

LITERATURE REVIEW OF FIBER REINFORCED POLYMER COMPOSITES

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ABSTRACT

Polymer-matrix composites (PMCs) have been used for a variety of structural memberships for chemical plants and airplanes, since they have outstanding performances, such as lightweight and good fatigue properties. To hold the long-term durability and to estimate the residual life of the composites under some hostile environments, it is an important issue to clarify the fracture and/or the failure mechanism in each service conditions. Degradation of components made from polymeric materials occurs in a wide variety of environments and service conditions, and very often limits the service lifetime. Degradation occurs as the result of environment-dependent chemical or physical attack, often caused by a combination of degradation agents, and may involve several chemical and mechanical mechanisms. The main concern of this review will be to examine the causes of degradation of polymeric components from the completion of fabrication to ultimate failure.

KEYWORDS: Degradation, oxidation, Hydrolysis, moulding

I. INTRODUCTION

Many polymers are prone to degradation caused by weathering in which photo-chemical reactions, involving ultraviolet solar photons and atmospheric oxygen, lead to chain scission. The chemical reactions may be accelerated by elevated temperatures caused by the warming effect of the sun. Alternatively, or additionally, the chemical reactions may be accelerated by the presence of stress that may be applied externally, or may be present in the form of moulding stress, or as the result of a temperature gradient or of differences in thermal expansion coefficient at different locations within the molding. Failure is often simply taken as the fracture of the component, but degradation of some other property, such as the transparency or surface gloss may render a component unserviceable. In broad terms, the majority of failures that are the consequence of polymer degradation can be attributed to one of three types of source such as 1] Molecular degradation caused during processing, usually due to elevated temperatures (as in melt processing) and often in combination with an oxidizing atmosphere, 2] Degradation in service caused by the natural environment and 3] Attack by an aggressive chemical, again during the service lifetime. The type of degradation referred to in third one includes as the major problem environment-sensitive fracture, in which contact with a liquid chemical leads to craze initiation and growth. This can be a particular problem with consumer goods, where the service conditions are not under the control of the supplier; the end-user may employ an inappropriate cleaning fluid, for example. Significant research has been conducted in this area over the past 20 years and several test procedures have been developed. It will be necessary to examine the mechanisms of failure and the features of the environment that control them, and then to look for possible remedies. The methods of testing are discussed with reference to their application in establishing ranking orders for plastics with respect to their weather resistance in determining the effectiveness of additives such as anti-oxidants: in providing data for lifetime prediction; and in research into the mechanisms of failure and the development of improved materials. There are elements of degradation behaviour that are common to all polymers and elements that are peculiar to a particular polymer. Much research has been

conducted on the important commodity polymers poly (vinylchloride) PVC), polyethylene, and polypropylene, and these materials are used by way of example in this review.

II. POLYMER DEGRADATION

2.1. Chemical mechanisms of degradation:

In an aggressive chemical environment polymer molecules- break (chain scission), cross-link, or suffer substitution reactions. Substitution is the least common and causes the smallest property changes and will not be considered further in this review. Scission and cross-linking both occur under natural weathering conditions, and molecular degradation can also take place during processing. There is general agreement that molecular degradation occurs almost exclusively at defects in the molecule. Much research has been conducted into the chemical reactions involved and there are many papers and several reviews on this topic [1-7].

2.1.1 Degradation Studies

A Universal Testing Machine is an instrument used for the measurement of loads and the associated test specimen deflections such as those encountered in tensile, compressive or flexural modes. It is used to test the tensile, flexural and Inter Laminar Shear Strength (ILSS) properties of materials. The flexural strengths of the specimens were determined for different alkali exposure durations using the three-point bending test as per ASTM-D790. The specimens (80 X 8 X 3mm) were tested with a span length of 50 mm in air using an instrumented 10 ton capacity UTM (M/s Kalpak, Pune).

Table 1 Degradation of Flexural strength at T=70°C

No. of hr of exposure	Carbon- Epoxy	Carbon -Vinylester	Carbon -Isopolyester
0	834.452	432	370
120	765.92	380	304
248	732	348	264
365	684	320	232
480	648	300	216
600	636	294	208

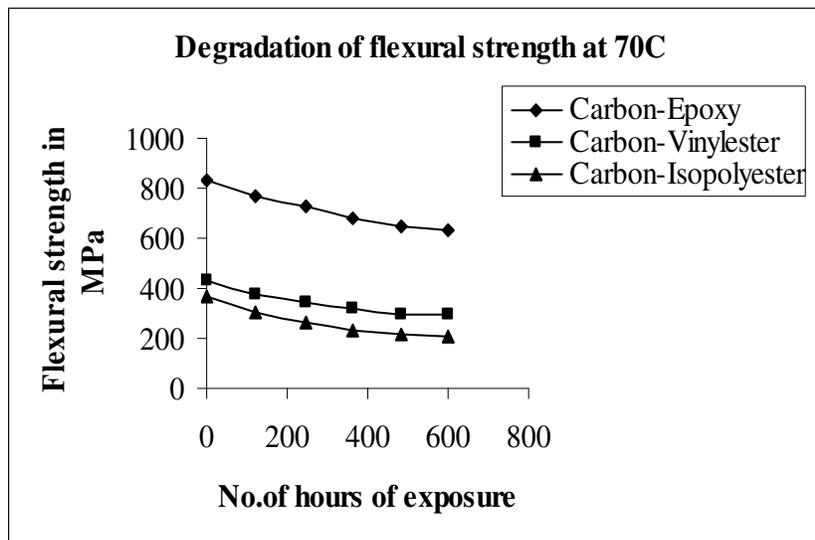


Fig 1 Degradation of Flexural strength at T=70°C

The specimens were tested for tensile strength as per ASTM-D638 the specimen dimensions of Length: 216mm, Thickness: 3mm and Width: 19mm at a cross head speed of 1 mm/ min.

Table 2 Degradation of Ultimate tensile strength at T=70°C

No. of hr of exposure	Carbon- Epoxy	carbon -Vinylester	carbon -Isopolyester
0	508.014	358.666	295.183
120	468	336.667	256
240	446	314	225
360	430	298	203
480	415	273	180
600	393	260	172

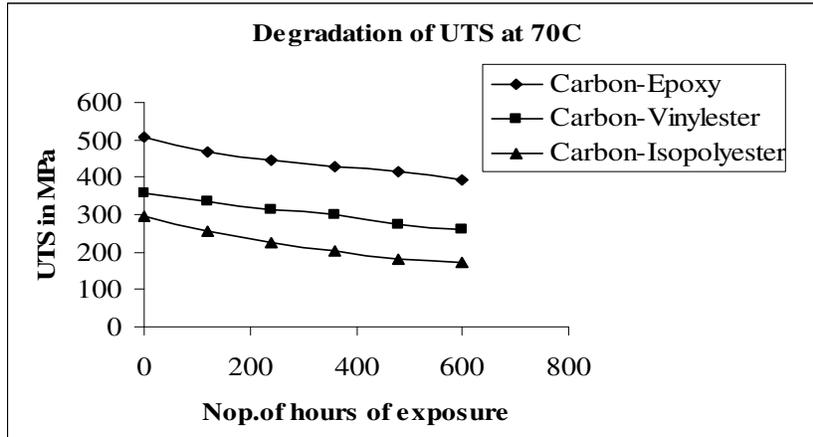


Fig 2 Degradation of Ultimate tensile strength at T=70°C

Three-point bend test was carried out to determine the ILSS values of the specimens in accordance to ASTM D2344. The testing was done at a crosshead speed of 1mm per minute.

Table 3 Degradation of Inter laminar shear strength at T=70°C

No. of hr of exposure	Carbon- Epoxy	Carbon -Vinylester	Carbon -Isopolyester
0	51.2396	22.4003	16.7829
120	50	21.7016	15.1
240	48	20.921	14.7
360	46.6	20.1743	14.3
480	45.3	19.2967	13.4
600	44	19.009	12.9

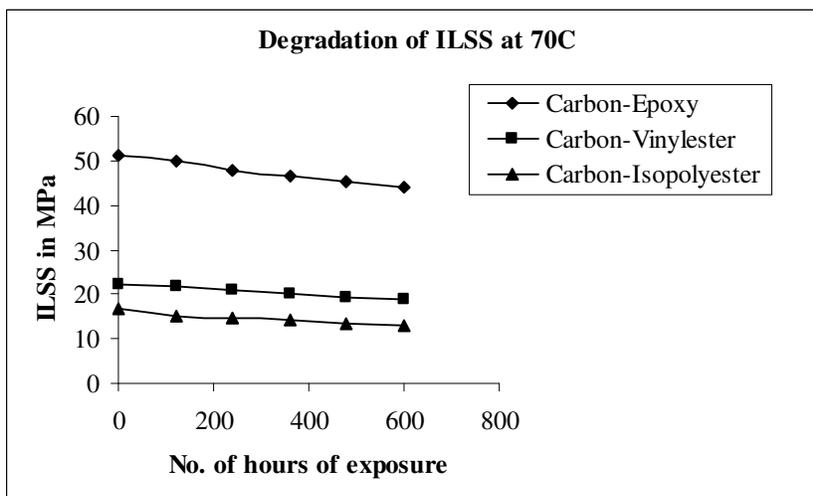


Fig 3 Degradation of Inter laminar shear strength at T=70°C

2.2. Photo-Oxidation:

Of major importance is the process of photo-oxidation is proceeds by a radical chain process initiated either by dissociation caused by the collision of a photon with sufficient energy with a polymer molecule, or as the result of some impurity present, for example trace metals from the polymerization catalyst. Once initiation has occurred, converting the long- chain polymer molecule, PH, into a radical, P, the reactions are as listed by Davis and Sims [8]:

Termination is then normally through the reaction of pairs of radicals. The reaction schemes are affected by trace metal impurities such as polymerization catalyst residues or contaminants from processing machinery, for these may catalyse some of the degradation reactions, for example Reaction 4 [11]. Degradation can still occur slowly in the dark through the formation of hydroperoxides through intermolecular back-biting hydrogen abstraction by peroxy radicals [12].

The reactions listed above would not cause serious degradation of the engineering properties of the material as they stand because the long-chain nature of the polymer molecules is preserved almost unchanged. Degradation occurs because the radicals are unstable and may undergo scission reactions. Discussion of scission reactions for polypropylene is given in a recent paper by Severini et al. [13]. Hydroperoxides produced by Reaction 2 or by other means can be decomposed by u.v. radiation with wavelength below 360 nm giving a PO radical, as shown in Reaction 3. The decomposition of hydroperoxides is generally acknowledged to be a key feature in the degradation of polyolefins, though their behaviour in polyethylene, in which they do not accumulate [14]. (Note that hydroperoxides accumulate both in polyethylene and polypropylene on thermal oxidation [15]). The presence of carbonyl groups in a degraded polymer indicates that oxidation has taken place and also warns that the material is vulnerable to further deterioration because they are photo-labile. Aldehyde and ketone carbonyl groups are common products during processing and the effect of processing on the subsequent degradation behaviour has been identified as of significant importance [15]. Although most studies of photo-oxidation have centred on U.V radiation, the need for information on the behaviour of polymers for insulation (polyethylene) and jacketing (PVC) in nuclear installations has stimulated study of the effect of γ -radiation. Clough and Gillen [16, 17] found that radiation dose and temperature act synergistically in promoting degradation.

2.3 Thermal decomposition and oxidation:

Thermal degradation is of relevance here because damage suffered by the polymer during processing at elevated temperature can lead subsequently to further deterioration under the conditions of photo-oxidation. Thermal degradation is a serious problem with PVC and has been the subject of much research. The initial step in the process of degradation is dehydrochlorination, with hydrogen and chlorine atoms on adjacent chain carbon atoms stripping off to form HCl and leaving behind a double bond in the polymer backbone, adjacent sites become less stable, more HCl may be stripped off, and a conjugated polyene structure develops. This causes yellowing of the material. HCl catalyses the reaction which is therefore auto- accelerating unless steps are taken to remove the HCl. The process is accelerated in oxygen but can occur in the absence of oxygen at temperatures above 1200 C [18]. Troitskii and Troitskaya [19] conclude that abnormal unstable fragments have a major influence over thermal degradation of PVC.

Mechanico-chemical degradation may occur during processing, producing free radicals that may then initiate dehydrochlorination in PVC [20, 21]. It is expected that dehydrochlorination will initiate at preexisting defect sites in the polymer, though there is evidence that it may not be restricted exclusively to them [20]. The small amount of oxygen present during processing allows the formation of hydroperoxides by reaction with radicals. After thermal degradation the polymer will suffer further degradation during later stage in processing, or under other conditions favouring thermal oxidation, or under conditions of photo-oxidation [22]. Even though the shearing action during processing is generally believed to promote molecular damage, the inclusion of lubricants to reduce the viscosity during processing does not produce any significant reduction in the vulnerability of the product PVC to oxidation [23]. The susceptibility to further degradation will depend on the amount of HG present, the degree of unsaturation and on the hydroperoxide content [20].

Although generally regarded as a lesser problem than with PVC, degradation of polyolefins occurs during processing as well. Mellor et al [24] found that the lifetime under U.V. exposure was very

sensitive to the degree of oxidation that took place during processing on a two-roll mill and that the rate of UV degradation was related to the increase in melt flow index that occurred in the material.

Billiani and Fleischmann [25] used the weight-average molecular weight, M_w , to assess molecular degradation during injection moulding of polypropylene and found that it was more sensitive to increases in melt temperature than to increases in shear rate. There was no significant difference between the molecular weight of material taken respectively from the skin and the core, and they deduced that degradation occurs in the plasticizing system and/or in the sprue. Amin et al. [26] claimed that processing low-density polyethylene (LDPE) at 160 °C produces hydroperoxides that have a photo-initiating effect, whereas those produced by thermal oxidation in the range 85-95 °C do not [26]. This has been examined further by Lemaire and co-workers [27, 28] and by Gugumus [29] who discuss the chemistry of oxidation and the nature of the oxidation products. Gugumus further claims that the mechanisms may be adapted to other polymers including non-olefinic polymers such as polystyrene and polyamides [29], though this may not be so because Ginhac et al. [27] report that hydroperoxides which initiate new oxidation reactions form in polypropylene under thermal oxidation conditions that do not cause the formation of active hydroperoxides in polyethylene.

2.4 Hydrolysis:

Hydrolytic attack can cause chain scission in some polymers, leading inevitably to deterioration in properties. A general hydrolysis scheme can be summarized as follows:

Polymers susceptible to this kind of attack include polycarbonate. The reaction can be unacceptably fast at elevated temperature and can be a problem with articles that need to be sterilized; Some polymers absorb water, leading to other problems. Nylons become plasticized and their Young's modulus can fall by as much as an order of magnitude. Some relevant references are given in a recent paper by Paterson and White [30]. When water is absorbed in polycarbonate in sufficient quantity it can form disc-shaped defects that act as stress-concentrating flaws and cause a serious fall in toughness. A review of the literature and some new results has been presented recently by Qayyum and White [31].

2.5 Attack by pollutants:

The attack of polymers by pollutants has been reviewed by Rånby and Rabek [32]. Some of the pollutants themselves are photolytic, leading to further products that may cause degradation. For example, SO_2 is oxidized and reacts with water to produce H_2SO_4 .

2.6 Mechanical degradation:

If a chemical bond is placed under sufficient stress it will break. It may not always be easy to apply such a stress because deformation mechanisms intervene. For a polymer chain bond to be broken, the segment in which it is contained must not be able to uncoil (i.e. it must be extended between entanglements or cross-links already) nor slip. Such constraints may be present in a cross-linked polymer, where the short chain segments become fully extended at fairly low extension in a highly oriented polymer, or possibly at the tip of a growing crack. Molecular fracture has been shown to occur in this way using electron spin resonance to detect the free radicals that are produced when chain scission occurs.

2.7 Stress-aided chemical degradation:

The phenomenon of mechanico-chemical degradation (or sometimes more specifically "mechanico-oxidative" degradation) has been known to occur in rubbers for many years [33]. The effect of stress on the rate of chemical degradation in a much wider range of polymers has been reviewed by Terselius et al. [34] and Popov et al. [35]. Unlike the case of mechanical degradation dealt with in the previous section in which very high stresses are needed to break a chain bond, a more modest stress may accelerate scission caused by chemical reaction. The most highly stressed bonds will still be the most likely to react [36, 37] so that bonds contained within short segments or highly strained bonds near entanglements will be most vulnerable. Highly oriented polymers are generally more resistant to this type of attack than when in more randomly oriented form because the molecules tend to share the load evenly, so that the chance of overstressing is much less. Nevertheless, the rate of oxidation of oriented polypropylene at 130 °C was found to increase with load at high loads [38, 39].

III. EFFECTS OF PROCESSING:

Much of the discussion of thermal degradation is related to the problem of molecular degradation during processing, when the temperature required to produce the desired flow properties for a moulding operation is often high enough to promote significant degradation, especially if oxygen is present. There will often be circumstances during processing operations in which stress-aided chemical degradation will occur this problem in the formed product, but some degradation of this kind may have already occurred during processing. There is a further aspect of processing that has not yet been dealt with and that is the morphology of the moulded or formed polymer. The rate of cooling is often quite high in molding operations and varies considerably from one position within the moulding to another. As a consequence the morphology of a semi-crystalline polymer varies substantially within an injection moulding, which normally contains equiaxed spherulites in the core and an oriented structure near to the surface. This is discussed further in section 6.9. The important point to note here is that degradation reactions occur almost exclusively in the amorphous phase because it takes up oxygen much more readily than the crystal phase [64] and that there can be a strong influence exercised by the morphology. It is further suggested that oxidation may occur preferentially at the crystal--amorphous boundary where the effects will be most damaging [65—67]. Nishimoto at [68] found that the crystal structure of their polypropylene samples varied with the quenching conditions and that there was a marked variation in property deterioration even though the (γ) radiation-induced oxidation did not differ. The diffusion rates of the various reactants are very different in the crystal and non-crystal regions of most polymers.

Another morphological feature is molecular orientation, which can occur in either crystalline or amorphous regions. There have been several studies of the effect of orientation on the degradation of polymers and some of them are referred to in section 2.3, in which the effect of orientation on stress-aided chemical degradation was discussed. Photo-degradation is slower in oriented polyethylene in the unstressed state as well as when an external stress is applied [69]. Some of these topics are discussed further by Slobodetskaya [70], who observed that hydroperoxides accumulated at a lower rate in oriented polypropylene than in unoriented material.

IV. CREEP FRACTURE

The mechanism of polymer-matrix composite is complicated than the other materials, since it can fail under a constant load that is significantly lower than its static strength even at room temperature and its degradation mechanism has not been fully discussed yet. McLean [4] described the creep behavior of unidirectional composites. It was assumed that a fiber was elastic and a matrix was viscoelastic. The matrix stress transfers to the fiber stress with time and makes the fiber strain increase equal to the composite strain. Curtin [5] predicted the rupture strain and the maximum fiber stress of unidirectional composites in view of estimating the probability of fiber breakages in its own cross section. Du and McMeeking [6], Sofronis and McMeeking [7] and Ohno et al. [8] also predicted the creep rupture time of unidirectional composites under tensile loads. They discussed about the relaxation of the interfacial shear stress that could decrease the unidirectional composite's strength. Among the above studies, although only the fiber breakages were considered as the fatal damage, the interfacial debondings that were likely to progress even for the normal PMC were not examined.

This time-dependent failure would promote fiber breakages and degrade the mechanical properties of composites [6–10]. From this point of view, Beyerlein and co-workers [11,12] investigated the interfacial debonding propagation and verified that the interface failed with time in a single fiber composite under a constant strain. In this paper, fragmentation tests were conducted tests with a single fiber composite to examine the interfacial debonding.

V. PREDICTIONS OF FATIGUE LIFE

FRP laminates have been subjected to the variable amplitude loading. The linear cumulative damage rule and Palmgren–Miner rule were used for the prediction of fatigue life under the variable amplitude loads. However, the linear cumulative damage rule for the materials is not useful for describing the complicated fracture mechanism [13–15]. Therefore, the cumulative damage was evaluated using residual strength or residual stiffness as the parameter of damage [13, 15, 16]. Recently, Yao and

Himmel [17] assumed that the cumulative damage was proportional to the decrease of strength, and they modify the analysis by considering the residual strength caused fatigue damage in FRP. In this paper, the variable amplitude cyclic loading tests of two-stage were conducted with quasi-isotropic [45/0/-45/90]S CFRP laminates Stress-corrosion crack problem of FRP Considering the degradation of a fiber embedded in a composite near the crack tip caused by the solution diffusion, the fragmentation test using a single fiber model specimen was employed. Fragmentation tests were conducted to investigate the degradation mechanism using a single fiber composite. The specimen was consisted of ECR-glass/vinylester and an E-glass/vinylester. Effects of environmental solution diffusion into a matrix on interfacial shear strength have been evaluated with immersion time.

VI. FRAGMENTATION TEST AND INTERFACIAL SHEAR STRENGTH

The specimen was constituted by an E-glass fiber as the reinforcement and a vinylester resin as the matrix. And the geometry of a specimen is shown in Fig. 2. The interface exposing to the solution was sealed at the end of the specimen in order to reduce the water uptake through the interface. The maximum interfacial shear strength was calculated by the Cox formula, which assumed that the fiber/matrix interface was perfectly bonded. Fig. 3 shows the maximum interfacial shear strength as a function of the water absorption rate. And the interfacial shear strength decreased against the water absorption rate. The maximum interfacial shear strength was influenced by matrix Young's modulus. Therefore, the interfacial shear strength decreased as a function of the water absorption rate, and it depended on the mechanical degradation of the matrix.

VII. RESULTS AND DISCUSSIONS

The effect of Alkali exposure for neat casting of Epoxy, Vinylester, and Isopolyester at 70°C are shown in Figure (1,2,3). The specimens after alkali exposure show increase in degradation substantially with increase in time of exposure. The percentage drops in UTS for epoxy/carbon, vinylester/carbon and Iso-polyester/carbon after 600 hours of exposure were 24.81, 26.73, and 63 % respectively The percentage drops in flexural strength for epoxy/carbon, vinyl ester/carbon and Iso-polyester/carbon after 600 hours of exposure were 28.73, 41.17, and 71.29 % respectively at RT and 31.20, 46.93, and 77.88 % respectively at 70°C. Carbon /epoxy show better performance than the others and carbon/Iso-Polyester exhibiting least ILSS.

VIII. CONCLUSIONS

Composite materials have a great potentiality of application in structures subjected primarily to compressive loads. Composite materials have attractive aspects like the relatively high compressive strength, good adaptability in fabricating thick composite shells, low weight and corrosion resistance. But, material characterization and failure evaluation of thick composite materials in compression is still an item of research. Glass reinforced plastics have wide application to naval & other vessels accompanied by application of conservative design safety factors due to limited durability data and to account for underwater shock loading. Increasingly GRP is being proposed for critical marine components such as masts, submarine control surfaces, transmission shafts propellers, & superstructures, submarine casings, radomes, etc.

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