Hybrid Composites of Polyamide-Imide and Silica Applied to Wire Insulation

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ABSTRACT Aromatic polyamide-imide resins (PAI) based on Trimellitic anhydride and Diphenylmethane-4,4'-diisocyanete were modified by an oligo-alkoxysilane compound to obtain novel silane-modified PAI. Then, PAI-silica hybrid films were prepared by drying and curing the silane-modified PAI. Increasing the silica unit content gives the hybrid films a higher Young's modulus and a higher tensile strength than the original PAI film, however, maximum elongation is maintained. Furthermore, they have much lower moisture absorption than PAI film under wet conditions. Moreover, this hybrid film has been found to have a different dielectric constant than the conventional film. This report describes properties of silane-modified PAI varnish, which is used as an insulation layer for enameled wire.

1. INTRODUCTION

PAI dissolves in polar solvents such as N-Methyl-2-pyrroridone, and it is widely used for insulation, such as a magnet wire insulation and polymer-coated copper foil for circuit boards. Moreover, PAI is a heat-resistant resin, which can be molded. It is used industrially for its outstanding electrical properties and mechanical characteristics as an engineering plastic. Thus, PAI has both processability and heat resistance, and its synthesis is comparatively easy. That is why further improvements to its various characteristics can be expected. PAI have the features mentioned above, but their moisture absorption is still not as low as other plastics. On the other hand, PAI is used as enameled wire insulation, because it exhibits good heat-resistance and moisture-resistance [1]. However, its heat-resistance should not be compared with a polyimide resin, for example Kapton. Moreover, PAI has good chemical stability against water, but water absorption is high, therefore, it cannot be used for materials that require a stable dielectric constant.

We examined these various problems associated with PAI. We synthesized a novel silane-modified PAI prepared with an oligo-alkoxysilane compound. The novel silane-modified PAI was cured to make a PAI-silica hybrid. Some properties of the PAI-silica hybrid are better than those of conventional PAI. Comparing enameled wires made from conventional PAI and PAI-silica, we found that the PAI-silica hybrid has different properties from the conventional PAI. In particular, silica has an effect on Young's modulus, maximum elongation rate, and glass transition temperature of cured film.

This paper describes the developments and the properties of PAI-silica hybrid, and the properties of this enamel when applied to magnet wires.

2. SYNTHESIS OF PAI-SILICA HYBRID

The methods of adding silica powder to enamel are well known. The general method is to mix fine silica powder to a varnish for dispersal in a polymer film. In this case, as it is often difficult to disperse silica and varnish, a surfactant is used in combination. This method has a wide range of applications, because it is possible to use any kind of polymer solution. Hitachi Cable Company Japan has introduced an example of silica blended with polyimide for the insulation of magnet wire. In this example, silica is added to raise the heat-resistance of the polyimide.

We recently developed a new method of preparing PAI silica hybrid. According to this method, it is possible to introduce silica selectively to the polymer end of the PAI.

This method is superior to mixing silica powder to varnish, except for restrictions on the kinds of solvent and the polymer. The film properties prepared by this method were superior to the above-mentioned method.

The following are the fabrication hybrid of the PAI and silica, and the properties of PAI-silica hybrid [2)~4)].

3. EXPERIMENTAL METHOD

3.1 Preparation of Varnish and Wires

Generally, PAI varnish is polymerized by Trimellitic anhydride and Diphenylmethane-4,4'-diisocyanete in polar solvents [5]. The base polymer was made to react at an

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excess molar ratio of TMA (Scheme 1), because there are carboxyl groups at the end of polymer chain to be modified with a silane. The oligo alkoxysilane compound was reacted with the carboxyl groups. As a result, a silane-modified PAI with an alkoxysilane part at the polymer end was created (Scheme 2).

An enameled wire was obtained by coating and baking this silane-modified PAI varnish onto copper wire. During baking, the alkoxysilane part of a polymer end is cured to silica by a Sol-gel process. The PAI polymer chains are intermolecular cross-linked at the silica site of the chain end. Through a series of these steps, the PAI-silica hybrid insulation film was formed (Scheme 3).

We evaluated the magnet wires made from the conventional PAI and three kinds of PAI-silica hybrid. Table 1 gives the types of insulation layer of the magnet wires.

3.2 Evaluation

3.2.1 Wire Properties

Table 2 gives the general properties of the magnet wires. A conventional PAI varnish is used for the magnet wire used in the comparison. Table 2 shows that the abrasion resistance of PAI-silica hybrid wire is improved. However, a difference was not seen in electrical properties such as breakdown voltage.

3.2.2 Film Mechanical Properties and Others

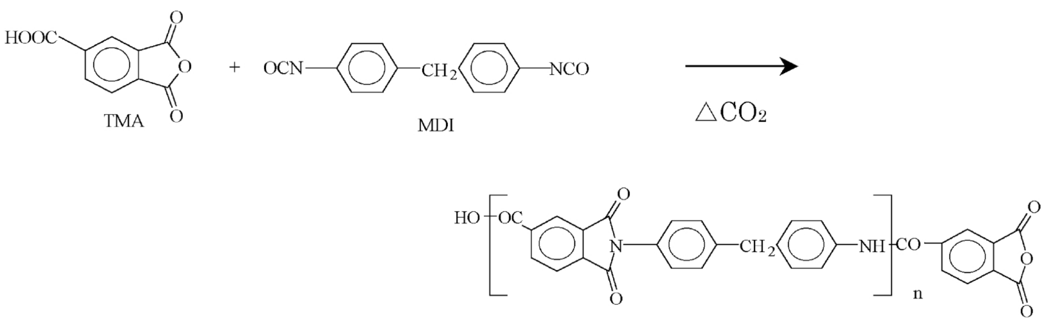
The insulation film of the magnet wire was evaluated after removing it from the wire. The evaluation method is as follows. A current etches the conductor and only the sheath survives. The insulating sheath is taken from the wire. This is the same method as that for making a tube for a pinhole test. Then, elongation and other properties of the sheath are evaluated.

Table 1 Type of magnet wires evaluated.

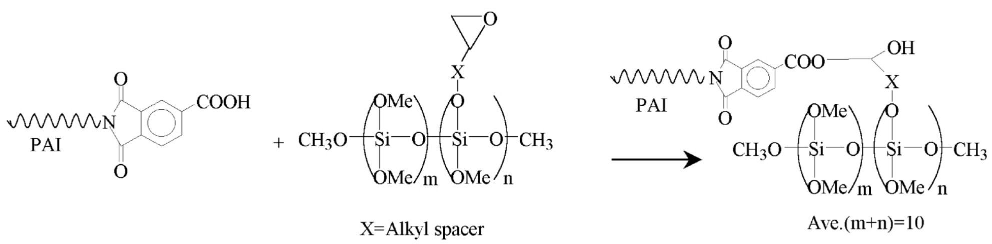
Type	Symbol
Conventional PAI	C-AI
Silica contents 1.4 % PAI	14Si-AI
Silica contents 2.0 % PAI	20Si-AI
Silica contents 4.0 % PAI	40Si-AI

Table 2 General properties of magnet wires evaluated.

	C-AI	14Si-AI	20Si-AI	40Si-AI
Bare wire (mm)	1.000	1.000	1.000	1.000
Film thickness (mm)	0.034	0.035	0.035	0.035
Break down voltage (kV)	12.2	13.3	13.5	13.6
Cut through temparature (°C)	420	425	435	435
Bi-directional abrasion (times)	210	245	280	310
Uni-directional abrasion (N)	26	30	33	33



... Scheme 1



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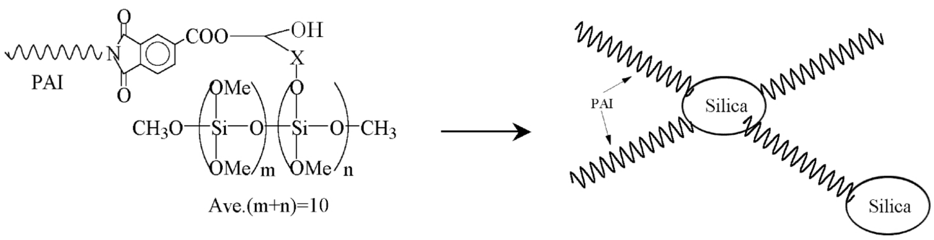

... Scheme 3

Table 3 gives the mechanical properties. From this result, the insulation film of the PAI-silica hybrid wire has a higher Young's modulus with the content of silica. In addition, the maximum elongation rate of the films is stable and there is no change with silica content, even when the modulus is higher.

Furthermore, a Differential Scanning Calorimeter (DSC) is used to measure the glass transition temperatures of these tubes. As a result, a difference in the glass transition temperature is observed with the content of silica. In addition, the energy gap at the glass transition is also different.

The moisture absorption of films was investigated. Table 4 shows the results. The result shows that the moisture absorption changed with silica content.

3.2.3 Electrical Properties

The dielectric constant of the film from the wire was evaluated (Table 5). The dielectric constant decreases as silica content increases. There is a large difference compared to the conventional PAI. General data on the dielectric constant show that polyimide is about 3.5 to 4.0 and PTFE is about 2.0 to 2.5. As a result of this evaluation, the dielectric constant was found to be higher than PTFE, but lower than polyimide (Figure 1).

4. DISCUSSION

The result shows the silica content and the intensity of the film are correlated. Because the correlation of the silica content and the Young's modulus of the film is especially clear, it can be presumed that the silica content is also related to abrasion resistance.

The silica content is related to the improvement in the Young's modulus, but it did not affect the flexibility of the film. It is considered that a difference exists in the cross-linking system of the polymer chain [6]. The PAI chains of the PAI-silica hybrid are cross-linked through the silica located at the chain ends, but its polymer does not make a cross-linkage at the middle of a polymer chain (Figure 2 (A) and (B)). We think the flexibility of PAI was maintained because movement of a PAI chain was not prevented. Consequently, the glass transition temperature measured using DSC may be influenced.

The moisture absorption of a film may depend on the silica content of the film. Another possibility is that it may be dependent on the location of entangled polymer chains. It is impossible to judge which is correct from these data.

The PAI-silica hybrid shows a lower dielectric constant than polyimide. It is known when silica particles are located in a polymer film, the dielectric constant decreases. The amount of silica may influence the dielectric constant. On the other hand, the influence of the cross-linkages of a polymer chain can also be considered.

Table 3 Mechanical properties of insulation films.

	C-AI	14Si-AI	20Si-AI	40Si-AI
Tensile Strength (MPa)	89.2	96.1	99.0	103.0
Elongation (%)	22.1	21.6	20.9	21.8
Young's modulus (GPa)	2.68	3.19	3.25	3.42

Table 4 Glass transition temperature and moisture absorption of insulation films.

	C-AI	14Si-AI	20Si-AI	40Si-AI
Tg (°C)	285.4	294.6	301.5	300.9
Moisture absorption at 20 % RH-24 h (%)	1.8	2.1	2.1	2.2
Moisture absorption at 95 % RH-24 h (%)	5.6	2.6	2.4	3.2

Table 5 Dielectric constant of insulation film.

	C-AI	14Si-AI	20Si-AI	40Si-AI
Dielectric constant at 1 kHz-23°C	3.9	3.7	3.3	3.0

5. CONCLUSION

The newly developed PAI-silica hybrid film (tube) prepared from magnet wire has an enhanced Young's modulus compared to conventional PAI film. This results in enhanced properties for unidirectional abrasion and bidirectional abrasion, thereby providing high windability. The improvement in Young's modulus has contributed to improving the abrasion resistance of the magnet wire.

The moisture absorption of the film may depend on the silica content of the film. A 2 % silica content minimizes water absorption.

The dielectric constant is influenced by the quantity of silica.

To protect the insulation layer from corona, various investigations are performed by adding inorganic powder to enameled wire insulation [7]. It is necessary to add about 10 to 30 % of inorganic powder to insulation resin to pro-

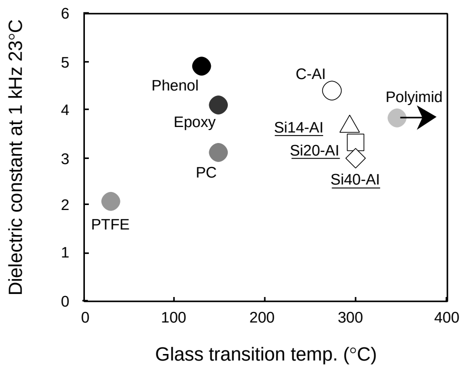

Figure 1 Relationship between glass transition temperature and dielectric constant of various polymers.

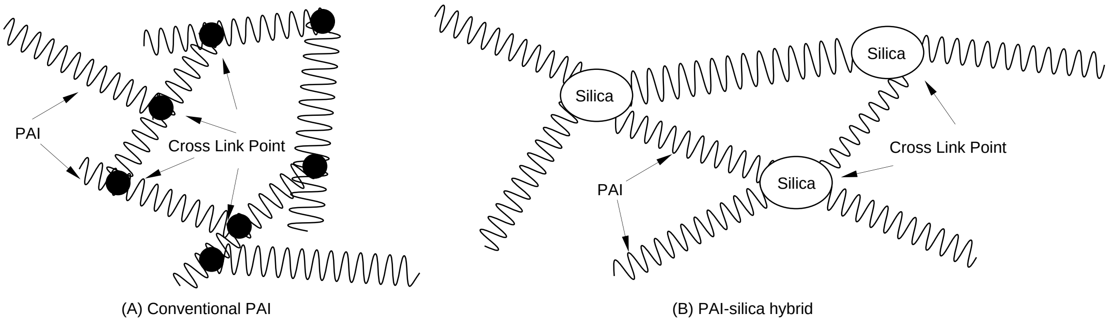

Figure 2 Cross-linking model of conventional PAI and PAI-silica hybrid.

tect the insulation from corona. When silica is formed the combination rate is 4 % at maximum in the examined silica hybrid PAI. It is thought to be possible by combining the method of adding inorganic powder for this silica hybrid PAI and others to protect the insulation from corona.

The magnet wires using the PAI-silica hybrid, which has improved abrasion resistance, are equivalent to conventional PAI wires in other properties, thus application to the same areas as before is possible. Furthermore, application to other areas, which efficiently employ the different low moisture absorption characteristics and dielectric constants, is also possible.

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