica and recombination with polymer matrix. The comprehensive performance of composite can be effectively improved with only a little volume fraction of inorganic nano-particle. As a result, modifying with nano-Silica will become an effective way to improve thermal aging and mechanical properties of the polymer. Liu added nano-Silica into epoxy resin to study mechanical property of the composite. The experiment results indicate that tensile strength of 3wt% loading nano-Silica composite increases by 27.25% and flexural strength increases by 24.56%[4]. Zhu also finds out that 3wt% loading nano-Silica composite has a better performance in flexural, impact and breakdown strength[5]. Martin Reading added nano-Silica into a kind of epoxy resin with very high breakdown strength, although the breakdown strength of the composite does not increase obviously, a better shape parameter is found[6]. In this paper, nano-Silica was added into Epoxy Tung-maleic Anhydride Adhesive Resin in order to improve the properties of heat aging resistance and toughness of the matrix.

## II. EXPERIMENT

In this paper, 1wt% and 2wt% content of nano-Silica were mixed into Epoxy Tung-maleic Anhydride Adhesive Resin to study the electrical and mechanical properties of the composite. The diameter of nano-Silica is 15nm. Adhesive

resin is a mixture which contains curing agent and a large quantity of acetone, and this makes adhesive resin have a very low viscosity. It is investigated that particles surface treatment also affects the performance of composite. In order to make the composite have a good dispersion, nano-Silica was modified with coupling agent before mixing into adhesive resin. A change occurred at the nano-Silica surface because of the action of the coupling agent, and a layer was formed around the filler to prevent agglomeration. This is a common method of surface modification to nano-Silica.

### A. Preparation of Test Sample

Firstly, a certain quantity of nano-Silica was dispersed into acetone by ultrasonic method to produce dispersion liquid, and stir the dispersion liquid at a constant speed made it form into a paste. Secondly, the paste was mixed with adhesive resin, after 2 hours mechanical stirring, the mixture was put into a vacuum drying oven. Air bubbles and a small amount of residual acetone, which is volatile liquid of boiling point 56°C, were removed at 120°C in 2 hours. At last, the composite was poured into molds. The curing time was 10 hours and curing temperature was 170°C. Samples with 50mm in diameter and 1mm in thickness were made for breakdown property test. The same method was used to make the samples with 120mm in length, 15mm in width and 10mm in height shown in Fig.1 for mechanical property. Smooth surface and without bubbles and impurities inside were required for all the samples.

### B. Thermal Aging Test

Thermal aging test is taken according to IEC 60216-8 and the recommendation of dielectric heat resistance level[7,8]. Epoxy Tung-maleic Anhydride Adhesive Resin is a class F insulation, and the accelerated thermal aging test was proceeded at 180°C. A thermal aging process of the composite which lasted for 168 hours, 336 hours, 504 hours and 672 hours was proceeded respectively, and the breakdown strength after each thermal aging period was measured subsequently. Four samples were measured after each thermal aging period. During the test, both of the sample and electrodes were put into the transformer oil to make sure a good insulating environment[9].

**C. Flexural Property Test**

Flexural property test is taken according to ISO 178:1993[10]. The sample was supported on the testing machine as a beam. A force was exerted on the center of sample at a constant speed of 1mm/min. Data was recorded in the computer when the pressure reached to a maximum. Stress is calculated as in equation(1)

$$\sigma_f = 3FL/2bh^2 \quad (1)$$

where,  $\sigma_f$  is stress, MPa; F is force, N; L is span, mm; b is sample width, mm; h is sample thickness, mm.

In each nano-Silica content, five samples were tested, and the average flexural strength was calculated.

**D. Impact Property Test**

Impact property test is taken according to ISO 179-1:2000[11]. The sample was impacted by a pendulum bob at a constant speed. The impact line was through the midpoint of the sample that was supported at two ends. The work consumed in thrusted sample was recorded. Stress is calculated as in equation(2)

$$\sigma_i = A/(b \times d) \quad (2)$$

where,  $\sigma_i$  is stress, kJ/m<sup>2</sup>; A is work, kJ; b is sample width, mm; d is sample thickness, mm.

Five samples of each nano-Silica content were tested, and the average impact strength was calculated.



Fig.1 Sample for mechanical property

**III. EXPERIMENT RESULTS AND ANALYSIS**

**A. Results of Breakdown Strength**

Samples that before and after the thermal aging are shown in Fig.2. During the aging, the color of sample changes from light yellow into dark brown. The variation of color is obvious. Results of breakdown strength are shown in TABLE.I. A curve according to TABLE.I is shown in Fig.3. When the breakdown strength of the composite is measured at room temperature, the experiment results indicate that with the increasing of nano-Silica content, the breakdown strength

of the composite increases. The aging time is separated into four periods. In the first period, breakdown strength of three kinds of sample decreases slightly. In the second period, breakdown strength keeps decreasing. And the drop rate is much bigger than before. During the first two periods, the breakdown strength of adhesive resin and composite both decrease. 1wt% loading nano-Silica composite decreases from 34.65kV/mm to 28.68kV/mm, 17.3% lower than that before aging. 2wt% loading nano-Silica composite decreases from 36.5kV/mm to 30.9kV/mm, 15.3% lower than that before aging. But adhesive resin decreases from 32.95kV/mm to 30.51kV/mm, only 7.4% lower than that before aging. Breakdown strength of the composite decreases faster than the unmodified adhesive resin. At the end of the second period, breakdown strength of 1wt% loading nano-Silica composite is lower than that of adhesive resin, while breakdown strength of 2wt% loading nano-Silica composite and adhesive resin are almost in the same level. So 2wt% loading nano-Silica composite is superior to adhesive resin in breakdown strength in the first two periods.



Fig.2 Samples that before and after the thermal aging

TABLE I. RESULTS OF BREAKDOWN STRENGTH

	0%	1%	2%
0 hour aging	32.95	34.65	36.49
168 hours aging	31.42	33.05	35.71
336 hours aging	30.51	28.68	30.90
504 hours aging	33.84	32.91	30.03
672 hours aging	30.46	29.96	30.20

Unit: kV/mm

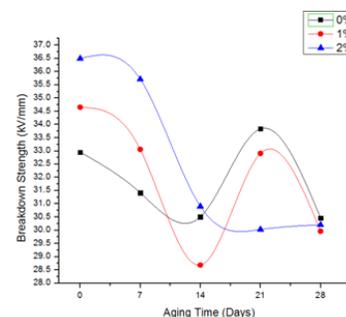


Fig.3 Results of breakdown strength

After the third aging period, the breakdown strength of 1wt% loading nano-Silica composite and adhesive resin increases over the 2wt% loading nano-Silica composite which keeps constant. However, the breakdown strength of the three kinds of sample are almost in the same level at the end of thermal aging.

To sum up, the addition of nano-Silica improves the breakdown strength of adhesive resin before and the early period of thermal aging.

**B. Results of Flexural and Impact Strength**

The flexural and impact strength of the composite are measured at room temperature. Results of flexural and impact strength are shown in TABLE.II. A curve according to TABLE.II is shown in Fig.4. The experiment results indicate that flexural strength increases from 49.78MPa to 61.89MPa, 24.32% higher than that of the matrix. Impact strength increased from 19.3kJ/m<sup>2</sup> to 21.3kJ/m<sup>2</sup>, 10.36% higher than that of the matrix. Nano-Silica has many active groups at its surface, and therefore it has a close integration with the adhesive resin. When affected by external force, the particles do not fall off easily. Besides, a lot of micro deformation zones are formed to absorb external energy of developing crack because of the interaction with stress field[12]. The micro deformation zones have an effect on dispersing stress which can produce main crack, and achieving the goal of inhibiting the main crack. Another theory about the micro deformation zones was put forward by Liu who believes that the matrix around the particles will yield to absorb much external energy with the formation of micro deformation zones[13]. So the composite is better than the matrix to pass on the external stress to trigger the composite yield. This behavior consumes a large amount of impact energy or flexural energy so as to get the function of enhancing toughness. So we conclude that addition of nano-Silica improves the toughness of adhesive resin.

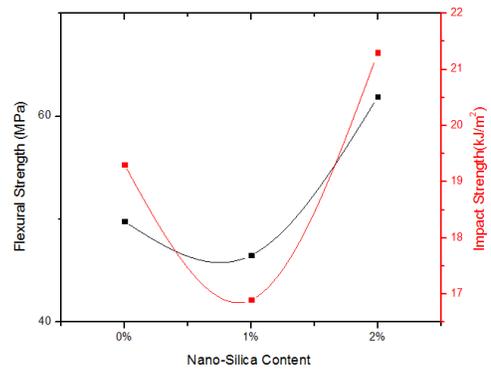


Fig.4 Results of flexural and impact strength

It also can be seen that strength of the composite decreases when 1wt% nano-Silica is added to the adhesive resin. This may be because of the stress concentration according to Tahir who believes that the ultimate strength of a composite is determined by the fracture path throughout the materials[14].

The 1wt% loading nano-Silica composite may not be cured completely as discussed in the breakdown strength analysis. So, the curing agent usage may also be a factor of reducing the mechanical property of 1wt% loading composite.

**IV.CONCLUSIONS**

In this paper, contents of 1wt% and 2wt% nano-Silica were added into Epoxy Tung-maleic Anhydride Adhesive Resin to study the thermal aging and mechanical properties of the composite. Thermal aging test, flexural property test and impact property test were conduct according to the corresponding standard.

Addition of nano-Silica improves the breakdown strength of adhesive resin before and the early period of thermal aging. However, the breakdown strength of 1% and 2% nano-Silica composite and adhesive resin trend to the same at the end of 28 days thermal aging. The mechanical property test indicates that the flexural and impact strength property of 2wt% loading nano-Silica composite increase by 24.32% and 10.36% respectively.

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TABLE II. RESULTS OF FLEXURAL AND IMPACT STRENGTH

	0%	1%	2%
Flexural Strength	49.78	46.5	61.89
Impact Strength	19.3	16.9	21.3

Unit:MPa kJm<sup>2</sup>

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