Penerbit Akademia Baru

Journal of Advanced Review on Scientific Research
ISSN (online): 2289-7887 | Vol. 11, No.1. Pages 13-36, 2015

Matrix Cracking in Reinforced Polymer Nanocomposites: A Review.

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Abstract – *This paper presents a summary about the problem of matrix cracking in reinforced polymer nanocomposite materials. The matrix cracking is reviewed under condition of fatigue loading, tensile loading, thermal loading, and flexural loading mechanism. Moreover, it discussed the effect of the matrix cracking on the material damping property and how the crack growth in the matrix nanocomposite materials is predicted. Fatigue life of the nanocomposite is also discovered after the crack propagation in matrix of nanocomposite is predicted. Continuum damage mechanics are discussed and used to prove the behaviour of the cracked matrix nanocomposite. Current development in this area is briefly discussed.*

Keywords: Matrix Cracking, Nanocomposites, Polymer, Reinforced Materials

1.0 INTRODUCTION

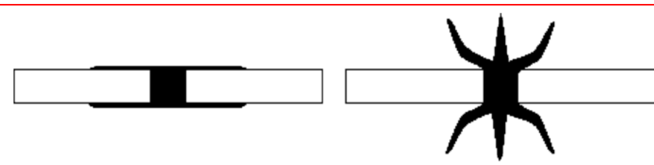
Nowadays, many light duty metal gears and bearings are replaced with polymer product. It is because of the economical and technical advantages of the polymer materials. Generally, polymers are light in weight and exhibit good environmental and chemically resistance. The durability and toughness of the polymers are beyond those of metals. Due to their low strength and modulus, the applications are limited to light duty parts. In recent studies, Liu *et al.* [1], Manjunatha *et al.* [2], Manjunatha *et al.* [3] and Wetzel *et al.* [4] indicate that there are ways to improve the strength and stiffness of polymer composites such as by incorporation of nanosize clay or filler into the virgin matrix. Then, nanocomposite becomes a common material after incorporation of clay or fillers into the matrix. It has reinforcement size of the order of 1-100 nm with the aspect ratio of 10-100.

Compared to standard structural materials, nano-sized non-metallic materials become considerable interest because of their potential for large gain in mechanical and physical properties. For example, carbon nanotube/polymer composites might provide superior properties such as increase in strength and stiffness compared to a standard typical carbon

Journal of Advanced Review on Scientific Research
ISSN (online): 2289-7887 | Vol. 11, No.1. Pages 13-36, 2015

fibre/polymer composites. Knowledge of the prediction bulk mechanical properties of the composite as a function of molecular structure of the polymer, nanotube, and polymer/nanotube interface must be obtained to facilitate the development of nanotube-reinforced polymer composites [5].

High-performance composites can be damaged and eventually fail. All in all, this is a very complex process. It is because of the materials anisotropy and heterogeneity. Damage for homogenous materials such metals, polymers, or ceramics can be monitored by observing the growth of the dominant crack. It is possible to construct a general analysis for stress distribution near the crack tips and it is shown that the same stress distribution has the same form for all cracks. In contrast, heterogeneity of composites leads to a variety of damage models. The damage initially starts by matrix cracking, filler breakage, interfacial debonding between matrix and filler [6]. Fig. 1 show damage modes in single fibre composite fragmentation test for interfacial debonding when the interface in weak and matrix cracking when the interface is strong. There are two causes that might increase the damage of the composite materials which are from the propagation of the initial damage or from the initiation of new type of damage. Compared to homogenous materials, composite materials have many types of cracks. The concept of stress intensity factor is less significant in composite materials. This is because it cannot reduce the description of a crack to a single scalar quantity in composite materials. Crack-tips in composite materials will depend on local environment and crack orientation [7&8].



a). Interface debonding b). Matrix cracking

Fig. 1: Damage modes in single fibre composite fragmentation test: (a) debonding when interface is weak; (b) matrix cracking when interface is strong [9].

Deep knowledge of the fracture mechanics is a must to predict the conditions of composite cracks growth that caused the failure. There are some factors that lead the composite to failure such as when the composite is exposed to fatigue loading, under tensile loading, thermally-induced and under flexural damage. Compared to homogeneous materials, there are many fracture mechanics problems to be solved in composite materials. Moreover, one type of crack might influence the initiation and propagation of another type of crack. To solve the work that involves the new problem, we must consider the interactions between all possible crack types [10].

There are two common methods for analysing damaged composite; continuum damage mechanics and micromechanics of damage [11]. Ricard *et al.* [12] agreed that the process of analysing the damage and prediction of its initiation is called micromechanics of damage. There are relationship between stress, strain and damage for composite under continuum damage mechanics. The mechanical properties of the composites need to be expressed as a function of damage. There are steps for a complete micromechanics damage analysis; observation of the damage, micromechanics in the current damage, damage propagation prediction and comparison to the experiment [13]. Lifetime of the composite materials can be determined by finding out the damage level of that composite [14]. Moreover, continuum damage mechanics only describe and find the relation between the level of damage and mechanical properties of the composite. Besides that, it needs the additional experimental input and normally in the form of measured mechanical properties with the level of damage is known.

In this paper, the studies of the characteristics of damage accumulation in a matrix of polymer nanocomposites subjected to fatigue, tensile, bending and thermal loadings were reviewed. By using several methods to perform the stress analysis of the polymer nanocomposite, prediction of failure of the materials subjected to the increment of loadings can be made. As mentioned earlier, the damage of the materials is determined by several methods such as continuum damage mechanics.

2.0 MATRIX CRACKING UNDER FATIGUE LOADING

It requires detailed understanding when designing against the fatigue, the fundamental fatigue mechanisms and the factors influencing them. As mentioned by Bellemare *et al*. [15], the main cause that contributed to the failure of the most machine parts subjected to variables load was fatigue. They also said the natures of polymers have the viscoelastic property and with the occurrence of the mean load during load controlled fatigue resulting a cyclic creep. Thus, it is important for the material engineer to consider such effects while designing plastic components and keep the level of consequences within its permissible limits.

Experimental study on cyclic fatigue has been made by Wang *et al.* [16], on two polymer nanocomposites in two common failure modes: mechanical failure in epoxy nanocomposites and thermal softening in polyamide (PA, nylon 6) nanocomposites. Results show that the fatigue life of polymer nanocomposite was increased with the addition of hard particles but the soft particles decreased the fatigue life of the materials compared to the original epoxy. The fracture was largest in rubber nanocomposites and least in pure epoxy when applying the same stress amplitude. PA6 (polyamide-6) nanocomposites exhibited fatigue failure due to thermal softening when the maximum local temperature of the specimens subjected to cyclic loading reached the glass transition temperature, Tg, of the material. In addition, Bellemare *et al*. [17] investigated the fatigue properties of clay nanocomposites. They noticed that gains were observed in the mechanical properties due to the inclusion of clay nanoparticles under fatigue loading conditions to ultimate failure. Moreover, Ramkumar and Gnanamoorthy [18], studied the effect of nanoclay addition on the temperature rise and modulus drop during cyclic loading in PA6 and its nanocomposite base. They found that the addition of nanoclay improved the modulus retention compared to the original polymers. Nanocomposite samples sustained more than 105 cycles at all stress levels examined. When at high stress levels, PA6 samples exhibited a rapid drop in modulus, unstable rise in temperature, localized neck formation, and thermal softening which were not observed in nanocomposite samples. Additional of nanoclay decreases the cyclic stain amplitude of PA6 matrix at a given stress amplitude condition and results in less heating and modulus drop.

Bathias [19] discovered that high performance composite materials that reinforced by long fibres of carbon, glass, boron or Kevlar have excellent reputation for a good fatigue behaviour. He also found composites were damaged in fatigue under shear or compression loading more than the metals. Furthermore, the fatigue resistance of composite materials is much lower in compression-compression than in tension-tension. In addition, Bathias and Legorju-jago [20] explored the fatigue initiation and propagation in natural and synthetic rubbers under fatigue loading. They used fracture mechanics approach but found difficulties because of deformability issues. They applied a tension-compression loading to a thick edge specimen with two lateral grooves and found that for a high Load Ratio (R), the fatigue crack would not propagate. In contrast, if R=-1, the threshold disappears. Furthermore, there were also studies conducted by Charles et al. [21] on the polymer nanocomposites that were Under Rolling Contact Fatigue (RCF). They found the surface temperature raised during rolling depends on the test load, speed and influences of the failure mechanism. Moreover, the failure of materials in under low Pressure-Velocity conditions is under the fatigue loading.

There were efforts done by Herrera Ramirez and Bunsell [22] to investigate the effects of fatigue on the accumulation of damage in polystyrene (PET) fibres, in which the destructive tests were performed. In this experiment, the ultimate failure of PET fibres after 4.22 million cycles was due to the presence of an inherent flaw attributed to antimony trioxide (Sb2O3) catalyst particles.

In terms of polymeric composites, Mallick and Zhou [23] have examined the effect of mean stress on fatigue performance of nylon 66 composite fibres reinforced with E-glass fibres. By performing the stress-controlled experiments at a maximum stress ranging from 55% to 80% of the tensile strength, an ensuing creep strain was observed for the experiments at a frequency of 1 Hz for various R (rmin/rmax) values ranging from 0.1 to 0.8. The fatigue creep was shown to be closely correlated with creep engendered from static creep tests.

Poly(ethylene terephthalate) (PET) and PET fibres with embedded vapor-grown carbon nanofibers (PET-VGCNF) were exposed to the cyclic loading and monotonic tensile tests. Results indicated a strengthening mechanism in the low residual strain limit for fatigued PET samples but not for fatigued PET-VGCNF samples. Compared with the unreinforced PET sample, the PET-VGCNF fibres showed greater degradation of mechanical properties as a function of residual strain due to fatigue when cycled at 60% of the fracture stress [24].

As summary for this subtopic, it is proved that the importance to have knowledge in matrix cracking under fatigue loading is crucial. As mentioned earlier, nowadays almost all materials are subjected to fatigue loading, thus with the deep knowledge in this area, researchers can predict the life of the materials and can prevent the matrix cracking by applying the additional materials to the matrix polymer.

Journal of Advanced Review on Scientific Research
ISSN (online): 2289-7887 | Vol. 11, No.1. Pages 13-36, 2015

3.0 MATRIX CRACKING UNDER TENSILE LOADING

Besides the cracking that is caused by fatigue loading, tensile loading also contributes to the matrix cracking. Masters [25] found that under quasi-static or cyclic tensile load, matrix cracking is always the first defect that occurs in composite laminate.

Lim *et al.* [26] investigated the result of loading rate and temperature on tensile yielding and distortion mechanisms of nylon 6-based nanocomposites. They used tensile dilatometry technique to characterize the tensile deformation of the materials. It was shown with the presence of rubber particles grafted polyethylene-octene copolymer maleic anhydride (POE-g-MA) did not change the shear deformation mode. But, the presence of organoclay altered the tensile yielding deformation behaviour of nylon 6 matrix. Therefore the presence of organoclay will distort the growth of cracking in the composite materials. In contrast, there was an experiment done by Hadal *et al.* [27] and they studied the effect of wollastonite and talc on the micromechanism of tensile deformation in polypropylene composites. The tensile modulus showed an increment for mineral-reinforced polypropylenes but the yield strength were still remaining the same. Wolastonite or talc altered the main micromechanism of deformation. It change from the deformation bands/crazing to wedge/neat tearing in the mineral-reinforced polypropylene composites. For the reinforced polypropylene, the final fracture occurs by combining two modes which fibrillation and brittle mode. While for neat polypropylene, the fracture mode was crazing-tearing and brittle deformation. There was an improvement shown in the fracture toughness as reported by Battistella *et al.* [28]. They examined the behaviour of nanocomposites based on epoxy resin modified with fumed silica nanoparticles (0.1, 0.3 and 0.5 vol.%) under tensile loading. They discovered the improvement in fracture toughness of the materials is increased because of the addition of the fumed silica and it shows strong crack propagation resistance in the matrix.

Sandler *et al.* [29] performed some uniaxial tensile experiments on melt-spun polyamide 12 fibres reinforced with carbon nanotubes and nanofibers. The carbon nanotubes involved is arc-grown nanotubes (AGNT), aligned catalytically grown nanotubes (aCGNT), and entangled catalytically grown nanotubes (eCGNT). They used catalytically grown nanofibers (CNF) for nanofibers. It was shown the increment in modulus and yield strength with increased filler content. At filler content of 10%, the finding shows that eCGNT reinforced polyamide 12 composites had significant improvements in modulus (1.6 GPa) and yield strength (45 MPa). In contrast, for unreinforced polyamide 12 had a modulus of 0.8 GPa and yield strength of 21MPa. Thus shows how the cracking will be distorted inside the materials with the addition of the nanofiller. Shindo *et al.* [30] also studied the mechanical responses of cracked carbon nanotube (CNT)-based polymer composites under tensile loading. They constructed an analytical model based on the electrical conduction mechanism of CNT-based composites to forecast the resistance change resulted from crack propagation and the result shows similarity with the experimental results. Moreover, this method was well applied to the observing of the crack behaviour in the nanocomposites and shows a strong relationship between the resistance and the crack length.

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Studies of the crack initiation mechanism was carried out by Adolfsson and Gudmundson [31]. They studied the matrix crack initiation and progression of glass/epoxy laminates with different stacking sequences subjected to bending and tensile load. They observed that the crack happened during the extension test were basically two different layup angles which are 90º and 45º and in plies with the fibres orientation. They concluded that the crack initiation mechanism is mode 1 type. They did the evaluation of the energy release rates and found the similar result. However, from the calculation they found, the energy rate is not suitable to be used in case of critical crack progression parameter for the glass/epoxy composite material. In addition, there is a good result shows between the numerical and analytical data under all cases of the extension of thin composite laminates with transverse matrix cracks [32]. They are combining the bending and extension load in this experiment. Composite that containing two plies in this experiment both experienced surface crack. There are changes in thermoelastic properties with increasing crack density and very rapid for low crack densities. This properties lead to the phenomenon of matric crack closure. They suggested that by including crack closure would result higher stiffness of the composite and recommended to combine their model with a crack initiation criterion to predict the damage development of the composite materials.

Keller *et al.* [33] examined the performance of pultruded glass fibres reinforced polymer (GFRP) in three forms; plate strips as-delivered, plate strips with tabs and plates in tapered form under tensile fatigue. From the experiment, they observed that the specimen temperature increased in line with the increment of tensile test depending on the loading frequency and loading range. Compared to as-delivered specimens, tapered specimens showed higher fatigue life but tabbed specimens showed lower fatigue life. There are degradation rate of the ultimate tension strength (UTS) and a loss of specimen stiffness were observed. This might due to the fibre failures. Their finding is consistent with the previous studies that fatigue failure of fibre-reinforced polymer composites is basically starts at the fibre.

Khiat *et al.* [34] developed a probabilistic strength model for unidirectional composites under tensile loading with fibres in hexagonal arrays. The objectives of their model are to find relation on how the temperature and moisture may affect the ineffective length and the stress concentration of unidirectional composite materials with respect to number of broken fibre. Results showed that the fibre strength and the modulus not really affected by temperature and moisture concentration. However, the matrix composite is influenced by temperature and moisture concentration. After gone through the experiment, they concluded that the proposed model is really affected by the fibre reference strength and shear parameter, but less affected by Young’s modulus of fibre and matrix.

Romanowicz [35], studied an effect of damage that caused by the interfacial debonding on the post initial failure behaviour of unidirectional fibre-reinforced polymers. This material is subjected to transverse tension and examined by using finite element approach. They constructed a micromechanics model and their model is strongly believed to stimulate the evolution of the damage and can explain the softening mechanism. Moreover, the interface strength and interphase stiffness become the main controlled to the post failure behaviour of material. In addition, the local fibre array irregularities contribute the most to matrix cracking

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through the local concentrations and the existence of localization. Fiedler *et al.* [36] also used finite element approach in their studies. They used it as an approach of initial matrix failure in carbon fibre-reinforced polymer (CFRP) under static transverse tensile loading. It shown the hydrostatic tensile stresses in matrix are accountable for the low strain to failure in the loaded composites. The Von Mises criterion is said to be unsuitable in predicting the initial failure in fibre-reinforced composites.

Gupta and Raghavan [37], did some experiment on the creep of plain weave polymer matrix composites under on-axis and off-axis loading to investigate their long-term durability. They constructed model prediction and it was parallel with the experimental results. Moreover the effect of stress and temperature gave less significant under both loading.

Manjunatha *et al.* [38] did some tensile fatigue test on an anhydride-cured thermosetting epoxy polymer incorporating with 10 wt% of well-dispersed silica nanoparticles. Tensile fatigue tests were executed on these composites, throughout which the matrix cracking and stiffness degradation was examined. They discovered that, additional of silica nanoparticles increased the fatigue life, suppressed matrix cracking and reduced crack propagation rate of the anhydride-cured thermosetting epoxy polymer. In contrast, result done by Ferreira *et al.* [39]. They did some experiment of organo-montmorillonite, Nanomer I30 E reinforced epoxy resin composites under tensile loading. They observed a decreased in fatigue strength and fatigue life of the composite material with the incorporation of 3 wt% of nanoclay into the matrix. As a consequence, some of the particle agglomerates, which promote easier fatigue, crack initiation in matrix.

It is crystal clear that the inclusion and addition of the nanofiller to the polymer composite materials will suppressed the growth of matrix cracking, reduce crack propagation in matrix, increase yield strength and improve fracture toughness of the polymer composites. Thus, it is really important to know the right content of the additional materials to the based composite materials in order to increase the life of the composite materials.

4.0 MATRIX CRACKING UNDER THERMAL LOADING

There are consequences on the condition of the composites structures during or after exposed to the thermal condition. During the exposed to the thermal condition, matrix composite materials will loss in stiffness, strength and creep resistance and might cause to failure long before they burn. Nowadays, researchers conducted experiment to explore how the composite materials behave under the thermal loading and they found a way to improve the thermal property of the composite materials by altering the matrix properties either by chemical modification or by inclusion nanoparticles to the based composite materials.

Katsoulis *et al.* [40] studied the consequence of silicate nanoclays and double-walled carbon nanotube sand micro-sized flame retardants (FRs)on the post heat/fire flexural performance of glass fibre-reinforced (GFR) epoxy composites. They found that the increasing incident heat flux, the flexural stiffness and modulus values of GFR composites decreased rapidly. The post-

Journal of Advanced Review on Scientific Research
ISSN (online): 2289-7887 | Vol. 11, No.1. Pages 13-36, 2015

fire flexural properties of the composite retain only 20% of their room temperature flexural properties. It was observed that the formed char network are not consolidated enough to effectively constrain the fibre reinforcements. Moreover Blasi *et al.* [41] created model of thermal degradation of one-dimensional disk of poly (methyl methacrylate) (PMMA) with and without the dispersion of carbon nanotubes exposed to a fire-level heat flux. The existence of nanotubes decreases the peak in the mass loss rate and rises the conversion time, owing to the insulating effects of the surface layer and much larger surface re-radiation losses. They concluded to enhance the effect of flame retardance by increasing the growth rate and/or the thickness of the surface layer.

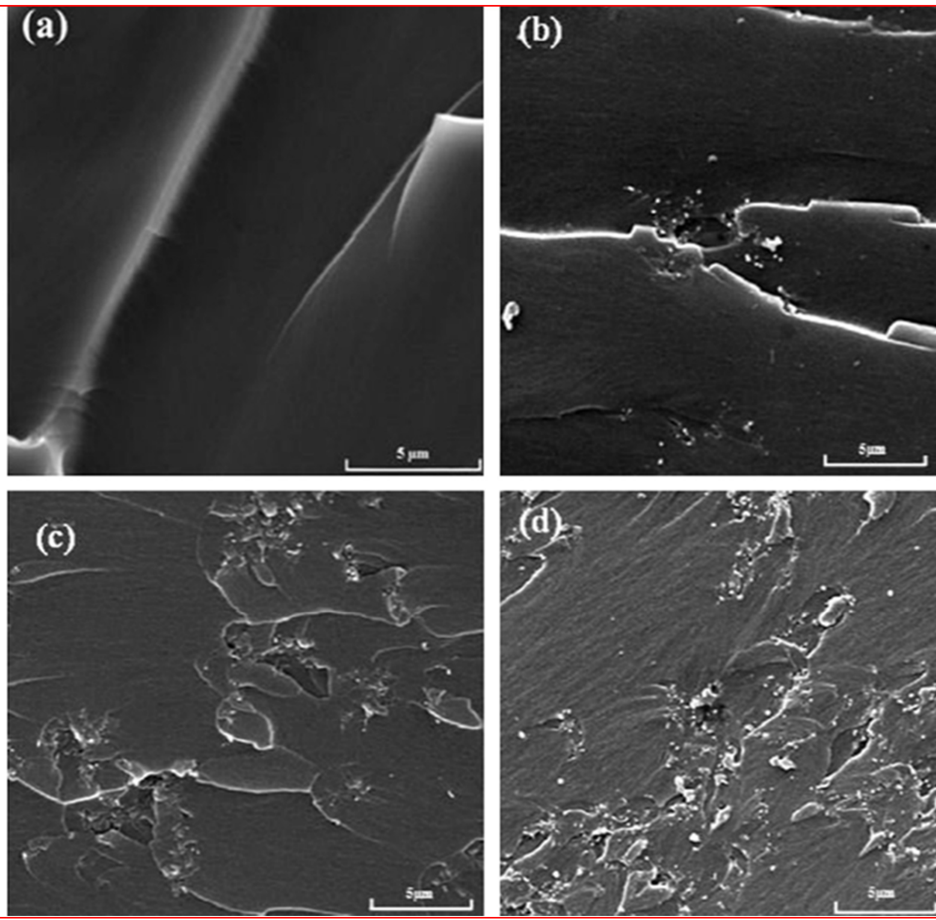

Fig. 2: Fracture surface of (a) neat epoxy (b) 0.1 wt.% (c) 0.5 wt.% and (d) 1 wt.% MWCNTs.[43]

Chu *et al.* [42] studied the thermal expansion properties of 30% chopped glass fibres reinforced Polyetheretherketone (PEEK) composites at cryogenic temperatures. This glass fibre reinforced PEEK was investigated from 77 K to room temperature resulting the thermal expansion coefficient (CTE) of PEEK was closely continuous in this temperature region and it can be reduced by adding more glass fibre. They also found from the thermal expansion

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coefficient measurement indicate that the temperature has little effect the thermal expansion property of PEEK matrix. They concluded that the glass fibre as the filler that can considerably decreases the CTE of PEEK matrix. In other research, Shokrieh *et al.* [43] examined the effect of addition 0.1 wt.%, 0.5 wt.% and 1 wt.% multi wall carbon nanotubes (MWCNTs) in ML-506 epoxy nanocomposites using thermo-mechanical test. They found that due to addition of MWCNTs, the coefficient of thermal expansion (CTE) of epoxy resulted in reduction in both micro and macro levels. Figure 3 shows addition of MWCNTs to the epoxy .Fig. 2(b–d) shows that MWCNTs were equally dispersed in the matrix resin blend expected thus proved MWCNTs played its part as a nano-additive.

It is very clear that the addition of the nanoclays and the dispersion of carbon nanotube to the based polymer composites will increase the flame retardant. This gives us the luminosity for future development in polymer based composite materials to be used in the related industries. Moreover, in term of the CTE, researchers found a way to reduce it by applying additional chopped glass to the PEEK and addition of MWCNT to the epoxy. Thus, it will decrease the CTE both at micro and macro levels.

5.0 MATRIX CRACKING UNDER FLEXURAL LOADING

Strength members that are in service are exposed to the flexural damage. It is common typical stresses and can be described as flexural fatigue under a pre-tension force. The expected life of the materials that are in service consist of the upper limits which is fatigue life under tension-tension fatigue and lower limit which is under flexural fatigue stresses. Under pure tensile cyclic, the service life-times are expected to be longer than the life-time achieved. But, it is expected to be less under the pure flexural fatigue.

Kukureka and Wei [44], studied the damage development in pultruded composites for optical telecommunications cables under flexural fatigue. The objective of their study is to investigate the long-term behaviour of pultruded glass-fibres composites. They proposed a definition for fatigue failure under the flexural fatigue. The development of fatigue damage is found to be dependent based on the stress level. From the finding, when the modulus loss reaches 1.5%, then the flexural fatigue is considered to occur. With this finding, they developed an analysis of failure analysis to estimate the life-time estimates and reliability of the composite materials. They suggested S-log N relation fitted experimental values for predicting the fatigue life at low cyclic stresses of the composite materials. In addition, research done by Rajeesh *et al.* [45] at cantilever bending fatigue test at different humidity levels of polyamide 6 nanocomposites. They found after the fatigue life for polyamide 6 nanocomposites was increased at high humidity and found the crack appears more at low humidity levels.

Lau and Hui [46], did some experiment of carbon nanotubes surrounding by polymer and they found a weak bonding between them. Thus the carbon nanotubes could be pulled out easily and will leave cavities. These cavities lead to a lower flexural strength of a nanotube/epoxy beam compared with the pure epoxy. In contrast, Vodenitcharova and Zhang [47], studied the

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ISSN (online): 2289-7887 | Vol. 11, No.1. Pages 13-36, 2015

effect of bending and local buckling of a nanocomposite beam reinforced by a single-walled carbon nanotube (SWNT).In the thicker matrix layers, the SWNT buckles occurs locally at the smaller bending angles and greater flattening ratios. Thus, it caused the degradation of the nanocomposite strength and strains/stresses in the surrounding matrix become higher. Lyon *et al*. [48], did some experiment on flexural fatigue behaviour of unidirectional glass fibre reinforced polymer (UDGFRP). They processed the epoxy-glass fibre by three techniques which is filament winding, press moulding of pregreg, and pultrusion. In term of flexural damage, the filament winding process has less flexural damage compared to press moulding and pultrusion process.

Davalos *et al*. [49] did some experimental and analytical studies of flexural-torsional buckling behaviour of full-size pultruded fibre-reinforced plastic (FRP) I-beams. They found that buckling is the mode of failure for FRP structures and it could be shown by the experimental results, proposed analytical solutions and finite-element analyses. Mottram [50] investigated the flexural-torsional behaviour of pultruded E-glass composite I-beams experimentally and compared with theoretical prediction. He decided that there was a possible risk in analysis and design of FRP beams without bearing in mind the shear deformation. Shan and Qiao [51], conducted analytical and experimental studies for flexural-torsional buckling of pultruded fibre-reinforced plastic (FRP) composite for open channel beams. Through transcendental function, they obtained eigenvalue solutions to solve problems related to the open channel beams. They also found and constructed analytical solutions to predict the buckling loads of FRP channel.

Jeng and Chen [52], conducted some experiments of flexural failure mechanisms in injection-moulded of short-carbon-fibre reinforced polyether ether ketone (PEEK).These composites were prepared with and without a transcrystalline interphase. Furthermore, these composites were subjected to three point bending test with different cross-head speed applied. There was deep shear cracks observed on the damaged specimen after test has been conducted. It was suggested that compressive cracks occurred first because the stress concentration under loading nose and the shear stress maximum induced the upper shear cracks. There was catastrophic failure that initiated from the tension region. The finding shows that transcrystalline interphase enhanced the fibre/matrix adhesion, improved the flexural deflection and strength of the composite while Dillon [53], examined the fatigue damage growth mechanism of laminated PEEK and subjected to three point flexure. It was observed by using scanning electron microscopy (SEM) machine. Fatigue pattern only can be seen on a coarse scale at very high stress levels and only in unidirectional samples. They can estimate the level of longitudinal ply matrix by the degree of matrix fatigue abrasion occurred. They found that cross-plied laminates resulting extensive transverse, longitudinal ply cracking and interlaminar shearing.

Caprino and Amore [54] performed static and fatigue tests in four point bending on a random continuous-fibre-reinforced polypropylene composite. By comparing their result with the data that available in literature for thermoset-based composites, they thermoplastic matrix does not sensibly affect the fatigue sensitivity of the material, because of the constraint action of the reinforcement. Their data was outstanding parallel with the theoretical predictions.

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Salvia and Vincent [55] performed some experiments and constructed a model of the fatigue damage in glass-fibre-reinforced polymers (GFRP) under deflection-controlled flexural test. They observed that there is a close connection among the stress ageing of the glass fibres, the viscoelasticity of the matrix, the shear strength of the fibre/matrix interface and the resulting fatigue performance of composites.

Researchers have found method and model to predict the life cycles of the polymer composite subjected to flexural damage and improve the flexural strength by adding the carbon nanotube to the based polymer composites. Moreover, flexural damage in composite materials will shorter the life of materials and there is a need to discover new area of polymer composite materials subjected to flexural damage for the improvement.

6.0 INFLUENCE OF MATRIX CRACKING ON MATERIAL DAMPING OF NANOCOMPOSITES

Few references deal with influence of matrix cracking on material damping. Vantomme [56] carried out some parametric studies of material damping in fibre-reinforced plastics. He focused on three parameters which is the matrix, fibre damping and the effect of the interface. In addition, he used an energy balance method and shows that the fibre or matrix contribution to damping lies in the partition of elastic strain energy and highly dependent on the elastic properties of the fibres and matrix. Moreover, he developed a three-phase model and this model shows clearly a poor quality interface and has an important consequence on the energy damping capacity of the unidirectional laminate. While Zhang and Hartwig [57] studied the relation of damping and fatigue damage of unidirectional AS4 carbon fibre reinforced PEEK and fibreglass reinforced epoxy composite. It shows the fatigue cycles in epoxy composite really affecting the damping property. In conclusion, the damping property on material is affected by the elastic properties of fibre or matrix and also by fatigue cycles that exposed to the composite materials.

7.0 CRACK GROWTH PREDICTION IN NANOCOMPOSITE

Filler particles such as carbon in polymers are expected to be the areas in which crack initiation and crack growth were most likely to develop. Thus the particles' presence, triaxial stresses are generated directly above and below the filler particles, restricting shear distortion and encouraging crack initiation. Besides that, the filler particles usually have low surface adhesion to the matrix, and separation from the matrix will generate surface cracks around the particles.

Ivankovic *et al*. [58] studied some prediction of crack initiation and propagation using a cohesive zone model and finite-volume method in three grades of polyethylene of different toughness. Originally, it was unable to provide satisfactory crack growth prediction using a fixed rate traction-separation curve in this model. But there was better agreement of the prediction with the experiment by using more physically realistic family of curves that measured at different rates for the tough polyethylene and also for the brittle polyethylene.

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They suggested for an incorporation of the effects of variations in constraint and perhaps also in temperature for more accurate prediction of the crack growth of tough polyethylene. Huang and Talreja [9] performed some numerical simulation based to predict the micro-cracking in short fiber reinforced polymer composites. They found that the conventional linear elastic fracture mechanics approach does not suitable for their case but the Rice-Tracey ductile fracture model is shown to work well in their case. Using this model, it can predict the measured conical crack including the crack initiation angle and kink formation successfully. Through the analysis, they can predict the propagation of matrix crack from the debonded fiber end towards the next fiber at an oblique angle to the fiber axis. They also discovered the crack is diverting gradually towards the fiber axis before it reaches the neighboring fiber.

Saintier *et al*. [59] studied the crack initiation and propagation under multiaxial fatigue in natural rubber. It was shown that the crack initiate from the pre- existing flaws in the material. They found that, if the material plane rotations are correctly taken into account, it can predict the crack orientation correctly. They also proposed a simple fatigue model that suitable for fatigue loading where there is no reinforcement is observed and used Zebulon FE code. This model can locate the crack initiation, predicting the fatigue life and the crack orientation on structures. May *et al*. [60] studied and constructed an advanced model for matric cracking initiation and propagation under the fatigue loading. They used acoustic emission (AE) and dye-penetrant enhanced X-rays to determine the onset matrix cracking in composites with thick ply blocks. The result X-rays shows a good correlation to the number if matrix cracks shows by AE. Their proposed model is shown suitable for both mixed-mode loading and pure mode loading. The predictions were compared with the experimental data obtained and both were parallel especially for the case of the mixed-mode configuration. Therefore, their model is proven to be suitable for predicting the crack initiation and propagation in composites under the fatigue loading.

Bernal *et al*. [61] studied the failure and failure prediction analysis for polyethylene flawed pipes using the EPRI/GE procedure under tension test. This experiment was conducted under full scale structures. There is agreement between the actual burst pressures determined with both the predicted critical pressures values from limit load analysis and EPRI scheme for the axially cracked pipes under internal pressure. Even though there is plastic collapse occurred during the experimental observation of the failed structures, the same experiment been applied to the circumferentially externally cracked pipes, the result parallels with the experimental data. In contrast, the predicted critical loads from the EPRI procedure overestimates measured value of failure for the circumferentially internally cracked polyethylene pipes. Frank *et al*. [62] investigated the remaining life of four different polyethylene pipes used after 30 years in gas and water distribution services in Austria. They used an extrapolation concept to predict the creep crack growth by means of fatigue tests. As a result, all the pipes are in a good condition and proves it can prevent a quasi-brittle failure to remain in good condition for 50 years.

Boonyapookana *et al*. [63], carried out fatigue crack growth tests of epoxy resin composite reinforced with silica particle under various load ratio R and to investigate the consequence of R-ratio on the crack growth in the composite material. The crack growth arranged by ΔK

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indicated clearly R-ratio's dependence even under zero crack closure. But, crack growth curves arranged by K_{max} displayed that crack growth behavior of the present composite was time-dependent. They also discovered the mechanism of the crack grows it starts at micro-cracking nearby the interface between silica particle and resin matrix. Moreover, it was occurred ahead of a main crack and then micro-cracks coalesce with the main crack to propagate.

As discussed here, there are many methods to predict the crack initiation and propagation depending on the situation. To predict the crack initiation of the brittle polyethylene, we can use a cohesive zone model and finite-volume method. In the other hand, Rice-Tracey ductile fracture model can predict the micro-cracking in short fibre reinforced polymer composites. In addition, some of the researchers here constructed a model to predict the crack propagation and its starting location under the fatigue loading. Thus, it can be used for future research that is related and making improvements to the existing model. There also some research done here that used the crack propagation to estimate the life of the material. By doing this, we can estimate the life of the materials and take the necessary action if the material is believed to damaged. Lastly, some of the researcher found the crack in nanocomposite starts from the filler in the matrix nanocomposite materials.

8.0 CONTINUUM DAMAGE MECHANICS OF MATRIX CRACKING IN NANOCOMPOSITES

Continuum damage concepts can be useful to simulate the failure behaviour of engineering structures. There will be theories and constitutive equations in the continuum damage concepts. The damage parameter will be incorporated into the constitutive equation. The need of an experimental data is to quantify a damage criterion and damage evolution law. Common finite element techniques will be performed to elaborate the mathematical model formulation. If there is no special precaution taken, the numerical results shows to be unacceptably dependent on the measure of the spatial discretization. Thus it shows that a simple but effective method leads to preservation of objectivity [64].

Lurie *et al*. [65], developed a multi scale model of the continuous media and use the second-order unified gradient model to describe a spectrum of cohesion and superficial phenomena within a unified continuum description. The proposed model of the interphase layer cannot be approximated by analysing the problem using spherical inclusion and even by the classical Eshelby solution for a three-phase sphere. It is because this model gives both more general and detailed description of local boundary interactions. They used asymptotic averaging technique of homogenization for filled composites and were generalized for a higher-order model. A general Eshelby solution was established and this method presents the real way of predicting dependence of effective properties of composites on the manufacturing process. Lurie *et al.* [66], were contributed in kinematical variational method. They proposed the virtual work of internal forces and they constructed the mathematical formulation of the interphase layer model for multified problems in composite materials.

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Odegard [67] developed a constitutive models for polymer composite system reinforced with single-walled carbon nanotubes (SWNT). The interaction of the polymer/nanotube interface is highly dependent on the local molecular structure and bonding due to their same size scale. The lattice structures of the nanotubes and polymer chains at these small length scales cannot be considered continuous. Moreover, the traditional micromechanical approaches that were formulated by using continuum mechanics cannot be used to determine the bulk mechanical properties of the materials. Thus he proposed an effective continuum fibre by using an equivalent-continuum modelling two SWNT/polyimide composite systems. Some problems in calculating the overall elastic properties of composite materials was studied by Sevostianov and Kachanov [68]. They calculated the elastic properties of matrix composite materials that contain two different populations of inclusions are the three-phase hybrid composites.

There are few studies that using continuum damage concept to simulate the damage behaviour in the composite materials. The classical Eshelby model cannot be used to describe a spectrum of cohesion and superficial phenomena with spherical inclusion. Some of the researcher proposed a mathematical model to overcome the problems in composite materials. In addition, the researcher used the continuum damage concept to determine the mechanical properties of the materials. It is clearly shown that continuum damage mechanics can be used to simulate the damage in the composite materials and determine the mechanical properties in the composite materials.

9.0 FATIGUE LIFE OF MATRIX NANOCOMPOSITES

The fatigue life of polymer composite materials can be predicted by knowing the crack growth rate. For most geometry, crack growth rates increased in size, and since the stress intensity, K at the tip of the crack increases. Swanson *et al.* [69] demonstrated that the stress intensity controls the crack growth rate by decreasing the load during crack growth at various rates. The calculation of the crack growth rate of composite materials by using the Paris Power Law as in Eqs. (1).

$$\frac{da}{dN} = A(\Delta K)^n (K_{mean})^m \tag{1}$$

Where c = crack length

N = number of cycles

$\Delta K = K_{max} - K_{mean}$

A, n = experimentally determined constant

Hence, the lifetime of the composite materials can be calculated if the fatigue load is constant. Direct integration may be performed by finding an analytical expression for the da/dN curve.

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The fatigue life of the composite materials will affected by temperature, fracture properties of the composite materials, molecular changes during fatigue and loading conditions of the composite materials.

There are many methods to predict the fatigue life of the composite materials other than using the Paris Power Law. Saintier *et al.* [70] carried out some investigation of multiaxial fatigue life prediction of natural rubber. The investigations involved the fatigue crack initiation with push-pull, torsion and tension-compression tests. They proposed two fatigue crack criteria; first one is based on the first and second invariant of the Cauchy stress tensor, and the second one is, based on the micromechanisms of crack initiation, which contains a critical plane method under large strain conditions using a micro to macro approach. They found that the second criterion give the best result in term of predicting the fatigue life. Morevoer, Shariati *et al.* [71] performed an experimental study on the behaviour of polyacetal or Polyoxymethylene (POM) under uniaxial cyclic loading. They calculated strain ratcheting, strain range, strain energy density and the slope of the stress–strain hysteresis loops based on the obtained stress-strain data. In addition, they calibrated the material constants using the stress, strain and energy approaches. The mean stress was used in the equivalent damage parameter and was included as the effect on the fatigue life of polyacetal. Moreover, they discovered that the stress and energy they used were successful in predicting the fatigue life of polyacetal. Furthermore, Hutař *et al.* [72] developed a new methodology for a lifetime assessment of internally pressurized polymer pipes using a linear elastic fracture mechanics approach. They realized that, this method need to be combined with the experimentally determined creep crack growth in order to successfully use this method as lifetime assessment. Evaluation of simulated lifetime with experimental data from tests of internally pressurized pipes verified the principal applicability of the concept suggested. Thus, they believed that the combination of the creep crack growth tests and numerical simulations can be a very great instrument for lifetime estimation of plastic pipes under different loading conditions.

Liu *et al.* [1], found the neat epoxy (E), nanosilica (S6, S12) increased in fatigue threshold but nanorubber (R6 and R12) did not. There is some effect observed on the fatigue threshold when both silica and rubber nanoparticles were added into epoxy and these nanoparticles were decreasing fatigue crack propagation. In other words, the live of the composite materials were increased. Moreover, they found the hybrid composites which combined rubber/silica/epoxy possesses well-balanced elastic stiffness and fracture toughness thus increases the life of the composite materials. Moreover, the same agreement is also reported by Manjunatha *et al.* [2]. They presented their (S–N) results on nanosilica particle and micro rubber particle altered epoxies. They found that adding either 9 wt-% micro rubber particles or 10 wt-% nanosilica particles into epoxy gave almost identical fatigue life improvements. Close examination of the test data suggests that a positive hybrid effect exists, especially at low applied stress amplitudes. In addition, Wetzel *et al.* [4] discovered by adding aluminium oxide(Al2O3) nanoparticles significantly reduced the fatigue crack propagation, matrix plastic deformation and crack pinning of epoxy materials. Therefore, it leads to the increment of the epoxy materials fatigue life. Again, Manjunatha *et al.*[3] fabricated two types of glass fibre reinforced

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plastic (GFRP) composites, GFRP with neat epoxy matrix (GFRP-neat) and GFRP with hybrid modified epoxy matrix (GFRP-hybrid) containing 9 wt-% of rubber micro particles and 10 wt-% of silica nanoparticles. They found that the fatigue life of GFRP-hybrid composite has 4-5 times higher than the GFRP-neat composite.

Kane *et al*. [73] examined the effects of the hydroxyapatite (HA) reinforced high density polyethylene (HDPE) and its fatigue behaviour subjected to four-point bending fatigue test. They used 20 and 40 vol% of either whiskers or powder of HA as reinforcement in the composites. The results show the composites containing 40 vol% HA exhibited decreased fatigue life compared to those with 20 vol% HA and it shows less stiffness loss and leads to more tolerant of fatigue damage. The results also show that HA whiskers can improve the fatigue life and damage tolerance compared to HA powder and it can be used as the reinforced polymers for synthetic bone substitutes.

Jones *et al*. [74] suggested by using Hartman–Schijve equation representation of delamination growth in nanocomposites, there are similarities between crack growth in metals, delamination growth in composites and the environmental degradation of adhesive bonds. This equation also may be useful for the damage tolerance assessment of small occurring defect in nanocomposite structures. They concluded that this equation can be used to predict the fatigue life of materials. In addition, Zhang *et al*. [75] carried out some investigations on fatigue life prediction of fibre reinforced concrete (FRC) under flexural load. They used semi-analytical method to predict the fatigue behaviour based on the equilibrium force in the critical cracked section. They found good correlation between experimental data and the model constructed. Their finding reveals that the fatigue performance in flexure of FRC materials is strongly influenced by the cyclic stress-crack width relationship within the fracture zone. Furthermore, they found that by optimising the bond properties of aggregate-matrix and fibre-matrix interfaces can optimise the fatigue behaviour of FRC structures in bending.

Loos *et al*. [76] carried out some studies on the effects of additional carbon nanotube (CNT) to the polyurethane composites. The composite materials were exposed to the tension-tension cyclic fatigue tests at various load levels. They observed that the tensile energy was increased to 38%, breaking the polyurethane composites. Moreover, the fatigue life of polyurethane composites in the high-stress amplitude was up to 248%. The mechanism for enhancement in fatigue life of polyurethane is expected because of the dispersed CNT in the composite materials and they were parallel with the results of Yu *et al.* [77]. They explored the fatigue life of multi-walled carbon nanotube (MWCNT) reinforced epoxy-matrix composites. The result of carbon nanotube weight fraction was studied. The composite materials were subjected to cyclic loadings with stress amplitudes of 8.67MPa and 11.56MPa. With addition of 0.5wt%-MWCNT/epoxy composites, the fatigue lives are 10.5 and 9.3 times of the average fatigue life of neat epoxy respectively. They observed through the micrographs that fatigue behaviour of MWCNT-reinforced composites was significantly affected by the separation and even distribution of MWCNTs in the matrix composites. Moreover, Jen & Yang [78] carried out some experiments of carbon nanotube (CNT)/epoxy composites using the two-stage cumulative fatigue behaviour. They used Miner's rule to evaluate the cumulative fatigue life

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of the composite materials and compared the prediction results with the experimental data but it was failed. They found that the non-linear damage theories based on the variation of the stiffness and the electrical resistance gave good approximate results of the cycle ratio at the second stage.

There are tests done by some researcher to know the factor that innfluence the fatigue life of the composite materials. Fern *et al.* [79] predicted the fatigue life of calcium carbonate based composite paper coating. There were fatigue tests conducted to determine the influence of porosity, binder volume and binder type used. They found that porosity influenced by binder volume was known as the key cause to effect a change in fatigue life time. Moreover, they discovered by increasing the polymer content and lowering glass transition temperature of the polymer binder can increase the lifetime of the composite materials. In addition, Shi *et al.* [80] constructed fatigue life prediction for the fibre reinforced polymer lamina with two damage degrees. The fatigue test was on 0º and 90º unidirectional were conducted respectively. They observed matrix cracks occurred first, and then the fibre failure occurred immediately. Moreover, they discovered that the fibre cannot bear the tension besides the fibre initial direction. Timmaraju *et al*. [81] investigated the influence of environment such as still air and circulated water mist environment on flexural fatigue behaviour of polyamide 66/hectorite nanocomposites. There was a significant drop in fatigue life in mist compared with in air when there is high hysteretic energy dissipation per cycle and high induced stress in mist. The strain and hysteretic energy dissipation per cycle against fatigue life curves showed linear relationships on log–log scale in air. The failure analysis discovered a different deformation and fracture mechanisms in air and mist.

The use of Paris Power Law in predicting the fatigue life of the composite materials is becoming trend nowadays. But, it only can be used if the loading applied is constant. The addition of nanoparticles to the matrix composite materials will increase the fatigue life of the materials. In addition, inclusion of carbon nanotube to the matrix composite materials will increase the fatigue life of the material. In this topic also discussed the factors that affect the fatigue life of the composite materials such the environment, the orientation of the matrix composite, and fracture properties of the materials.

10. CONCLUSIONS

In conclusion, this paper reviewed the matrix cracking in reinforced polymer nanocomposites. There are many factors that caused the matrix cracking in reinforced polymer nanocomposites such as materials that were exposed to cyclic fatigue loading, tensile loading, thermal loading, and exposed to flexural loading. Researchers found the main cause that really contributes to the matrix cracking is the materials exposed to the fatigue loading. There are proof that many materials and products of polymer based materials in the related industries are exposed extremely to the fatigue loading. Researchers are aware of this problem thus they discovered how to suppress the matrix cracking by the addition and inclusion of the nanofiller such as carbon nanotube to the matrix nanocomposites. The more inclusion of the nanofiller to the matrix nanocomposites will delay and suppressed the crack growth and propagation of the crack in the matrix nanocomposites. In addition, the orientation of the fibre in the matrix composite also affects the crack growth. Therefore, by inclusion of the nanofiller to the matrix composite also increase the fatigue life of the polymer composite. This happened because of the relation between the crack growth rate and fatigue life of the material is inversely proportionate. There also discussion on the matrix cracking to the damping property of materials. Researchers discovered that the damping property on material is affected by elastic properties of fibre or matrix and fatigue that applied to the composite materials. Moreover, this article also discussed how researcher predicts the crack growth in the reinforced polymer nanocomposite. There are many methods can be used such as continuum damage mechanics in which it simulates the failure behaviour of the materials. It has theories and constitutive equation in the concept. Researcher also used FEA method to analyse the damage level in the composite material. Therefore, they can predict the location of the crack and also predict the crack growth in nanocomposite materials.

ACKNOWLEDGEMENTS

The authors wish to express their gratitude to Universiti Selangor, Malaysia (UNISEL) for the financial assistance, the Department of Manufacturing and Materials Engineering of International Islamic University Malaysia for assistance in conducting this study and grant PRGS12-003-0003 awarded by Malaysia of Higher Education.

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