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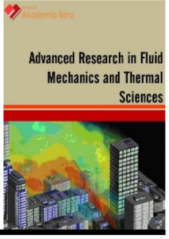

Effect of Silica (SiO_2) Filler on Thermal Stability of Polysiloxane Composites

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ABSTRACT

Thermal stability of composites are influenced by the types and properties of filler used. In this study Silica (SiO2) namely CS was used to improve the thermal stability of Polysiloxane (PoS) panel. The PoS/CS composites were fabricated by mixing PoS and CS with filler loading of 2wt% to 12 wt%. PoS/CS composites are then cast using close mold technique and cured at room temperature (RT), 65˚C and 100˚C. The CS were characterized for X-Ray Diffraction (XRD), X-Ray Fluorescence (XRF), and Fourier Transform Infrared (FTIR) technique. The thermal stability of PoS/CS composites were identified via Thermal Gravimetric Analysis (TGA). It was found that, CS improved the thermal stability of PoS by increased decomposition temperature and decreased relative mass loss percentage. CS as a filler, replaced and reduced the hydroxyl group in the composites panel to significantly improved the thermal stability. Hence, the good thermal stability of CS also helps to improve PoS composites thermal properties. The interference of hydroxyl crosslink during curing was also found to affect the thermal stability. Thus, the high temperature curing (65˚C and 100˚C) were indeed unstable due to disruption of cross link process and thus affect the filler dispersion and cause aggregations. PoS/CS/RT composites were found to show the stable and linear profile of thermal stability compare to PoS/CS/65˚C and PoS/CS/100 ˚C. Thus the thermal stability of polysiloxane had improved by using CS as a filler and cured at RT.

1. Introduction

Polysiloxane (PoS) are categorised as an elastomer material in the silicon rubber group. Silicon rubber service temperature ranged from 200˚C to 250˚C with decomposition occurring 395˚C to 410˚C [1]. In previous studies, polysiloxanes were mixed with various filler in order to increase the properties. The thermal properties of Polysiloxanes are also able to be decreased by mixing the matrix of polysiloxanes with filler to develop composite materials [2].

Thermal properties explains the characteristic of material during application high temperature. The high thermal properties of material means higher ability of material to service at high

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temperature. Thermal properties of materials were determine through material decomposition temperature and its relative mass loss via thermal gravimetric analysis (TGA) test [3].

Silica (SiO_2) are widely used as a rubber filler especially in the tyre industries to increase wear and tear properties of rubber by improved bonding mechanism especially at high temperature service [4]. SiO_2 is one of the ceramic material that suitable to be used as a filler.

Silica is an elementary component of granite, soil, sand and other minerals [5]. The most common phase of Silica is Quartz. Tridymite and Cristobalite are the two other phases of Silica. The Silica used as filler may be in particulate form. Silica offer high thermal properties with melting point at 1700˚C to 1800˚C, and as a filler, the high thermal properties were required to enhance the thermal stability of materials [6].

The present of PoS/CS composites work is mainly concerned with thermal stability. In this paper, we first reported the preparation of PoS/CS composites with filler loading at 2wt% - 12wt% and cured at room temperature (RT), 65˚C and 100˚C. Thermal stability of those fabricated composites were investigated through Thermal Gravimetric Analysis (TGA).

2. Methodology

2.1 Material

Filler of Silica (CS) powder used in this study was purchased from Unahan-Chem, Korea. Polysiloxane (PoS) manufactured by Dow corning from USA were used as a composite's matrix

2.2 Preparation of Composites Panel

Firstly, polysiloxane (PoS), catalyst, and CS were agitated. Air bubbles were removed through vacuum chamber. Homogeneous mixture of PoS/CS were casted *via* closed mold technique. The fabricated samples dimension were $300x300x3mm^3$. The PDMS/CS composites then were cured at different curing temperature i.e; room temperature (RT), 65˚C, and 100˚C. Those PoS/CS composites were designated PoS/CS/RT, PoS/CS/65˚C and PoS/CS/100˚C

2.3 Test Method

X-ray Diffraction (XRD)

The purpose of this analysis is as to characterise the silica phase and structure of CS. XRD Bruker D8Advance (USA) was used to conduct the analysis.

X-ray Fluorescence (XRF)

The compositional element of CS were determined via XRF of Bruker S4 Pioneer (USA)

Fourier Transform Infrared (FTiR)

Molecular bonding characterization was determined through Spectrum peaks and molecular wavelength data which constructed through conducting FTiR *via* Perkin Elmer FTIR Spectrum 100 Spectrometer, USA

Thermal Gravimetric Analysis (TGA)

Thermal decomposition and relative mass loss of polysiloxane composites to determine thermal properties were identified through TGA using Linseis Thermobalance D8762 SELB (Germany)

3. Results and Discussion

3.1 Phase Analysis

Crystalline phase analysis was determined *via* XRD method. Spectrum curve and peak patterns of intensity vs 2 theta were constructed by using analyzed data. XRD spectrum pattern of CS as showed as per Figure 1. The patterns showed eight of main peaks that signifies phase of CS. Those peak patterns meets JCPDS file 00-046-1045 for Quartz. Eight peaks of crystalline phase of quartz at 21°, 26°, 36°, 39°, 43°, 46°, 50°and 55. As shown in Table 1. this pattern trends and intense peak degree of CS is similar to the findings in [7], which also used CS as their study. In their study eight main peaks were found from 20˚-60˚ and 2-theta degree almost similar to this research findings.

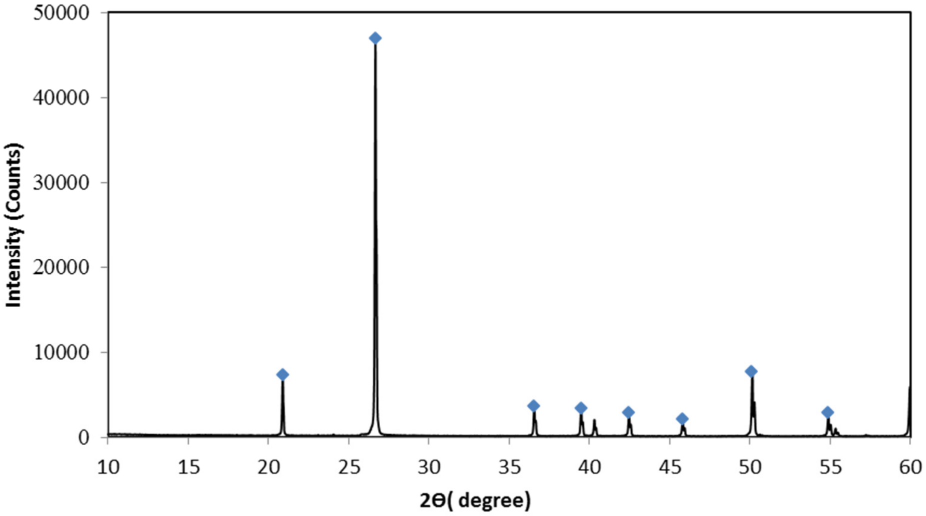

Fig. 1. XRD pattern for CS

Table 1
Peak of XRD for Silica

Sample	Silica Phase	Intense peak 2Θ
CS	Crystalline, Quartz	21°,26°,36°,39°,43°,46°,50°,55°

3.2 Elemental Analysis

Elemental analysis conducted *via* XRF method was performed to define the element composition of CS. Nine elements were identified. Silica (SiO_2) purity was detected at 99.60% while total impurity detected were 0.4% which are CaO, Na_2O, K_2O, MnO, Al_2O_3, Fe_2O_3, TiO_2, and Cr_2O_3, the compositional percentage content is as per Table 2.

Table 2
CS element concentration

Element	Concentration (%)
SiO_2	99.60
CaO	0.1533
Na_2O	0.1148
K_2O	0.0736
MnO	0.0233
Al_2O_3	0.0181
Fe_2O_3	0.0093
TiO_2	0.0040
Cr_2O_3	0.0037

3.3 Molecular Bonding Analysis

Molecular analysis of CS was analyzed *via* FTiR method to determine the molecular bonding in Unahan-Chem CS. The FTiR spectrum of CS is as shown in Figure 2. The characteristic of FTiR spectra molecular band were studied and three main vibration bands were identified. Si-O-Si asymmetrical stretching vibration of CS was detected at 1056 cm^{-1} and at wavelength of 796 cm^{-1} the vibration band of Si-O-Si symmetrical stretching vibration was determined. Vibration band of Si-O symmetric stretching at 777 cm^{-1} detected as Si-O. FTiR spectrum of CS at 1056 cm^{-1}, 796 cm^{-1} and 777 cm^{-1} is indeed relatively similar to pattern of CS [8, 9]. The comparison are as summarized in in Table 3.

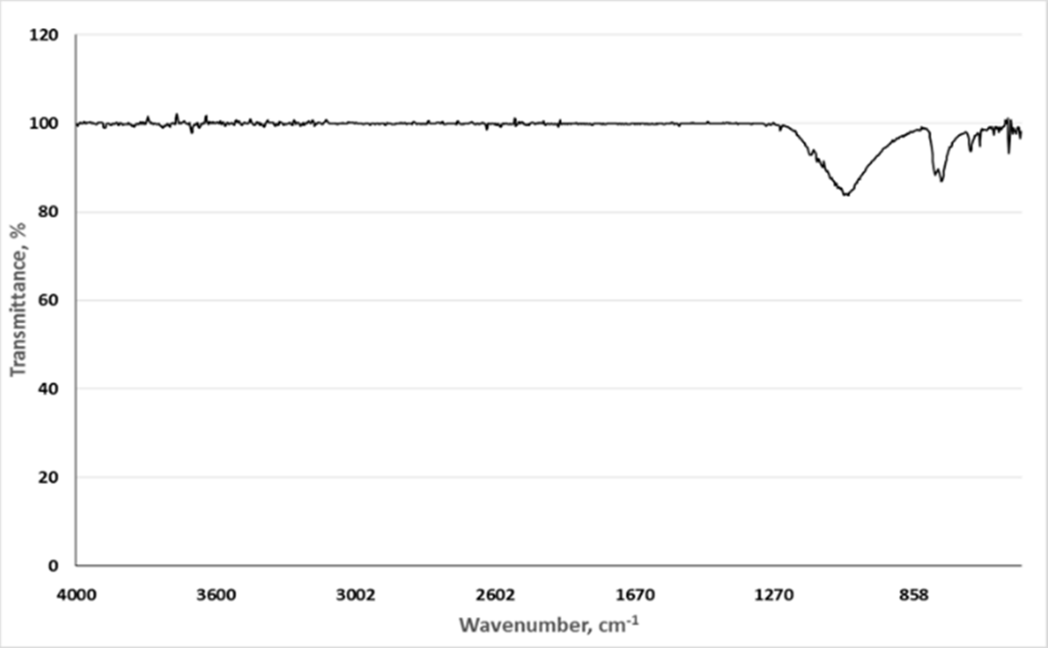

Fig. 2. FTiR of Unahan-Chem CS

Table 3
Band assignments of CS

Wave number (cm^{-1})	Band Assignments	Literature Value (cm^{-1})	Ref.
1056	Si-O-Si asymmetrical stretching vibration	1098	[8]
796	Si-O-Si symmetrical stretching vibration	780-799 779-798	[8] [9]
777	Si-O Symmetric stretching	779-798	[9]

3.4 Thermal Behaviour

To investigate fillers effect on the thermal stability of the PoS/Cs composites, their thermal degradative behaviors were evaluated by TGA. Thermal behaviour were identified by analysis on thermal decomposition temperature and relative mass loss. In general, CS as filler addition and curing at room temperature leads to increment of thermal stability. Figure 3 shows the relation constructed graph between relative mass and temperature of unfilled PoS and filled PoS with 2wt% to 12 wt% of CS. The depicted profile represents the thermal stability behaviour of PoS/CS composite. It was clearly seen that only a single step of degradation was observed between 300 ˚C - 450 ˚C.

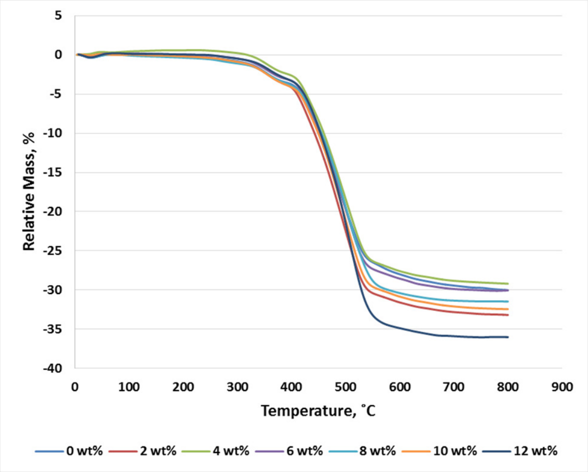

Fig. 3. TGA of CS-polysiloxane composites fabricated via casting method (Cured at RT)

The thermal decomposition temperature behaviour is as showed in Figure 4. Decomposition temperature of PoS/CS composites was found to increase with addition of CS as a filler. The highest decomposition temperature was observed at 12 wt% filler loading. The PoS/CS composites at 12 wt% degraded at 433˚C, 430.3˚C and 422.3˚C for PoS/CS/RT, PoS/CS/65˚C and PoS/CS/100˚C respectively. The higher percent of filler loading, promotes the composites resistant to degradation. The percentage of thermal stability enhancement towards decomposition temperature of PoS/CS composites at 12 wt% compare to unfilled Pos are 3.7%, 3.9% and 3.9% for PoS/CS/RT, PoS/CS/65˚C and PoS/CS/100˚C respectively.

Relative mass of PoS/CS composites measured the percentage of mass loss to air. It is measured as the material reach at thermal decomposition temperature. Relative mass percentage behaviour would signifies the ability of material to resist mass loss during high temperature exposure. Less relative mass percentage indicates the material is more stable at high temperature. The PoS/CS composites at 12 wt%, relative mass loss percentage were 0.2%, 0.4% and 0.68% of the initial mass for PoS/CS/RT, PoS/CS/65˚C and PoS/CS/100˚C respectively. As shown in Figure 5, the relative mass percentage showed that the higher filler loading used will decrease relative mass percentage. The relative mass loss percentage improvement for PoS/CS/RT, PoS/CS/65˚C and PoS/CS/100˚C at 12 wt% filler loading compared to unfilled PoS are 50%, 65.5% and 76.7% respectively.

Comparison of filled and unfilled PoS proved that CS as the filler helps to stabilize PoS/CS composites by increased decomposition temperature and relative mass [10]. The improvements were contributed by using CS as filler which offer thermal stability of having melting point at 1700˚C to 1800˚C. During the temperature exposure, matrix-filler bonding plays main role in order to hold their panel to avoid material loss due to the decomposition temperature. The composites panel also react by transferring the thermal exposed to the filler [11]. Thermal stability offered by CS had improve the matrix filler-bonding to become more stable compare to unfilled PoS. The use of high thermal stability filler had indeed improved the decomposition temperature and relative mass percentage of composites [12].

The amount of hydroxyl group in the composites panel affect the thermal decomposition. The hydroxyl group referred to the hydroxyl bonding in PoS. Increasing the amount of filler would cause the thermal decomposition to significantly increase, due to the decrement of hydroxyl group in the composites panel which was replaced by the filler used, moreover with the excellent thermal stability of CS, it would then enhance the thermal stability of the composites [13].

Both thermal stability behaviours showed that the curing temperature of PoS/CS composites affect the thermal properties. Figure 4 and Figure 5 show that, PoS/CS composites cured at RT had highest thermal decomposition temperatures and lowest relative mass loss percentage. While PoS/CS/65˚C and PoS/CS/100˚C shows lower thermal stability characteristic. Thus, curing of PoS/CS composites at high temperature does not improve the thermal stability of the PoS/CS composites. The high curing temperature helps to shorten curing time and affect thickness of the composites, since the high temperature curing causes the shrinkage of the composites once cured [14]. The high temperature cured PoS/CS composites become less stable compared to RT PoS/CS composites due to disruption of PoS hydroxyl cross link properties which was affected by accelerated curing through high curing temperature[15].

Also Figure 4 and Figure 5 show that PoS/CS/RT constructed profile are more stable and linear compared to PoS/CS/65˚C and PoS/CS/100˚C. The non-linear profiles were caused by filler aggregations and uneven dispersion which was affected by accelerated curing [16]. As the amount of CS increased, the tendency of aggregations and uneven dispersion also increase, which explain the non-linear profiles with the increment of filler loading amount. It is clearly seen occurred in between filler loading at 4wt% to 10 wt% for PoS/CS/65˚C and PoS/CS/100˚C [13]

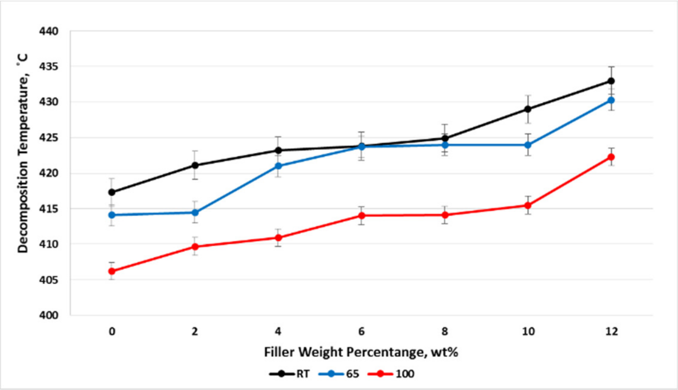

Fig. 4. Thermal decomposition of PoS/CS composites

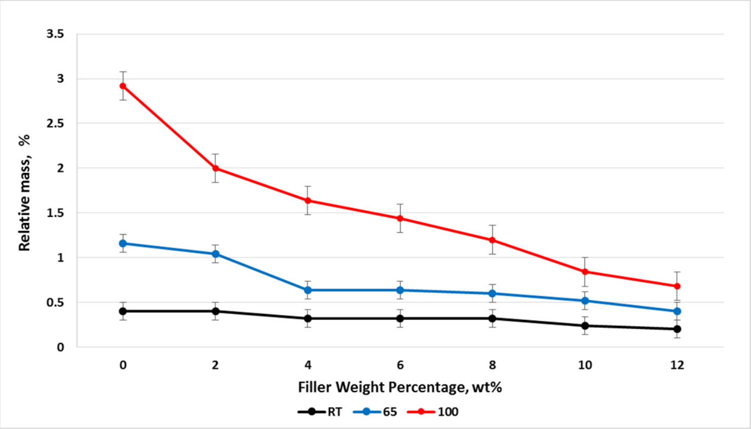

Fig. 5. Relative mass percentage of PoS/CS composites

4. Conclusions

Through observation in TGA it was found that CS had improved the thermal stability of polysiloxane. The increment of amount of filler loading, had improved significantly the decomposition temperature and relative mass loss percentage. Moreover, it was also concluded that curing temperature of PoS/CS/RT offer better thermal stability through well linked crosslinking of hydroxyl group which lead to good dispersion of filler.

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