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Research paper



The Use of Multi-Walled Carbon Nanotubes and Nanoclay for Simultaneously Improving the Flame Retardancy and Mechanical Properties of Epoxy Nanocomposites

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Abstract

In this study, the flame retardants epoxy nanocomposites were prepared by combining mechanical stir and sonication of epoxy Epikote 240 (EE240) resin, chlorinated paraffin, atimony oxide, multi-walled carbon nanotubes (MWCNTs) and montmotillonite clay. Resultants of CNTs, montmorillonite and flame retardant additives were investigated limiting oxygen (LOI) and UL-94, combustion rate. The SEM, FE-SEM, TEM were measured to analyze the dispersion of MWCNTs and montmorillonite clay in epoxy matrix. The mechanical properties including tensile strength, compressive strength, flexural strength and impact strength Izod were studied. The results of testing burning and mechanical properties indicated that CNTs were more efficient than clay in improving the flame retardancy of materials. The dispersion method combining of mechanical stir and sonication is the good choice to distribute additive agents into epoxy matrix.

Keywords: antimony trioxide, chlorinated paraffin, flame retardancy, epoxy resin, multi-walls carbon nanotubes, Epoxidised Linseed Oil.

1. Introduction

Epoxy resins are well-known thermosetting polymers and have been used widely in industrial applications for a long time. They exhibited many attractive characteristics such as high tensile strength and modulus, low shrinkage during cure, chemical and corrosion resistance, dimensional stability, high adhesion to many substrates and superior electrical properties. They were usually applied for surface coating, adhesive, composites, laminate and matrix materials in electronic, transport and aerospace areas [1, 2]. However, one of the main drawback of epoxy resins are their inherent flammability, restricting their applications in many fields because of safety consideration. Therefore, improving the flame retardancy of epoxy resins is an important issue needed to be concerned. This problem can be deal either by incorporating fire-retardant additives (the additive type) [3-6] or by copolymerization with reactive flame retardants (the reactive type) [7-9]. Among two strategies, the addition of fire-retardant additives is more simple and common one to achieve flame retardancy in polymers because of its low-cost and convenience.



There are some additives were used in literature such as halogen – or phosphorus – containing flame retardants [10-13]. In our previous report [14], by introducing of 9 phr of antimony trioxide and of 11 phr of chlorinated paraffin to epoxy, obtained materials had not only high flammability but also maintained mechanical strength.

Recently, carbon nanotubes (CNTs) are considered as ideal agents to disperse into polymer nanocomposites because they are expected to enhance the mechanical, thermal and electrical properties of epoxy polymers [15-17]. To achieve the improvement of properties of epoxy polymers with adding CNTs, single-walled (SWCNTs) or multi-walled carbon nanotubes (MWCNTs), the most important things are the uniform dispersion and an effectively strong interaction between CNTs and the polymer matrix [18-19].

In this study, the dispersion effects of co-additives including antimony trioxide/chlorinated paraffin and MWCNTs were investigated for flame retardant and mechanical properties of EE240 resin. By using ultrasound at 65⁰ C with different times, MWCNTs or co-additives were dispersed into the epoxy resin with various weight percentages to find the optimized weight ratio for achieving uniform dispersion and good interaction of components. The final products were then investigated by scanning electron microscope (SEM), limiting oxygen index (LOI) and UL 94.

2. Experimental

2.1 Materials.

The epoxy Epikote 240 (EE240) was provided by Dow chemicals (USA). EE240 is a low viscosity, based on a belnd of bisphenol A resin and bis phenol B resin, contented epoxy group of 24.6 %, molecular weight (Mw) of 5100-5400 mmol/kg, density of 1.12 g/ml, viscosity at 25°C were to be 0.7-1.1 Pa.s. Diethylenetriamine (DETA) received from Dow Chemical (USA) has a density of 0.95 g/ml boiling point of 207°C, Mw of 103 mmol/kg and used directly without any further purification. MWCNTs with a diameter of 40-45 nm and a length of around 3 µm was provided by Showa Denko Japan Co. Chlorinated paraffin - S52 (CPs) was purchased from China and contained maximum of 52 % of chlorine. Antimony oxide (ATO) was received from China as a fine white powder, melted at 656°C, had a specific gravity of 5.7. Antimony trioxide has the content of antimony trioxide is over than 99.0%, amount of antimony metal is approximately to be 83 %. The average size of fine powder antimony trioxide was to be 1.5 µm. Epoxidised Linseed Oil (ELO) was obtained as

Lankroflex E2447 from Acros Chemical Limited (UK). It was liquid solidifier at low temperature, light (or pale) yellow-water-white color, in soluble in water. Its viscosity at 25°C was to be 800 cSt and contained 22.89 % of epoxy group in content. Nanomer® I.30E nanoclay (Nanocor USA) is a surface modified montmorillonite mineral which will disperse to nanoscale in epoxy resin systems. The dispersion creates a near-molecular blend commonly known as a nanocomposite. This new type of composite exhibits enhanced strength, thermal and barrier properties. I.30E is supplied as a white powder which disperses to particles so thin they are nearly transparent in the resin matrix. As reinforcement in composites, e-glass woven fabric with a density of 600 g/cm² and a single fiber diameter of 14–16 lm was procured from Fiber Glast Development Corporation.

2.2 Preparation of Samples.

2.2.1. Preparation of EE240 Contained Chlorinated Paraffin/Antimony Trioxide.

The powders of chlorinated paraffin and antimony trioxide were dried at 80°C in 60 min before mixing with the ratio of 11 phr and 9 phr, respectively, to EE240 and kept for 24 hours. After that, mechanical stirring was used with the rate of 1500 rpm (HS-100T, WiseStir, Korea) at 80°C for 3 hours in water bath. The mixture was then cooled to RT, added DETA and stirred with the rate of 150 rpm for 10 min. After the sample was removed bubbles in vacuum for 30 min, the bubble free mixture was casted on a mould and kept for 24 hours, dried at 80°C for 3 hours, and stabilized for 7 days before investigating the mechanical properties and flame retardancy.

2.2.2. Preparation of EE240 Contained Various Weight Ratios of MWCNTs.

MWCNTs with various weight ratios (0.01, 0.02, and 0.03 wt %) were mixed with EE240. The mixtures were then sonicated using an ultrasonic bath (Elmasonic S300 H, 37 kHz, Germany) for 6 hours at 65°C. After sonication, the hardener was added to the mixtures and stirred for 15 min at 200 rpm/min. Air bubbles were removed by placing the mixture under vacuum. The bubble free mixtures were then casted on a mould and cured at 80°C for 3 hours.

2.3.3. Preparation of EE240 Contained Nanoclay.

Nanoclay-I.30E was dried at 80°C for 1 hour before mixed with EE240. The quantity of nanoclay-I.30E was varied with the value

of 1 %, 2 %, 3 %, and 4 %, dispersed in EE240 at 80°C for 8 hours by mechanical stirring at 3000 rpm. After that, the mixture was sonicated (Ultrsonic homogenizer, Model 300V/T, Bilogics INC, USA) for 1 hour with the power of 50 % at room temperature. The mixture was then removed air bubbles by vacuum for 15 min, added hardener DETA, stirred mechanically with the speed of 60-80 rpm for 10 min. Then the mixture was poured into mold. After 24 hours, the sample was dried for 3 hours at 80°C (\pm 20°C), and tested the mechanical properties and flame retardancy after 7 days.

2.3.4. Preparation of E-glass/MWCNTs/Chlorinated Paraffin/Antimon Oxit/ EE240 Nanocomposites.

To produce samples for burning-off, after dispersion of MWCNTs into the epoxy resin, the conventional hand lay-up method was employed to fabricate different sheets of MWCNTs/chlorinatedparaffin/antimonoxit/EE240

nanocomposites. E-glass/MWCNTs/chlorinated paraffin/antimon oxit/EE240 nanocomposites were fabricated by using a combination of hand lay-up and compression hot press techniques. E-glass woven fabric layers were properly stacked into 10 plies and the orientation of fiber within the fabric was kept constant. The final thickness of the panel was measured to be 4 mm.

2.3.5. Preparation of E-Glass/Epoxy Nanocomposites.

E-glass/epoxy nanocomposites were fabricated by using a combination of hand lay-up and compression hot press techniques. E-glass woven fabric layers were properly stacked into 7 plies and the orientation of fiber within the fabric was kept constant. Epoxy resin modified with MWCNTs was spread uniformly on each fabric layer and the laminate was consolidated by applying 133 kN force in a hot press. The temperature of hot press was maintained at 60°C for 4 h for curing followed by post curing in oven for 4 h at 100°C. The final thickness of the panel was measured to be 4 mm.

2.3 Characterizations.

The morphology of the samples was carried out by scanning electron microscope (SEM, Evacseq error codes, S-4800, Japan). Structural characterizations were studied by X-ray diffraction (XRD, D8-Advance, Brucker, Germany). The transmission electron microscopy was performed on a Titan Cryotwin (FEI Company) equipped with a 4 k \times 4 k CCD camera (Gatan) at an acceleration voltage of 300 kV. An ultramicrotome (Leica microsystem) was used to prepare cut ultra-thin sections (80 nm) of samples before recovered on a copper grid. The flame retardant

properties was tested by the limiting oxygen index (LOI) (Yasuda Seiki Seisakusho Ltd, Japan) according to JIS K720-1976, with sheet dimensions of $120 \times 6.5 \times 3.2$ mm. The UL-94 rating was tested according to the UL-94 (ASTMD635-12) with sheet dimensions of 125 ± 5 mm long by 13.0 ± 0.5 mm wide, and provided in the minimum thickness and 3.0 (-0.0 + 0.2) mm thick. The combustion rate was measured by COMBUSTION RESISTANCE COD 6145000 according to ASTM D757-77 standard. Specimen's dimensions $3.17 \times 12.7 \times 121$ mm³. Mechanical properties were measured on an INSTRON-5582 100 KN (USA) according to ISO 527-1993 at an extension speed of 5 mm/min. All data were the average of five independent measurements; the relative errors committed on each data were reported as well.

3. Results and Discussion

3.1 Study the Mechanical Properties and Flame Retardancy of EE240/MWCNTs Nanocomposite.

3.1.1 Morphology of the EE240/MWCNTs Nanocomposite

Figure 1 shows the dispersion of MWCNTs was affected by ultrasonic time. When the ultrasonic time was 4 hours, MWCNTs was poorly dispersed. It can be seen that they gathered up small clusters and only a little tube of MWCNTs spread into the epoxy. Increase ultrasonic time to 5 hours, MWCNTs dispersed more evenly, the clusters became smaller compared to those of 4 hours. When ultrasonic time was increased to 6 hours, MWCNTs was dispersed properly and homogeneously covered by epoxy, indicating the good interaction of the background and additive. We think in this condition, MWCNTs have diffused into the gap made by cross-linkages of epoxy molecules and amino groups of curing agent which was shown in Figure 2. With this mechanism, the nature of plastic materials, especially the mechanical properties and fire resistance, will be definitely improved. Increase continuously the time of ultrasound solution to 7 hours, the dispersion of MWCNTs were not better than that of 6 hours. Therefore, 6 hours - ultrasonic time at 65°C was the optimized condition for dispersion of MWCNTs into EE240.

Please See Figure 1.

The effect of quantity of MWCNTs dispersed in EE240 was investigated with various weight ratios of 0.01%, 0.02% and 0.03% by using optimized condition above. High solution FE-SEM images of the fracture surface of obtained materials were shown in Figure 2. It can be observed that a uniform distribution was exhibited in the case of material containing 0.02% wt of

MWCNTs. With the case of material containing 0.03 % of additive, mass of MWCNTs was detected while the low density of them was seen in the case of material containing 0.01 %. Hence, the optimized quantity of MWCNTs for dispersion it into epoxy Epikote 240 was to be 0.02%.

Please See Figure 2.

Network structure of MWCNTs/EE240 plays an important role as a protective shield and reduces the proportion of coal combustion and polymer because MWCNTs in this network acts as a shield or barrier from fire. Combustion inhibition ability of material depends on the network structure of itself, consequently, the well distribution of MWCNTs in the polymer is considered as a mandatory requirement for improving flame retardancy. Application of optimized condition for dispersion of MWCNTs into EE240, the increasing of combustion inhibition ability of material can be predicted.

3.1.2. Mechanical Characteristics and Flame Retardant Properties of EE240/MWCNTs.

The mechanical characteristics and flame retardant properties of epoxy/MWCNTs depended on the processing time and the weight ratios of MWCNTs were shown in Table 1 and Table 2, respectively.

In Table 1.

In Table 2.

In the Table 1, the sample that sonicated in 6 hours shows high LOI of 23.2% with slow combustion of 21.70 mm/min. The slow combustion of this material can be explained by the formation of ash layer from MWCNTs prevented the contact with the polymer fire during the inflammation. Hence, the materials with combination of MWCNTs and ground polymer makes a protective layer during the burning of samples, plays as a barrier between materials, gas phase and flame. In addition, the presence of the carbon nanotube also increased the thermal conductivity of polymer. As the result, the inflammation time and heat release rate of obtained materials were increased with distribution ability of MWCNTs in EE240. It can be suggested that when MWCNTs were distributed regularly into ground polymer, the coal layers were formed quickly and covered over the surface. These layers, as barriers, minimized the inflammation ability and prevented the contacting of material with air, consequently decreased the re-burning trend and stopped the spreading of flame, led to turn it off.

Following the mechanism above, the quality of dispersion of MWCNTs in EE240 is a crucial factor effecting directly to the flame retardant ability of EE240/MWCNTs. Fig. 2 showed the

results of the fire retardant ability of obtained materials, appreciating the agreement with theory. As the prediction above, the material sonicated for 6h has the higher fire retardancy than that of 4, 5, 7 hours and EE240. The LOI was to be 23.2%, the combustion rate following UL 94HB was to be 21.70 mm/min.

The poorer flame retardancy of the samples used the ultrasonic vibrating time of 4, 5 and 7 hours compared to that of 6 hours can be explained by the distribution of MWCNTs into ground polymer. In cases of 4 and 5 hours, the ultrasonic vibrating time were not sufficient for evenly distributing of MWCNTs, lead to appear concentrated areas of MWCNTs into inside polymer and formed defects. However, when the ultrasonic vibrating time was increased to 7 hours, the fire retardant ability of material was not improved, slightly lower than that of sample treated for 6 hours.

3.2. Study the Effect of Nanoclay Quantity to Mechanical Properties and Flame Retardant Ability of Nanocomposite EE240/I30E.

3.2.1. X-ray Diffraction (XRD) and Transmission Electron Microscopy (TEM) Analyses of EE240/MMT-I30E Nanocomposites

Nanoclay I.30E (1, 2, 3, 4 % by weight) was dispersed in EE240 by mechanical stirring with the speed of 3000 rpm at 80^oC for 8 hours, then sonicated continuously for 60 min with the power of 50 % at $25-30^{\circ}$ C.

To investigate the dispersed ability of nanoclay in EE240, structure of materials were examined by TEM, FE-SEM and XRD (Figure 3 and 4). The sample contained 2 % of nanoclay showed the flaking off and dividing completely of nanoclay when it was dispersed by high speed mechanical stirring and sonicating.

The sample contained 3 % of nanoclay was distributed incompletely because it can be found some places nanoclay layers was not devived (Figure 3).

Please See Figure 3.

Please See Figure 4.

XRD diagrams of nanocomposites EE240/I.30E was showed in Figure 3. It can be seen that nanoclay I30E appeared at the $2\theta = 40$, corresponding to the distance d = 22.128 Å. This peak was not appeared in the XRD diagrams of nanocomposite sample contained 2 % of nanoclay by weight. This phenomenon improved that EE240 molecule was poked into space of nanoclay layers, improving the distance d, lead to layered structure of nanoclay was changed, flaked off, and lost. Fig. 4 shows the dispersion for 2 wt.% of nanoclay, I-30E within the epoxy matrix, Separated single layers of clay platelet can be also observed. The TEM image

(Figure 4) clearly shows the montmorillonite expansion Particles (intermediate measurements mean 3.3 and 3.4 nm). The upper left part of the image consists of split montmorillonite Class, that is the exfoliating beads. Stacks alternate between 3 and 6 Layers are visible from the top left to the bottom right of the image. So, The nanoparticles contain interstitials and exfoliants such as Proved by TEM.

3.2.2. Mechanical Properties and Flame Retardancy of Nanocomposite EE240/I.30E.

The effect nanoclay I.30E amount to mechanical properties of material was shown in Table 3.

In Table 3.

It can be seen that at the sample with 2 % by weight of nanoclay, the mechanical properties of material were significantly improved. The tensile strength was improved up to 63.05 MPa, the flexural strength was achieved 116.80 MPa, compressive strength was obtained 179.67 MPa, and Izod impact strength was 12.81 KJ/m², which were increased of 13.59 %, 34.63 %, 15.11 %, and 80.16 %, respectively, in comparison to those of original material.

To study the reason improved the mechanical strength of materials, FE-SEM was took (Figure 5). At the surface of fragment, it can be observed the difference between materials which contained different nanoclay amounts. The fragment surface of EE 240 was smooth and deviation of cracks was not clear.

Please See Figure 5.

When adding nanoclay in to EE240, the fragment surface was rough, the cracks on the surface of material were changed. The spread of cracks was difficult because of nanoclay adding, lead to avoid the concentrating of power, contributing to reduce the appearance of cracks inside material.

However, when the quantity of nanoclay was increased up to 3 or 4 %, nanoclay particles in some places on the surface of nanoclay-EE240 (because nanoclay was excessed, they existed in individual phase, the homogenous structure was broken) were found. Therefore, the cracks were increased and concentrated at these places, lead to reduce mechanical properties. The sample with 1 % by weight of nanoclay has the fragment surface which nanoclay was not appeared much because the adding amount is very small.

It can be seen from FE-SEM images, the fragment surface of original EE240 was smooth, proved that the improvement of cracks affected by external force was convenient.

However, by adding nanoclay in small portions with various ratios, the fragment surfaces of materials were rougher than that of EE240. The cracks can not continue because the nanoclay barriers, so they have to changed direction or discontinued. To continue cracking or change the initial shape, they need to support higher external force, lead to improve mechanical strengths.

The sample with 2 % by weight of nanoclay, some cracks can be seen clearly and they tend to make bend. If the amount of nanoclay over the threshold (3 and 4 % of weight), the compatibility of nanoclay and EE240 was reduced and the concentration of nanoclay was appeared (Figure 3.20). The excess nanoclay made some holes inside the material, the main reason reduced mechanical properties compared to those of sample contained 2 % by weight of nanoclay. Therefore, 2 % by weight of nanoclay is the optimal weight ratio of nanoclay I.30E to disperse into EE240 to make nano layered and interfered structure inside the nanocomposite, which will increase the mechanical properties of material.

The effect of nanoclay amounts to flame retardancy of nanocomposite EE240/I.30E was presented in Table 4.

In Table 4.

The sample EE240/I.30E (2 %) was achieved the highest flame retardancy, as shown in Figure 5. The dispersion of nanoclay in to EE240 affected significantly to improvement of flame retardant ability of material. A protective layer acted like a barrier was formed by nanoclay, which was well known as a heat-resistance and heat-storage material. This layer will reduce the diffusion of oxygen and prevent the evaporation of flammable compounds contained inside the material, limiting the transference of heat comes inside of material, hence decrease the time of burning.

3.3. Investigation of Flammability of Materials Based on EE240 Resin Matrix Cured by DETA in the Presence of Fire Retardants MWCNTs/Chlorinated Paraffin/Antimony Oxide.

As above research, the addition of antimony oxide and chlorinated paraffin lead to increase the fire retardant ability of material because of the simultaneous fire controllable mechanism happened [14].

In Table 5.

Please See Figure 6.

FE-SEM image of EE240/MWCNTs (0.02 wt%)/ Sb₂O₃ (9 phr)/chlorinated paraffin (11 phr) was shown in Figure 7a. It can be seen that Sb₂O₃ was distributed evenly, percolated and combined well with EE240. When the material was broken, the particles of Sb₂O₃ were remained in fracture surface of fragmentation and covered by EE240 (no any space was found). On the other hand, MWCNTs was distributed uniformly in regions

of fire retardant additives (arrow positions), proved for the good compatibility of MWCNTs – fire retardants – EE240. The good dispersion of MWCNTs in EE240 will form coal (ash) layers when material was burned. These layers will cover surface of material, protect it from oxygen, lead to prevent the development of flame. They were also spread to keep material was not dripped, improved the fire retardant ability of material. As we discussed above, the combination of fire controllable mechanisms of several components will improve flame retardancy of materials. To obtain the improvement of flame retardancy of material when mixing several components, the more important thing is the good compatibility of them. The more evenly in distribution of additives, the more improvable in fire retardant ability and mechanical properties of materials.

The particles of Sb₂O₃ were distributed equally and shown clearly the absorption as well as the linkage with ground epoxy. After breaking materials, Sb₂O₃ particles were remained in surface of fragmentation and covered by EE240, and no any space around the particles was observed. In addition, MWCNTs were also dispersed uniformly and sneaked perfectly in the flame retardancy additives (arrow positions). All of them proved the high compatibility of MWCNTs – flame retardants – EE240.

Figure 6 shown the FE-SEM images of nanocomposites at flame – out after burning. The surface of the sample after burning was relatively smooth, no any pore or bubble. The black coal layer was found on the outside surface of material demonstrated clearly the protective effect of the internal structure, the prevention of heat transferring and the reduction of fuel gas when it was burned. Please See **Figure 7**.

The addition of antimony oxide and chlorinated paraffin provided the improvement of fire retardant ability of material due to various fire controllable mechanisms of additives were occurred resonantly and simultaneously. The layered network formed by Sb₂O₃, chlorinated paraffin, e-glass and MWCNTs played an important role as a shield, re-emitting O* radical from the surface of EE240 to outside, declining the degradation rate of polymer.

3.4. Demonstrating the synergistic effects of oxit antymon /chlorinated paraffins/MWCNTs/nanoclay-I30E additives.

Please See Figure 8.

It can be seen form Figure 8, when the sample of composite EE240/MWCNTs/ELO/ oxit antymon /chlorinated paraffins was fractured, the fiber was draw from inside of EE240 ground resin and then broken. On the surface of fiber, a number of resin in combining to other components were remained. In figure 8-a,

showing the sample of MWCNTs/EE240/e-glass when it was broken, the quantity of resin on e-glass was poor, appeared the segregation and burst. This phenomenon proved that the material was became more brittle than original material and smooth surface caused by less adherent.

Specially, in the case of the sample of MWCNTs/nanoclay/EE240/e-glass shown in Figure 8-c, a part of epoxy was broken at the chink where the fiber was draw. This was the evidence for good compatibility of e-glass and ground material. The surface of broken e-glass was rough and harsh, depicted the breaking which needed to use more energy than previous one (MWCNTs/EE240/e-glass).

In Table 6.

Table 6 shows the effect of both MWCNTs and nanoclay I.30E to flame retardancy of materials. The flammability of MWCNTs/EE240/ Sb₂O₃/chlorinated paraffins/e-glass was significantly improved. Additives with nano size were dispersed uniformly in EE240, formed carbon and silicate layers which were thin and heat - durability, covered materials. Adding flame retardancy system including antimony oxide and chlorinated paraffin, the flame retardancy was improved cause by effect of simultaneous flame retardancy mechanisms.

4. Conclusions

The flame retardancy of materials was improved by the combination of nanoclay, MWCNTs, flame retardant complex of chlorinated paraffin/Sb₂O₃ and glass fiber while mechanical properties were maintained as regulations. The flame retardancy was investigated by using UL-94 machine with UL-94V method. The material contained EE240 as ground material, 2 % by weight of nanoclay, 0.02 % by weight of MWCNTs and chlorinated paraffin/ Sb₂O₃ shown the flame retardancy reached to level of V-0. In addition, mechanical properties of material were also improved. One special thing is the quantity of MWCNTs using in this research, it is much lower than other reports in literatures.

The dispersion ability of MWCNTs and nanoclay affected expressively to flame retardant property of nanocomposite material. MWCNTs will increase the formation of carbon layers, which will form a thermal barrier. Nanoclay prevents the transference of heat into inside part of material because it has high specific heat. Finally, the flame retardancy of epoxy resin was improved significantly by using MWCNTs/nanoclay/chlorinated paraffin/ Sb₂O₃ complex.

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Figures

Figure 1. FE-SEM micrographs of the fracture surface of MWCNTs in EE240, ultrasound solution: NB (4 hours); NB(5 hours); NB(6 hours); NB(7 hours).

Figure 2. FE-SEM images of the fracture surface MWCNTs/EE240 nanocomposites: 0.01 wt.% of MWCNTs, 0.02 wt.% of MWCNTs and 0.03wt.% of MWCNTs.

Figure 3. XRD diagrams of initial nanoclay (a) and nanocomposite materials: EE240/I.30E (1%) (b); EE240/I.30E (2%) (c); EE240/I.30E (3%) (d); EE240/I.30E (4%) (e).

Figure 4. TEM images of nanocomposites EE240/I.30E with difference of nanoclay 2 % weight.

Figure 5. FE-SEM images of nanocomposites EE240/nanoclay I.30E.

Figure 6. FE-SEM images of nanocomposites: (a) EE240/MWCNTs (2wt.%)/ Sb2O3 (9phr)/chlorinated paraffins (11phr) ; (b) EE240/ Sb2O3 (9phr)/chlorinated paraffins(11phr).

Figure 7. FE-SEM images of nanocomposites at flame-out after burning.

Figure 8. FE-SEM images of materials: (a) MWCNTs/EE240/e-glass; (b) MWCNTs/EE240/Sb2O3/chlorinatedparaffins/e-glass; (c) MWCNTs/nanoclay/EE240/e-glass.













Figure 4.



Nano clay 2YI30E.006 Print Mag: 312000x @ 51 mm 4:05:08 p 03/06/14 TEM Mode: Imaging

20 nm HV=80.0kV Direct Mag: 150000x EMLab-NIHE

Figure 5.





Figure 7.



Figure 8.



Tables

Table .1. Flame retardant properties of EE240/MWCNTs resin depended on the processing time.

Time Ultrasonic Vibration (hours-h)	Limiting oxygen index,	UL-94HB,	Combustion rate,
	(LOI) (%)	(mm/min)	(mm/min)
Neat epoxy	20.6	28.41	-
4 hours [*]	22.8	27.41	25.78
5 hours	23.2	24.65	23.27
6 hours	23.2	23.03	21.70
7 hours	22.8	24.05	22.34

(*4 hours: EE240/MWCNTs when the ultrasonic time was 4 hours)

Table 2. Flame retardant properties of EE240/MWCNTs resin depended on the weight ratios of MWCNTs.

Samples	% I.30E	Tensile strength, MPa	Flexural strength, MPa	Compressive strength, MPa	Impact strength (Izod), KJ/m ²
Neat epoxy	0	55.90	86.75	156.08	7.11

NC1*	1	58.28	87.30	152.30	6.90
NC2	2	63.50	116.80	179.67	12.81
NC3	3	59.60	97.00	157.32	11.27
NC4	4	58.91	79.90	170.43	10.41

Table .3. Mechanical strength of nanocomposite EE240/I.30E with various I.30E quantity.

MWCNT content	Limiting oxygen index,	UL-94HB,	Combustion rate,
(wt%)	(LOI) (%)	(mm/min)	(mm/min)
Neat epoxy	20,6	28,41	-
0,01	21,9	24,78	23,56
0,02	23,2	23,03	21,70
0,03	23,7	23,90	22,34

(*NC1: nanocomposite epoxy Epikote240/nanoclay-I.30E (1 wt%))

Table .4. The flame retardance	y of nanocomposite EE240/I.30E.
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Nanocompozit epoxy Epikote 240/I.30E	Nanoclay I.30E,	LOI value	Combustion rate,	UL 94HB,
	% Wt	(%)	mm/min	mm/min
Neat epoxy	0	20.6	28.41	-
NC1	1	21.4	27.80	-
NC2	2	23.7	24.50	22.59
NC3	3	22.8	25.76	23.45
NC4	4	21.9	25.05	24.55

(*NC1: nanocomposite epoxy Epikote240/nanoclay-I.30E (1 wt%))

Table .5. Flame retardant properties of MWCNTs/nanoclay/EE240 nanocomposite

Material	LOI, (%)	UL-94HB,	Combustion rate,
		(mm/min)	(mm/min)
EE240/Sb2O3(9phr)/chlorinated paraffins(11phr)	22.8	13.22	20.55
EE240/MWCNTs (0.02 wt.%)	23.2	21.70	23,03
EE240/nanoclay-I30E (2Wt %)	23.7	22,59	24,50
EE240/MWCNTs(0.02wt.%)/Sb2O3(9phr)/chlorinated paraffins(11phr)	25.4	16.39	18.34
EE240/MWCNTs 0.02wt.%/nanoclay2wt.%/Sb2O3(9phr)/chlorinated paraffins(11phr)	29.8	V-1	11.34
		(Rating)	
EE240/MWCNTs(0.02wt.%)/nanoclay 2wt.%/Sb2O3(9phr)/chlorinated paraffins(11phr)/E-glass	33.3	V-0	10.05
		(Rating)	

	-				
Experiment No.	Components of material	LOI	Combustion rate	UL 94V	Whether the cotton be ignited
		(%)	(mm/min)		
1	EE240/e-glass	27.6	25.82	-	yes
2	MWCNTs/EE240/e-glass	28.9	18.56	-	yes
3	MWCNTs/EE240/Sb2O3/chlorinated paraffin/e-glass	31.0	12.11	V-1	sometimes
4	MWCNTs/nanoclay/EE240/e-glass	33.3	10.05	V-0	no

Table .6. Flame retardancy properties of EE240/additives using e-glass fiber

Table of Contents

The use of multi-walled carbon In this study, the multi-walled carbon nanotubes (MWNTs); nanoclay (I30-E) were added to nanotubes and nanoclay for the epoxy Epikote 240 resin as a reinforcement to fabricate MWNTs/epoxy Epikote 240 nanocomposites. Flame retardant additives of chlorinated paraffin/antimony trioxide and simultaneously improving the flame retardancy and mechanical properties multi-walled carbon nanotube (MWCNT); nanoclay (I30E) were embedded in epoxy Epikote of epoxy nanocomposites 240 resin to improve the resin's flame retardant properties. Epoxy sized woven e-glass was Nguyen Tuan Anh1*, Nguyen Quang used to establish a three-way interlink between e-glass, epoxy and MWCNTs; nanoclay; Tung¹, Bach Trong Phuc², Nguyen Xuan chlorinated paraffin/antimony trioxide; Epoxidised linseed oil for improving the interfacial Canh¹ bonding them. To the best of our knowledge, such a comprehensive study is not yet been reported and thus carries a high scientific significance. Res., XX, XXX (20XX)