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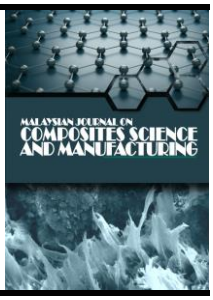

Preparation and Properties of Nanocomposite Based on K-153 Epoxy Reinforced T-13 Glass Fiber

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ABSTRACT

This study aims to investigate the influence of nanosilica content and the ratio of glass fiber/epoxy resin on the mechanical properties of nanocomposites based on K-153 epoxy resin (K-153) matrix reinforced by T-13 glass fiber (T-13). Thermogravimetric Analysis (TGA), Field Emission Scanning Electron Microscopy (FESEM) and Energy Dispersive X-Ray Analysis (EDX) were used to examine the results of the investigations. The findings showed that the nanosilica of 1.20 weight percent (wt.%) was suitable for making K-153/T-13/nanosilica nanocomposites. Compared to K-153/T-13 polymer composites, K-153/T-13/nanosilica nanocomposites with nanosilica content of 0- 1.2 wt.% and the ratio of T-13 glass fiber/K-153 epoxy resin at 70/30 wt.%, the tensile strength had increased from 215.41 MPa to 313.12 MPa and from 230.96 MPa to 325.35 MPa for flexural strength. Besides that, the thermal resistance of nanocomposites also increases compared to that of polymer composites based on K-153 epoxy resin. At 500 °C, the weight loss of nanocomposites and polymer composites was 34.01 % and 37.67 %, respectively.

1. Introduction

Polymer composite based on epoxy resin, reinforced by glass fiber is commonly used in transportation, electronics, mechanics, machines, buildings and chemicals [1-5]. However, disadvantages that limit this polymer composite's applicability are brittle and poor in their impact resistance [6-9]. Therefore, many studies on solutions to this problem have been carried out, mainly focusing on two methods, namely (i) using plasticizers for the epoxy-based phase or; (ii) choosing an appropriate curing agent [10-13]. Besides that, carbon nanotubes, nanoclays, nano-graphene, nanosilica, etc., have been added to polymer composites based on epoxy/glass fiber to improve their

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resulted flexural strength, tensile strength, electrical properties, thermal performance of polymer composites [11, 12, 14].

K-153 epoxy resin (K-153) is one kind of epoxy resin produced by modifying ED-20 epoxy resin simultaneously with thiokol and acrylate oligomer [15]. It is generally more flexible and less viscous than other types of epoxy resins. K-153 epoxy resin is a good matrix for polymer composite, but reports on this kind of polymer composite are quite limited. In the previous studies, authors had investigated the effect of drying temperature on the degree of curing, structural morphology, etc., of polymer composites based on K-153 with glass fiber [15-16]. Besides that, in other research [17-18], authors had studied the influence of nanosilica content on some properties of nanocomposite, based on K-153 epoxy resin, without the glass fiber addition. For example, tensile strength and flexural strength of nanocomposite with 1.2 wt.% of nanosilica were 79.81 MPa and 98,14 MPa, which were much higher than those of K-153 as 53.30 MPa and 60.81 MPa, respectively. Meanwhile, the thermal oxidation resistance and ash remaining of polymer composites based on K-153 epoxy resin also increased compared to those of K-153 epoxy resin.

In this study, the influence of nanosilica on the tensile strength and flexural strength of nanocomposite based on K-153 matrix, reinforced by T-13 glass fiber was evaluated and discussed further. Furthermore, the effect of glass fiber/epoxy resin ratio on nanocomposite tensile and flexural strength was also studied. In addition, the comparison in thermal resistance of produced nanocomposite and polymer composite based on K-153 epoxy resin and T-13 glass fiber was also investigated.

2. Materials and Methods

2.1 Chemicals

K-153 epoxy resin of TU 6-05-1584-85 (Russia), epoxy content: 19-22 %, molecular weight: 390 g/mol, mass fraction of volatile substances < 0.9%, and dynamic viscosity at 20 °C, Pas: 6-12. Polyetylenpolyamine (PEPA) of TU 2413-357-00203447-99 supplied by Chimex Ltd (Russia), molecular weight: 230- 250 g/mol, and third amine group: 5- 9. T-13 glass fiber roving of GOST 19170-2001 (Russia) with a mass density of 300 g/m^2. Nanosilica (CAS number: 7631-86-9) supplied by Sigma-Aldrich, fine powder, purity: 99.8%, average size: 12nm, specific surface area: 175- 225 m^2/g (based on BET method).

2.2 Paint Preparation

T-13 glass fiber roving was dried at 90^{o}C for 6 hours and then cut into 150 x 200 mm sheets. First, k-153 and nanosilica of 0 - 1.50 wt.% was mixed well until getting homogenous mixture. Then, a curing agent, PEPA, was added to the mixture with a ratio of K - 153 (100 wt.%) and PEPA (10 wt.%), respectively [16, 17].

Cleaning and spraying the non-stick coating prepared a glass sheet for sample making. First, a layer of K-153 was mixed with a curing agent and applied on the glass sheet, then a sheet of T-13 glass fiber roving was put on, and a roller was used to wet the epoxy resin onto T-13. These steps were repeated until the 4th layer of T-13 achieved the thickness of the nanocomposite sample of about 5 mm. The glass fiber/epoxy resin ratio varied from 0/100 wt.% to 80/20 wt.%. Nanocomposite samples were dried at 80 °C for 6 hours [16, 17] and kept for 7 days at room temperature before conducting the testing to examine the properties.

2.3. Analysis methods

The tensile strength was determined per ISO 527-1:2012 by using the Zwick device with a sample pulling speed of 5 mm/mins, at 25 °C, and humidity of 70 %. Flexural strength was determined in compliance with ISO 178:2010 by using an Instron 5582-100 kN machine at a bending speed of 5 mm/min.

The elemental composition of produced samples was determined on Energy Dispersive X-ray Spectrometry (EDX) analyzer model HORIBA 7593H. The samples were prepared as a plate with an accelerating voltage exposure of 20 kV.

For the thermal resistance, the thermogravimetric analysis (TGA) was conducted using NETZSCH TG 209F1 LIBRA in nitrogen gas with a temperature raising rate of 10 °C/min from room temperature to 500 °C. The morphological observation was conducted using the FESEM Hitachi S4800 machine with a magnification of 500 times and an accelerating voltage of 2KV.

3. Results and Discussion

3.1. Effect of nanosilica loadings on the mechanical properties of nanocomposites

Nanocomposite samples based on T-13/K-153/nanosilica were made with a T-13/K-153 ratio of 70/30 wt.%, and nanosilica content varied in the range of 0 - 1.50 wt.%. After making it, the samples were dried at 80 °C for 6 hours and kept for seven days at room temperature before determining the mechanical properties. The results are presented in Figure 1.

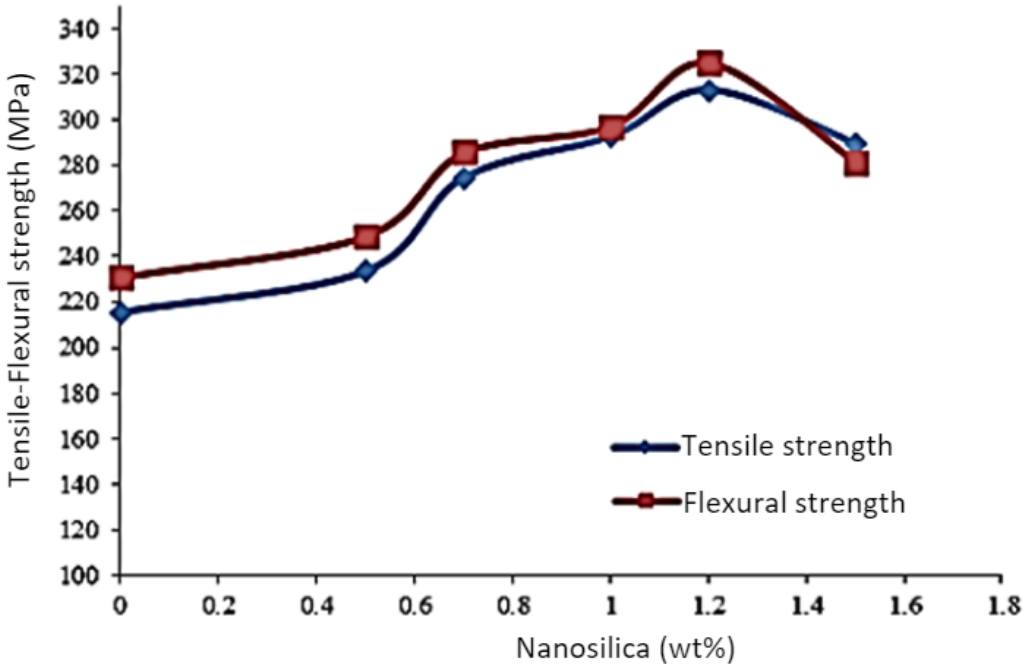


Fig.1. Effect of nanosilica content on the tensile strength and flexural strength of K-153/T-13/nanosilica nanocomposites (T-13/K-153,70/30 wt.%)

Figure 1 shows that with the content of nanosilica addition between 0 to 1.20 wt.%, the tensile strength and flexural strength of produced nanocomposite had increased significantly, from 215.41 MPa up to 313.12 MPa for the tensile strength. The flexural strength was increased from 230.96 MPa up to 325.35 MPa. However, once the nanosilica loadings reached 1.50 wt.%, the nanocomposite's tensile and flexural strength significantly decreased. This situation implied that to have a good adhesion

interaction between all components (K-153, T-13, and nanosilica), the added nanosilica particles must be uniformly distributed within the epoxy resin matrix. The added nanosilica nanofiller worked as a bonding bridge, increasing the bonding ability of resin matrix and glass fiber and improving mechanical properties [18-20]. When the nanosilica content was over 1.20 wt.%, the nanocomposite's tensile and flexural strength decreased rapidly. It was due to the relatively dense nanosilica at this content, causing the viscosity to become too high. As a result, it became difficult to wet the glass fiber to form a homogeneous nanocomposite. This finding on the effect of nanosilica content on the tensile and flexural strength of nanocomposites is similar to those reported for K-153/nanosilica nanocomposites without glass fiber [17]. In addition, agglomerates would form larger particles at higher concentrations of nanosilica particles, reducing nanoparticles' dispersion ability within the epoxy matrix. Thus, the material's mechanical properties were reduced [10, 14, 17].

3.2. Effect of glass fiber/epoxy resin ratio on the tensile strength and flexural strength of nanocomposites

The samples were prepared with different ratio of T-13/K-153 and nanosilica content of 1.20 wt.% to study the effect of T-153/K-153 ratio on the tensile strength and flexural strength of nanocomposites. After preparation, the samples were dried at 80^{o}C for 6 hours and kept conditioned for seven days at room temperature before determining material properties. The experimental results are presented in Table 1.

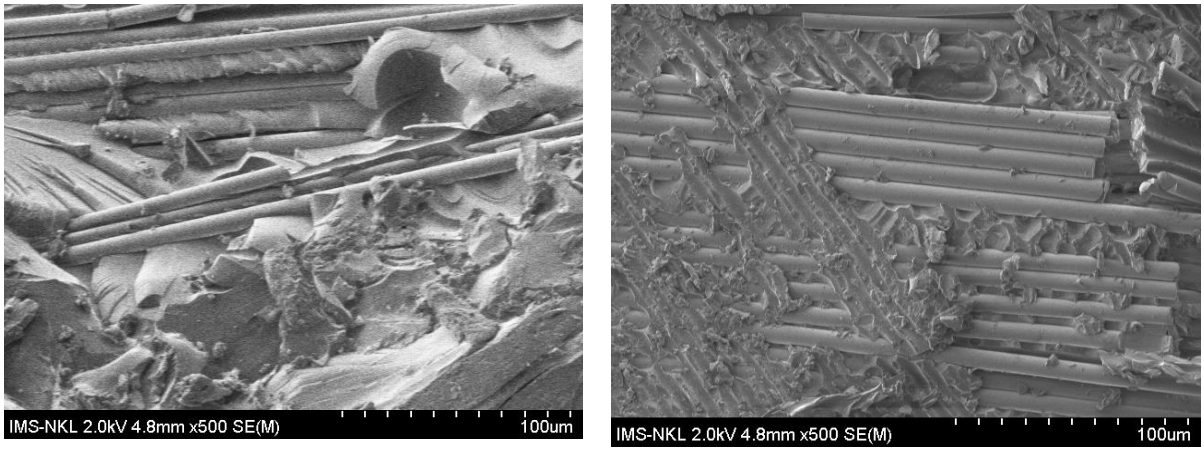
Table 1 shows that the increased glass fibre content increases nanocomposites' tensile strength and flexural strength. Then, it reached the highest maximum values when the glass fiber content used was 70 wt.%. Corresponding to tensile strength increased from 79.81 MPa to 313.12 MPa, the flexural strength increased from 98.14 MPa to 325.35 MPa. Further increased the glass fiber proportion, it was found that the tensile and flexural strength of nanocomposite rapidly decreased. This can be explained by the fact that the glass fiber had higher strength and stiffness, so increasing the ratio of glass fiber would improve the tensile strength and flexural strength of produced nanocomposite. However, nanocomposites' mechanical properties highly depended on interaction and adhesion between the glass fiber and matrix. The bonding dictates the stress transfer efficiency at the interphase [14-16]. The epoxy resin could not evenly cover the glass fiber surfaces at a more extensive glass fiber content of > 70 wt.%. The adhesion ability between the glass fiber and matrix was insufficient at this point. This situation led to uneven stress distribution in the nanocomposite material. Then, it reduces the nanocomposite tensile and flexural strength [17, 21, 22].

Table 1
Effect of glass fiber/epoxy resin to the tensile strength and flexural strength of K-153/T-13/nanosilica nanocomposites

No.	T-13/ K-153 (wt. %)	Tensile strength (MPa)	Flexural strength (MPa)
1	0/100	79.81	98.14
2	40/60	170.68	186.55
3	50/50	246.74	264.86
4	60/40	289.56	293.48
5	70/30	313.12	325.35
6	80/20	275.78	307.68

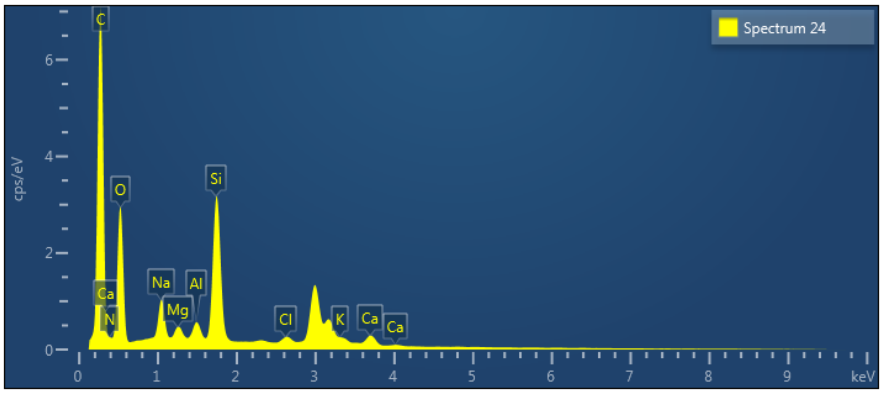
3.3. SEM observation and EDX analysis of produced polymer composites and nanocomposites

The SEM-EDX observation was conducted to study the influence of nanosilica particles on the morphology structure and chemical composition of K-153/T-13 polymer composite and K-153/T-13/nanosilica nanocomposite. The SEM images and EDX of the fractured surface of glass fiber/epoxy resin ratio of 70/30 polymer composite and nanocomposite samples with glass fiber/epoxy resin/nanosilica ratio of 70/30/1.20 are depicted in Figures 2 and 3. The analyses are detailed in Tables 2 and 3.

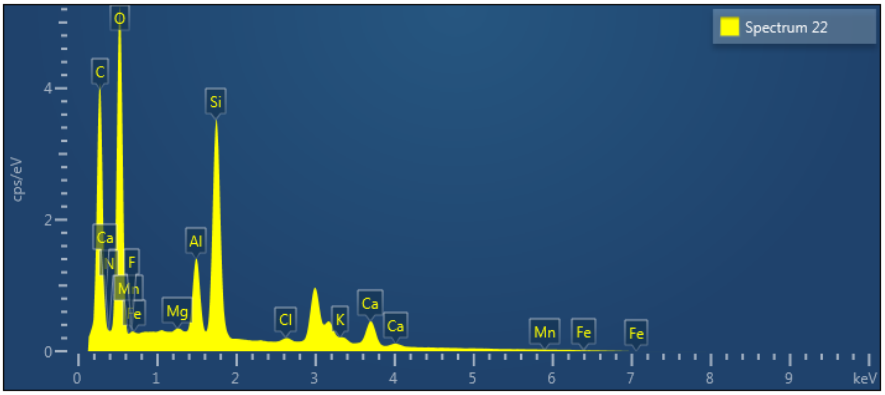


(a) (b)

Fig. 2. SEM images of (a) K-153/T-13 polymer composite and (b) K-153/T-13/nanosilica nanocomposite



(a)



(b)

Fig. 3. EDX of (a) K-153/T-13 polymer composite and (b) K-153/T-13/nanosilica nanocomposite

Table 2
Chemical compositions of K-153/T-13 polymer composite

Spectrum 24				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	51.81	0.61	62.60
O	K series	27.37	0.40	24.82
Na	K series	2.44	0.07	1.54
Si	K series	9.82	0.15	5.07
Mg	K series	0.70	0.04	0.42
Al	L series	1.05	0.05	0.56
N	K series	3.68	0.86	3.81
Ca	K series	2.41	0.11	0.87
Cl	K series	0.52	0.06	0.21
K	K series	0.21	0.10	0.08
Total		100.00		100.00

It is clearly shown in Figure 2(b) that the clustering nanosilica particles have appeared on the glass fiber. A similar feature cannot be seen in Figure 2(a). These observations were also consistent with EDX mapping results, as presented in Figure 3. Several elements, such as manganese (Mn) or iron (Fe) could be observed in Figure 3(b) but not in Figure 3(a). In comparison, Table 2 lists the percentages of Si and O, which are 9.82 % and 27.37 %, respectively. Meanwhile, in Table 3, those percentages are 11.28 and 41.35, respectively. These results explained the presence of nanosilica particles in the nanocomposites. These results also confirmed that the presence of nanosilica particles had significantly improved the mechanical properties of produced nanocomposite [19, 21, 23].

Table 3.
Chemical compositions of K-153/T-13/nanosilica nanocomposite

Spectrum 22				
Element	Line Type	Weight %	Weight % Sigma	Atomic %
C	K series	31.91	0.80	42.61
O	K series	41.35	0.89	41.44
Al	K series	3.65	0.10	2.17
Si	K series	11.28	0.25	6.44
N	K series	3.44	0.70	3.94
Cl	L series	0.20	0.05	0.09
Ca	K series	4.42	0.14	1.77
K	K series	0.27	0.08	0.11
Mg	K series	0.18	0.04	0.12
F	K series	0.63	0.16	0.53
Mn	K series	2.18	1.60	0.64
Fe	K series	0.48	0.81	0.14
Total		100.00		100.00

3.4. Thermal resistance of polymer composite and nanocomposite

To study the thermal resistance of polymer composite and nanocomposite, Thermogravimetric analysis (TGA) was used. Sample of glass fiber/resin/nanosilica nanocomposite with a ratio of 70/30/1.20 wt.% and polymer composite with epoxy/glass fiber ratio of 70/30 wt.% were compared. The TGA analysis was conducted in the nitrogen atmosphere with a heating rate of 10°C/min from room temperature to 500°C. The TGA results for comparisons are shown in Table 4 and Figure 4.

Table 4.
Effect of nanosilica on the thermal oxidation resistance of the coating

Samples	Weight loss (%)	
	350 °C	500 °C
K-153/T-13 polymer composite	7.51	37.67
K-153/T-13/nanosilica nanocomposite	12.15	34.01

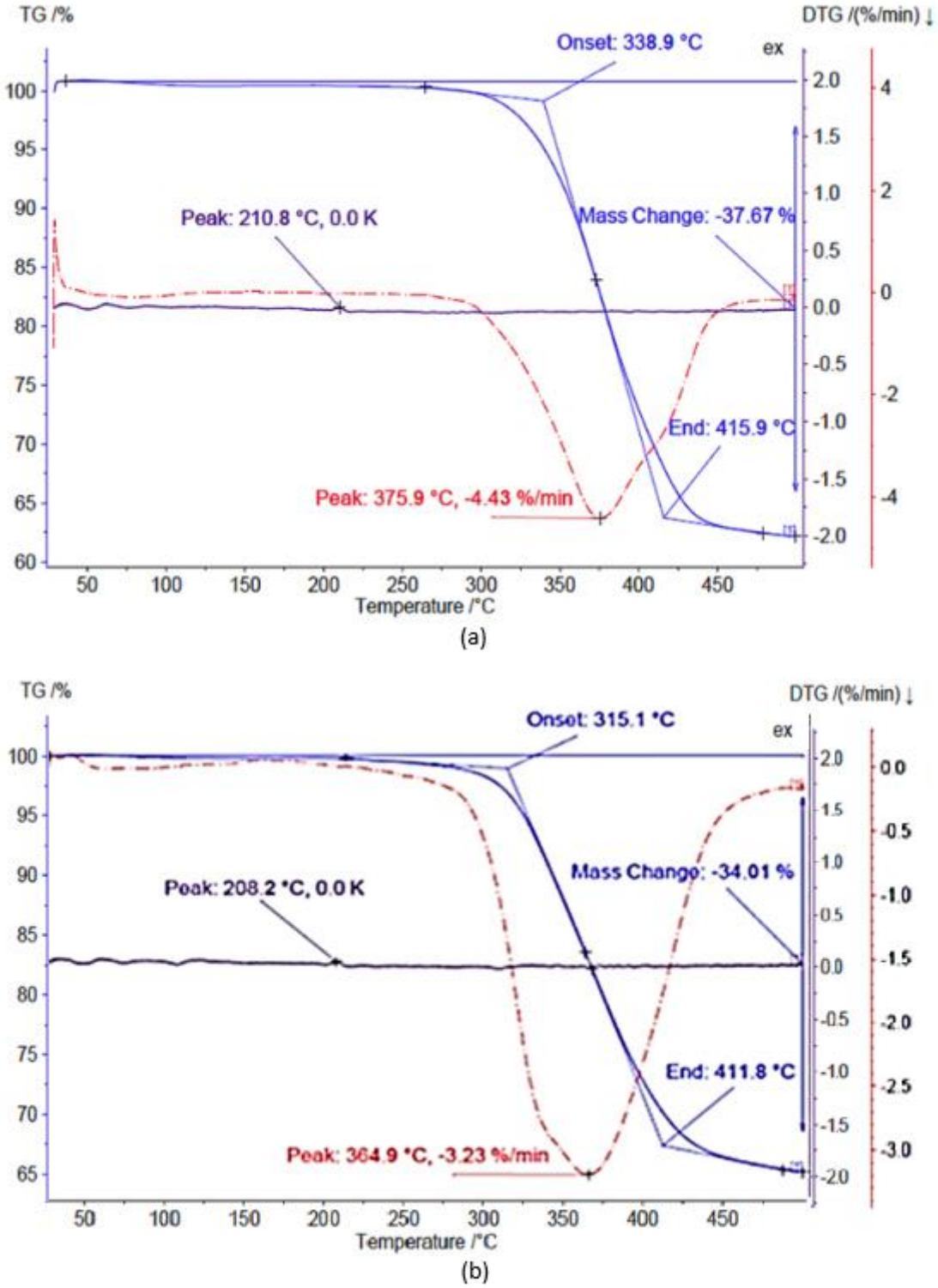


Fig. 4. TGA of (a) K-153/T-13 polymer composite and (b) K-153/T-13/nanosilica nanocomposite

The results presented in Figure 4 and Table 4 showed that the thermal resistance of nanocomposite had increased significantly if compared to polymer composite. Besides, the polymer composite has an ash content of 62.33 %. Meanwhile, with nanosilica, the ash content of nanocomposite is much higher, up to 65.99 %. Thus, it implies that the added nanosilica improved the thermal resistance of produced nanocomposite [19-21]. This observation can be explained by the fact that the added nanosilica particles efficiently worked as a "fence" to protect the material structure from heat and as an agent to prevent polymer chain decomposition, resulting in higher thermal resistance. In addition, under the thermal decomposition of nanosilica, coke would form a stable structure like ceramic. The role of nanosilica in improving the thermal resistance of nanocomposite has been mentioned elsewhere [18, 25-28].

4. Conclusions

Nanosilica content strongly affects the tensile and flexural strength of produced nanocomposites based on K-153 epoxy/T-13 glass fiber/nanosilica. Nanosilica content of 0- 1.20 wt.% had caused the tensile strength to increase from 215.41 MPa up to 313.12 MPa. Besides, the flexural strength was increased from 230.96 MPa to 325.35 MPa. Nanosilica of 1.20 wt.% is the most suitable for polymer nanocomposite with an ideal ratio of T-13 glass fiber/K-153 epoxy resin at 70/30 wt.%. The thermal resistance of glass fiber/epoxy resin/nanosilica nanocomposite is higher than that of glass fiber/epoxy resin polymer composite. At 500 ^{o}C, the weight loss of nanocomposite and polymer composite was 34.01% and 37.67%, respectively.

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