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# Effects of silane coatings in aqueous and non-aqueous media on the properties of magnesia filled PTFE laminates

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ABSTRACT

Silane coated on magnesia particulates in isopropyl alcohol (IPA) medium, precluded the formation of brucite phase, which hitherto existed in silane coated on magnesia filler in aqueous medium. Both the types of coated MgO powders were separately filled in the PTFE matrix, and microwave flexible substrates were made by the process comprising of sigma mixing, extrusion, calendering and hot pressing (SMECH process). Filler content in the PTFE matrix was varied from 10 to 70 wt% and microstructures of the composites were analyzed using SEM technique. The dielectric properties of the planar substrates were measured at X-band using waveguide cavity perturbation technique. The IPC-TM-650 procedure was employed to evaluate the moisture absorption characteristics.



## 1. Introduction

Ceramic powder filled polymer laminates are being used as base substrates for the fabrication of microwave devices. Some of their advantages over the conventional ceramic substrates are (i) the tailoring of dielectric properties by adjusting the fractions of the constituents and (ii) multilayer structure fabrication even in curved shapes [1]. The main applications of flexible substrates are in patch antennas, mobile base stations, global positioning systems (GPS), high power amplifiers, etc. [1,2]. In order to obtain filled polymer substrates having wide range of properties, various polymers such as Polyphenylene sulphide (PPS), Polytetrafluoroethylene (PTFE) and Epoxy, and fillers ranging from low dielectric constant ($\varepsilon_r \sim 3.8$) silica to high dielectric constant ($\varepsilon_r \sim 270$) strontium titanate have been explored [3–6]. It is found that compositional variations in fillers, their particle sizes, distributions, morphologies, processing techniques, etc. have significant influence on the electrical, thermal and mechanical properties of the composite substrates. Prior to incorporation in the polymer matrix, often particulate fillers are silane coated in order to have much effective filler and polymer inter-coupling and also to prevent moisture absorption by the filler, which adversely affects the substrate properties [7,8].

In our earlier work [7], various physical properties of the magnesia filled PTFE substrates, prepared using silane coated magnesia in aqueous medium, were presented. For the magnesia filled substrates, the results revealed much inferior properties such as low filling fraction, poor density and inferior dielectric properties when compared to that of the alumina filled ones. However, interestingly, the as-received-magnesia (uncoated magnesia) filled PTFE composites exhibited relatively less inferior properties than the hitherto silane coated ones. On XRD investigation of the silane coated magnesia, it was found that a secondary brucite phase essentially formed during the aqueous based silane coating process. Hence, it is concluded that the brucite phase was the reason behind the inferior properties seen in the silane coated magnesia filled composites [7]. The process of aqueous medium silane coating that was employed for magnesia filler was reported for the surface treatment of many inorganic oxides [5,6,8–11]. In the present study, silane coating of magnesia was carried out in isopropyl alcohol (IPA) medium instead of aqueous medium in an effort to avoid the formation of undesirable brucite phase. Similar to the earlier work [7], the silane coated MgO filler was incorporated in the PTFE matrix through SMECH process comprising of sigma mixing, extrusion, calendering followed by hot pressing, and composite laminates were fabricated. In the present work, the end properties such as dielectric constant, loss tangent and moisture absorption were evaluated and compared, for the filled PTFE substrates that were fabricated using MgO fillers silane coated in (i) the aqueous and (ii) the non-aqueous IPA media.

## 2. Experimental

### 2.1. Materials

The starting materials were 50 μm size PTFE powder, H71 grade (M/s Hindustan Fluoro Carbons, India), 2 μm MgO powder (M/s Dead Sea Bromine Group (DSBG), France) and short E-glass fiber (filament size ϕ10 μm, length 4–6 mm) (M/s Binani Glass, India). Phenyltrimethoxysilane (M/s Sigma–Aldrich, USA) and

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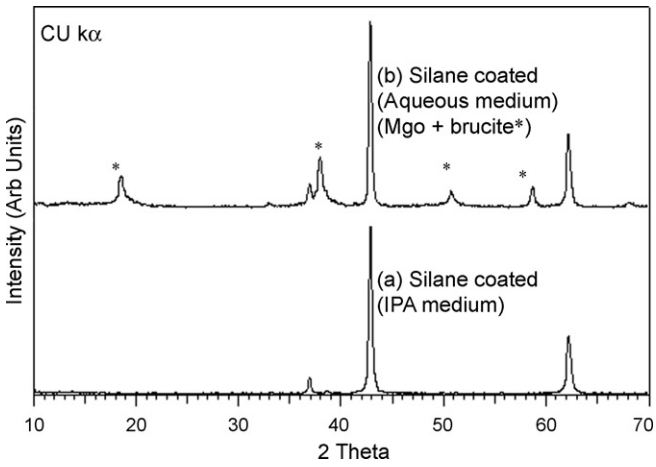


**Fig. 1.** Comparison of the XRD patterns of the magnesia fillers silane coated in (a) the IPA medium and (b) the aqueous medium.

Di(propyleneglycol) (M/s Merck, Germany) were used, respectively, as silane coating agent and lubricant. AR grade isopropyl alcohol (IPA) (M/s Merck, India) was used as silane coating medium.

### 2.2. Processing

The aqueous based sol–gel process of silane coating is a well established experimental procedure for the surface treatment of ceramic fillers, and is often employed to prevent moisture absorption in inorganic fillers such as fused silica, alumina, rutile and $CaTiO_3$ [5–11]. However, the aqueous base silane coating process employed for MgO filler resulted in substrates with inferior properties due to the formation of brucite phase. Since brucite has poor density and inferior dielectric properties, the composite system fabricated using aqueous based silane coated filler could considerably deteriorate the end properties of the composites made out of them. In order to avoid brucite phase formation during silane coating of MgO filler, the as-received-MgO was silane coated in non-aqueous isopropyl alcohol (IPA) medium and the amount of water was restricted just for hydrolysis of the phenyltrimethoxysilane (PTMS).

MgO filler, heat treated at 600 °C, was dispersed in distilled IPA and stirred well using a heavy duty stirrer. To the stirred suspension, calculated amount of PTMS and water were added. The amount of PTMS was just enough to give 1 wt% silane coating on the filler, and amount of water was controlled exactly for the hydrolysis of PTMS. The stirring was continued for 1 h. The slurry thus obtained was dried at 100 °C and powdered. The XRD pattern and DSC/TGA plots of the silane coated (IPA medium) magnesia filler were recorded to investigate the presence of brucite phase in it and compared these results with those of the MgO coated in the aqueous medium.

PTFE/MgO composites were fabricated through the SMECH process as described in our earlier work [5–7,12]. The filler fraction was varied in the PTFE matrix from 10 to 70 wt% in successive steps of 10 wt%; and near to the maximum observed value of $\varepsilon_r$, fine tuning of filler fraction was carried out to find out the optimum loading. Based on the earlier experiments [5–7,12], 2 wt% of microfiber glass was used as the reinforcing agent during composite preparation.

The microwave dielectric properties of the samples were studied using waveguide cavity perturbation technique [13]. Agilent make PNA E8362B Vector Network Analyzer was used in the X-band (8.2–12.4 GHz) region for the measurements. Surface morphology and filler distribution in the composite samples were analyzed using M/s Philips (The Netherlands) make SEM (Philips XL-30) with an accelerating voltage of 15 kV. Samples were sputtered with gold layer and planar surface was analyzed. Moisture absorption in the composites was determined as per IPC-TM-650 2.6.2 standard method.

## 3. Results and discussion

The XRD patterns of the magnesia filler silane coated in (a) IPA medium and (b) aqueous medium are shown in Fig. 1. It is clear from the figure that besides the MgO phase, there are peaks corresponding to the brucite phase in the silane (aqueous medium) coated magnesia, whereas silane coated magnesia (in IPA medium) shows peaks only due to MgO.

Fig. 2(a) and (b) show the DSC and the TG curves for magnesia fillers silane coated in aqueous and IPA medium, respectively. TG curves of both the fillers show that aqueous silane coated magnesia filler has 18% weight loss which is attributed to the expulsion of

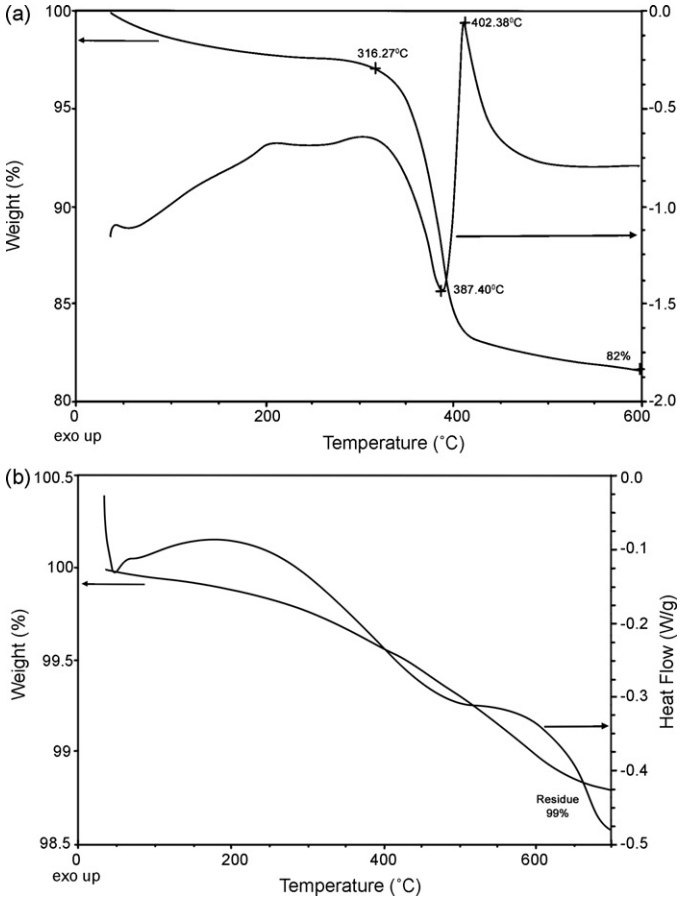


**Fig. 2.** DSC/TG curves of silane coated magnesia filler in (a) aqueous medium and (b) IPA medium.

adsorbed water molecules and the hydroxyl groups in the brucite phase. Whereas in the case of magnesia filler silane coated in IPA medium, the weight loss is merely 1 wt% and the brucite content has been considerably reduced. Hence, it can be concluded that the silane coated magnesia filler in IPA medium can be gainfully used as filler for the fabrication of filled PTFE substrates.

Variations in dielectric properties and moisture absorption of the filled PTFE composites with silane coated (IPA medium) magnesia were found out and compared with those of the composites fabricated using silane (aqueous medium) coated MgO. Fig. 3 shows the planar SEM micrographs of the both silane (aqueous and IPA media) coated magnesia filled [50 wt% loaded] composite substrates. The micrograph clearly shows highly dense microstructure with uniformly distributed MgO filler in the PTFE matrix. Compared to silane (aqueous medium) coated magnesia filled substrates, a better matrix-filler interface together with lesser pores are observed which should lead to enhanced electrical, thermal and mechanical properties.

Variation of dielectric constant of the silane coated (IPA and aqueous media) magnesia filled PTFE composites is shown in Fig. 4. The dielectric constant is found to increase non-monotonically as a function of filler loading in both the cases (Fig. 4). This is consistent with the trend seen in the other composites [5–8,12]. The optimum filler loading is a critical requirement for polymer–ceramic composite substrates to exhibit the best reproducible end properties. In order to find out the optimum filler loading for silane coated (IPA medium) magnesia filled PTFE composites, fine tuning has been carried out (>50 wt% loading) by varying filler loading with an interval of 2 wt%. It can be seen from Fig. 4 (inset) that the filling fraction has indeed been improved to 66 wt%, whereas, for