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Modified Epoxy for Fibre Reinforced Polymer Strengthening of Concrete Structures

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Abstract: Fibre-Reinforced Polymer (FRP) is a preferable material for repairing concrete structure due to excellent material properties and effective installation cost over the long-term maintenance of structures. The successful application of FRP strengthening system very much depends on the bond between the concrete substrate and the FRP material using epoxy adhesive. Epoxy acts as a bridge to transfer stress from the concrete to the FRP material. The use of wet lay-up technique to apply FRP onto concrete structure requires epoxy to undergo a curing process normally referred to as cold curing. This paper intends to give a review of the problems with cold-cured epoxy and its effect on structural performance. Cured epoxy is characterised as brittle; therefore, modifications of epoxy are required to toughen the epoxy to suit the purpose of repairing a concrete structure. The methodological approaches from previous studies on modified epoxy were collected and reviewed in this paper. This review also offers some important insights regarding the use of sustainable materials, as well as recommendations for new epoxy in the future.

Keyword : Modified epoxy, FRP strengthening, cold-cured epoxy

1. Introduction

Reinforced concrete is one of the most popular artificial materials on Earth widely used in constructing public structures such as buildings, dams and bridges. However, a large number of concrete structures deteriorate due to overloading, corrosion of reinforcement, fire, environmental exposure, the inadequacy of design and the lack of maintenance [1]-[4]. Therefore, the repair and rehabilitation of deteriorated concrete structure are essential not only to extend the service life of a structure, but also to ensure safety and serviceability.

Fibre-reinforced polymer (FRP) is one of the materials used for repairing public infrastructure which enhance the load-bearing capacity of a structure [4], [5]. Its properties such as high specific strength and specific stiffness, lightweight, recyclability, environmentally-friendly ease of handling, high durability against corrosion, and reduced manufacture time make FRP preferable to traditional construction materials such as steel [1], [2], [6], [7]. However, the performance and durability of FRP strengthening system do not only depend on its material but also upon the FRP adhesive substrate performance and adhesive bond interface.

A common technique for FRP application is wet lay-up whereby it involves the attachment of dry FRP material onto the affected concrete structure using epoxy [4], [7]. Epoxy is typically a combination of part A (resin) and part B (hardener) [8]. Both parts are usually mixed in different ratio as suggested by epoxy manufactures. The most common type of epoxy is cold-cured epoxy resin which hardens and achieves considerable mechanical properties at ambient temperature. However, uncertainty occurs when it is applied to structures whereby the durability of the material will be an issue.

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This paper presents a review of the use of cold-cured epoxy as an adhesive in the FRP strengthening system for repairing concrete structures. The information from recent studies in modifying epoxy by incorporating various materials provide insights into the suitability, performance and identification of several critical factors for the success of the application of modified epoxy.

2. Fibre-Reinforced Polymer Strengthening

FRP is an advanced material that has become a preferable choice for the strengthening and repair of reinforced concrete structure. The application of FRP for strengthening a reinforced concrete has gained attention over the last two decades. The FRP strengthening system is usually used when a structure is required to carry additional load (particularly old bridges), or when there is a deficiency in existing designs and construction. When the costs for demolition and reconstruction are high, rehabilitation for the structure may be the best option to restore the loss of strength and to improve the serviceability of the structure [1]-[3], [7]. The application of FRP is not limited to improving flexural properties for a structure, but also shear capacity and compression member.

FRP composites typically composed of continuous fibre embedded in a thermoset resin matrix (epoxy, vinyl ester, or polyester resin) that holds the fibres and transfers the load between them [7]. High mechanical and thermal properties make FRP widely used in various applications, from aerospace to sport [6]. FRP possesses high tensile strength-to-weight ratio and is not vulnerable to corrosion. The efficiency of FRP can be optimised due to the anisotropic properties of FRP, making it flexible in design [9]. Three commonly used FRP for strengthening are Carbon, Glass and Aramid [3]-[5], [10]. Among all the FRPs, Carbon Fiber Reinforced Polymer (CFRP) has been widely used for strengthening work due to its excellent mechanical properties compared to the others [4], [11]. FRP has two different forms: uni-directional and bi-directional arrangement. Uni-directional FRP has higher tensile strength and elastic modulus and is most suitable for increasing the structural capacity of an element [12].

FRP can be applied by two techniques: (i) the prepregs are adhesively-bonded as prefabricated elements on the concrete structure and (ii) the dry FRP is applied through wet lay-up onto the concrete substrate as shown in Fig 1. In the precured technique, as it is manufactured in industry, the properties of FRP are controlled in terms of its stiffness and strength compared to the wet lay-up technique which is commonly applied in situ. However, the latter techniques provide enormous flexibility following the geometrical configuration of the structure element required for the repair [4], [7], [13]. Apart from that, the strength of the FRP on the concrete structure depends on the adhesive that bonds the FRP to the concrete. The epoxy resin is similar to the matrix of the FRP that can form continuity between the FRP and the concrete. Careful monitoring during the curing process is necessary as it affects variation in the final performance [7].

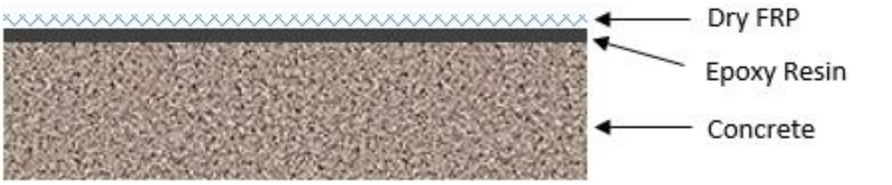


Fig.1 - Step by step installation by wet lay-up technique

3. Epoxy as a Bonding Agent

Epoxy has a wide application in the automotive, aerospace, marine equipment, chemical plants as well as in the construction industry [14-18]. The functions of epoxy in the construction industry are coating, self-levelling floor, bonding of prefabricated elements in bridges, grouting and structure repair [19]-[21].

Epoxy is a thermosetting polymer which offers excellent chemical and mechanical properties, good insulating properties and a good environmental and chemical resistance over a wide range of temperature [6], [15], [17], [20]. Epoxy is typically a combination of part A resin with part B hardener. The common epoxy resin is produced from combining epichlorohydrin and bisphenol A to give bisphenol A diglycidyl ethers [15], [20], [22]. On the other hand, the hardener is produced from polyaminoamides or amidoamines [15]. Both parts are usually mixed with different ratio suggested by epoxy manufactures. The combination of part A and Part B epoxy resin is usually in the form of solid, liquid, solution and semi-formulated pastes [14].

One of the most important properties in epoxy is the glass transition temperature (*Tg*). *Tg* is the temperature when the epoxy transitions from hard, glassy material to soft, rubbery material. Therefore, the curing temperature plays a crucial role [23]. *Tg* can be measured using dynamic mechanical analysis (DMA) which measures the viscosity of the polymer. DMA is one of the methods commonly used to determine *Tg* which provides mechanical properties information of a specimen when a small deformation is applied to the material as a function of time and temperature. The storage modulus E', the loss modulus E' and the loss factor, tan σ are the parameters found in determining the *Tg* using DMA [24]. These abovementioned parameters are depicted in Fig 2.

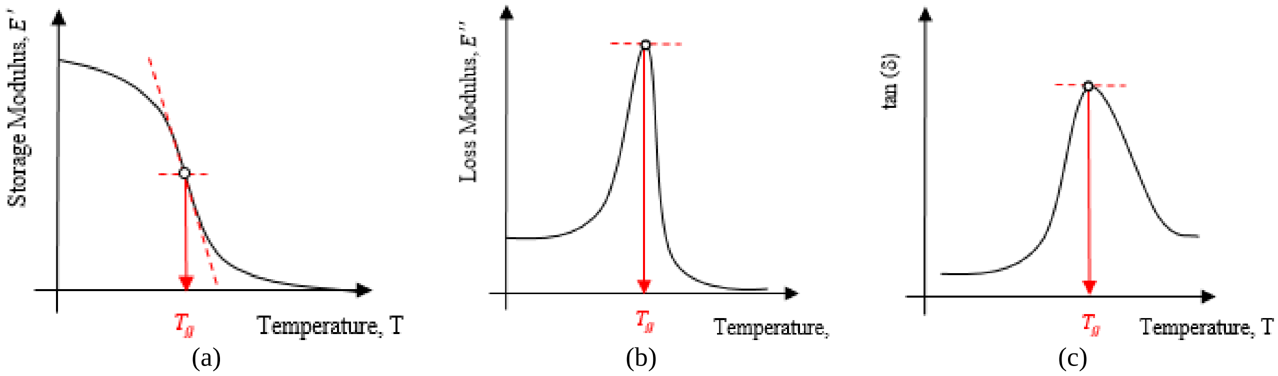


Fig. 2 - Schematic representation of several key temperature values during glass transition [25]

When *Tg* is exposed to mild external temperatures, the adhesion between FRP and concrete will be reduced. Besides, exposure to moderate temperatures (above *Tg*) of non-fully cross-linked thermoset polymer can lead to post-curing. Post-curing will increase *Tg*, stiffness and strength of the resin. The *Tg* value never exceeds 75 ˚C even if the cross-linking has been completed through post-curing procedures. These systems are operating in a non-equilibrium state, with the different properties evolving in time and as an effect of the variable external condition [7].

The efficiency of reinforced concrete strengthened with FRP depends on the proper bonding between the FRP and concrete with an epoxy adhesive. The adhesive plays an important role in bonding the fibres and the concrete together. Particularly, for interfacial debonding and concrete cover separation, the mechanisms are related to the adhesive mechanical properties and interlaminar fracture toughness [7]. The adhesives should not be applied in wet or damp places to avoid weakening or reducing their effects. Environmental conditions such as extreme temperatures, direct contact with dust or rain could retard or accelerate the resin curing time. The required properties of an adhesive for excellent strengthening are as follows: it should have compatible thermal properties with both FRP and concrete, easy to mix, apply and cure, not sensitive to normal variations in the moisture content of prepared surfaces and present low creep [1].

Although epoxy is an important composite in construction industries, it has some effects on health and is hazardous [5], [29]. Epoxy can cause health impacts on people who are living and working with this material. This is because epoxy fumes out toxic gases at temperatures above 82˚C [5]. In addition, it cannot be applied in humid conditions. Various plastic can give hazardous effects on the health and studies have shown that epoxy resin is one of the most hazardous polymers [30]. Contact of epoxy with dermatitis (upper layer of human skin) can affect the sensitisation of human skin by approximately 40.25% [31]. Besides, the epoxy polymer can cause angiosarcomas of the liver and asthma because of the initiator used with epoxy [32]. In addition, epoxy resin is difficult to recycle due to the cross-linked structure from curing and leftover epoxy also creates a waste disposal problem [33-35]. Therefore, safer epoxy resin and utilisation of sustainable alternatives resources are being studied to reduce the effect on human health and environment [36].

4. Curing of Epoxy for FRP Application

Epoxy has been widely used in engineering application as an adhesive for concrete due to their advanced mechanical and chemical properties. The performance of epoxy depends on the curing process which involves changes of properties of thermosetting plastic through chemical reaction [32]. The low viscosity of epoxy resin can be formulated and at room temperature when fully cured, it exhibits excellent mechanical properties, as well as resistance towards aggressive environment [7].

The polymerisation of part A of resin gives rise to a rigid network-type structure. It occurs when the hardener (part B) is present as a curing agent. The polymerisation depends on the heat or radiation on the ingredient and the curing mechanism. The amount of hardener used depends on the resin and the curing condition that affects the final performance of the cured epoxy [7]. The properties of the final solid-state epoxy are dependent on the density of the cross-link formed. High cross-link density can improve the glass transition temperature (*Tg*), tensile modulus and chemical resistance properties [32].

In the curing process of the epoxy, there are two stages known as gelation and vitrification. Gelation is the stage whereby the resin transforms from a liquid to a rubbery state. During the gelation process, the flowability of resin reduces due to the formation of cross-linking polymers and became insoluble but swell as it is imbibed in the solvent. The vitrification stage takes place when the resin change from a rubbery to a glass state, also known as the glass transition phase [36]. In this stage, the molecules are linked to each other, creating a three-dimensional network of epoxy.

The three methods for curing epoxy are cold-curing, heat curing and using ultraviolet light. Epoxy cured at elevated temperature is known as heat curing. In heat curing, there is a pre-cure step which cures at a low temperature and a post-cure step which cures at a high temperature. The need for post-curing of epoxy at elevated temperature is to ensure a complete cross-linking reaction to acquire the ultimate mechanical and physical properties of the resin [32].

For practical and economical reasons especially for FRP strengthening method, the use of epoxy as an adhesive led to the use of cold-curing method. In the cold-curing process, epoxy resin left in ambient temperature is able to achieve acceptable mechanical properties in a reasonable curing time [37]-[42]. Aliphatic amines act as curing agents which can react with epoxy at low temperature [44]. However, longer time is needed to cure the epoxy in the cold-cure method compared to a few hours of curing with a source of heat necessary to provide a satisfactory degree of cross-linking. The conversion of the epoxy group in the cold-cured method never reach completion and the *Tg* is moderate (not more than 60˚C) [44]-[47].

The curing of epoxy at a higher temperature than the ambient temperature can restart the cross-linking reaction with an increasing *Tg* and the stiffening of the system. The absorption of external water produce decrease in *Tg* which can affect the mechanical properties and enable post-curing even at a low temperature [48]. These thermodynamically unstable system can undergo physical aging which leads to densification process that can affect the mechanical and temperature properties [49]-[51]. Physical aging is a thermo-reversible phenomenon which can be eliminated by heating above its *T*g. When temperature exceed the *Tg* of aged cold-cured epoxy, the elimination of physical aging with the recovering of the initial properties take places. These cold-cured epoxies are constantly subjected to aging and de-aging process which depends on the actual meteorological weather [51], [52].

5. FRP-Concrete Interface

Long curing time in the cold-curing technique in FRP application exposed the structure to a humid environment which affects the bonding properties of FRP-concrete interface. Exposure to high temperature, as well as high humidity can result in bond degradation and eventually affect the mechanical and durability of the entire strengthening system [54], [56]. Most studies observed that the presence of moisture vapour transmission can cause air pocket under the FRP laminate. This relates to the trapping moisture in the concrete after exposure to high temperature. The formation of air pocket can affect efficiency and cause premature failure of the system. The surface moisture on strengthened concrete, extreme humidity as well as low temperature have major adverse effects on bond strength. Although high temperature insignificantly affects bond strength, it is recommended that FRP must be installed at a temperature between 4˚C to 49˚C to avoid set-time and saturate workability at high temperature.

The durability and performance of the epoxy bond have been studied by [57] on concrete and FRP in the marine environment. Four types of different environments were studied - wet/dry and hot/cold cycles in 15% salt-water, wet/dry cycles in 15% saltwater, outdoor condition and air-conditioned laboratory condition. The results show that bond degradation is the greatest under the wet/dry cycles caused by moisture absorption by the epoxy that is detrimental to bond durability. The amount of crosslinking agent used to make the epoxy network induced a high moisture absorption characteristic, hence affects the toughness and reduces the application of the system [58].

A simplified method for assessing the bond behaviour of the epoxy, FRP and concrete interface is using a direct tension pull-off test as shown in Fig 3. The test is carried out by applying FRP sheet on a concrete substrate, and then the steel plate (dolly) will be bonded with similar epoxy over the FRP sheet. A groove will be made around the bonded FRP and the load will be applied to pull the dolly [59]. A failure from the test is illustrated in Fig 4 and the description of the failure is described in Table 4 [60]. Failure mode G is the most desirable whereby the failure occurs entirely on the concrete structure and not in the epoxy or FRP [61].

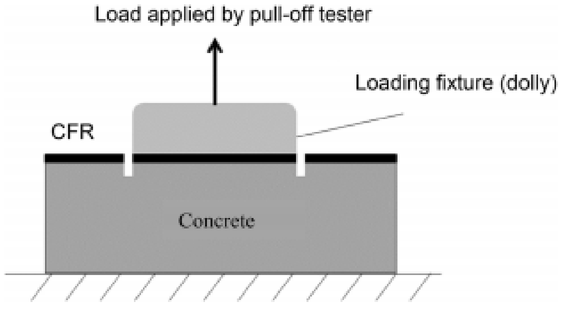


Fig. 3 - Direct tension pull off test mechanism [60]

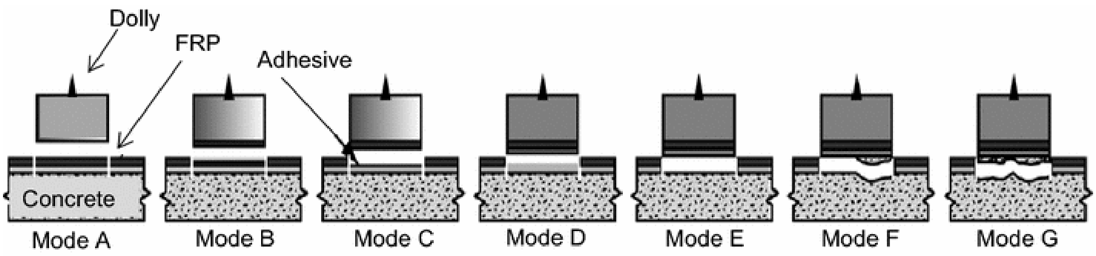


Fig. 4 - Failure modes for the pull of test [61]

Table 1 - Description of the failure modes [61]

Failure mode	Failure type	Possible causes of failure
A	Bonding epoxy failure at dolly (loading fixture)	Use of an inappropriate bonding epoxy system for affixing the dolly
B	Cohesive failure in FRP laminate	Incomplete epoxy saturation of the fibres or environmental degradation of the FRP material itself
C	Epoxy failure at FRP/epoxy interface	Improper selection of epoxy, contamination of epoxy, improper or incomplete epoxy curing, contamination or improper preparation or cleaning of adherent surfaces, or environmental degradation
D	Cohesive failure in epoxy	Contamination of epoxy, incomplete curing, and environmental degradation of the material.
E	Epoxy failure at FRP/concrete interface	Improper selection of epoxy, contamination of epoxy, improper or incomplete epoxy curing, contamination or improper preparation or cleaning of concrete surfaces or environmental degradation
F	Mixed cohesive failure in concrete and epoxy at the epoxy/concrete interface	Inconsistent FRP-concrete adhesion. Failure is partly in epoxy and partly in concrete
G	Cohesive failure in the concrete substrate	Proper adhesion of FRP-concrete. Desirable failure mode

6. Consistency of Modified Epoxy

The mix ratio for epoxy with supplementary materials either by additional or replacement, can alter the curing time, physical characteristic and eventually the mechanical properties of modified epoxy. Three important criteria can be gathered from the literature regarding the consistency of a modified epoxy: the mixing procedures, the method to ensure the homogeneity of the combined materials and the method to assess the flowability or viscosity of the mixture before tested for other properties.

The procedures for mixing epoxy with supplementary material vary among researchers. Liquid polymers named Amine Terminated Butadiene Acylonitirle (ATBN) and Carboxyl Terminated Butadiene Acrylonitrile (CTBN) have been used by [62] to produce modified epoxy. ATBN and hardener were mixed and heated before adding epoxy resin. On the other hand, CTBN and epoxy resin were mixed and heated before combining with hardener. Either resin or hardener heating technique produces a homogenous mix. Similar procedures were observed for [63] whereby fine sand was incorporated into resin or hardener before they were mixed. The study found that if the sand is added later after the hardener and resin is mixed, the workability of the mixture is reduced due to the interaction of epoxy preventing the penetration of sand. The epoxy cannot be mixed in large quantities because of the heat resulting from the mixing of the two parts composed of A and B, which promotes speed of hardness in addition to the nature of its texture.

A carbon nanotube (CNT) in liquid form has been used by [64] in commercially available epoxy resin. To ensure the homogeneity of the mixture, a mechanical stirrer was used, and the mixture was then subjected to Ultra Sonicator for 30 minutes. Ultrasonication is a method to achieve homogenisation and stability of the mixture, especially for nanomaterials. Lastly, the hardener is added into the CNT-resin mixture and was later mixed.

The use of sand and sand washing waste as supplementary materials in producing epoxy mortar have been studied by Yemam [65]. The amount of epoxy resin used is 10% to 25% by weight facture of the mortar which is based on the workability condition. The dry sand and sand washing waste were mixed for 2 minutes with an automatic mixer to attain uniform distribution. On the other hand, the epoxy resin was prepared, before being poured into the dry mixture and continued with further mixing for 2 minutes.

The consistency of epoxy resin can be determined using a test method for the flow of hydraulic mortar (ASTM C1437 or ASTM C230) [67]. The truncated cone is filled with epoxy on the flow table (Fig. 5), whereby the top can be raised and dropped by rotating the cam. Then, the cone is removed, and the cam is rotated so that the table can be raised and dropped 25 times in 15s. The diameter of the initial flow after the cone is removed and the final diameter will be recorded [67]. This method helps to control the flowability of the mixer when a supplementary material is used before assessing further properties such as the strength.

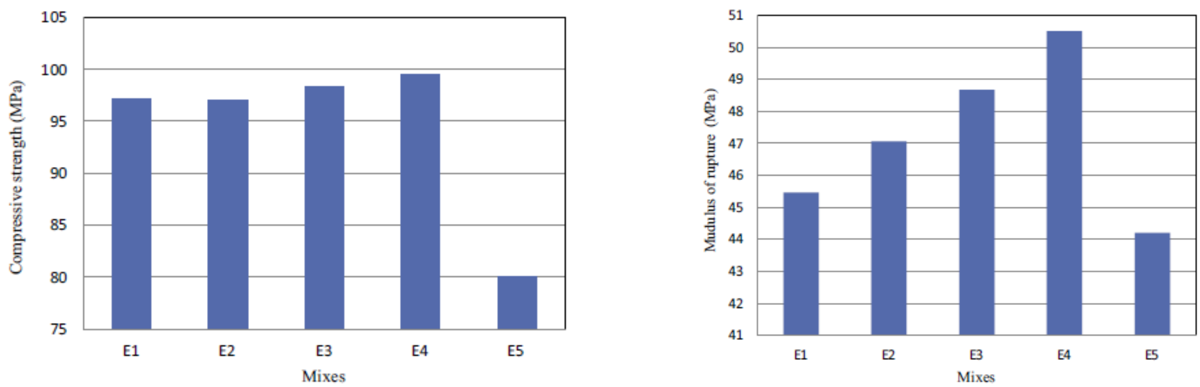
Fig. 5 - Flow table

7. Hardened Properties in Modified Epoxy

Cured epoxy is brittle, therefore modifications made to neat epoxy should be able to toughen it by improving its ductility and strength [60], [69]. In relation to bonding materials for FRP, problems such as thermal incompatibility to the concrete substrate, minimum application temperature, as well as curing condition, eventually affect the performance of the repaired structure [63], [70]-[72]. Generally, hardened properties of cured epoxy will be assessed through its compressive and tensile strength. Meanwhile, its bond strength is determined by bonding the material to the concrete substrate.

Strength properties of modified epoxy resin have been studied by [73] using nano-silica filler with a flexibiliser and is subjected to a maximum of 100 cycles of wet and dry conditions at temperatures ranging from 20 to 60 degrees Celcius. The tensile strength of the modified epoxy resin increases as the wet and dry cycle increased due to the high temperature during the cycle improving the degree of curing. However, after 25 cycles, the strength gradually decreased. The study also involves an investigation on the bond strength of FRP strengthened samples using modified epoxy resin with the same environmental exposure. The findings confirmed that the bond strength increased, followed by strength loss.

Fine sand has been used by [66] as an addition to neat epoxy to reduce the cost of epoxy for bonding with CFRP. Sand-to-epoxy ratio ranges from 0 to 1.5 and denoted as E1 to E6. The compressive strength and modulus of rupture for the mixture are presented in Fig. 6(a) and Fig. 6(b). The optimum ratio of sand to epoxy is 1, thus contributing to a reduced cost of bonding material. The use of sand-modified epoxy for structural strengthening was also carried out. Reinforced concrete beams with 150mm x 150mm cross-section and length of one meter were casted and strengthened with CFRP. Modified epoxy increased the stiffness, ductility and toughness for beams strengthened with CFRP.



(a) (b)

Fig. 6 - a) The effect of fine-sand ratio on the adhesive cubes compressive strength, b) The effect of fine-sand ratio to the modulus of rupture of adhesive the prisms [61]

The use of sand washing waste as a filler in epoxy resin mortar has been proposed and researched by [65]. Samples were tested with different mixture proportion by weight percentages which include 10%, 15%, 20% and 25% of epoxy, sand washing waste (0%, 10% and 20%) and sand (60%, 65%, 70%, 75%, 80%, 85% and 90%). From the results, it can be concluded that the compressive strength, flexural strength and modulus of elasticity increased as the filler percentage increased by an average of 1.08-1.66, 1.18-1.66 and 1.07-1.54 times, respectively. On the other hand, for the bond strength test, most of the samples of epoxy content showed an increase in bond strength as the filler increased. Overall, sand washing waste can be used as a filler for epoxy resin mortar to obtain better mechanical properties (compressive strength, flexural strength and modulus of elasticity).

A study by [64] focused on the bond strength and characteristics of carbon nanotube modified epoxy. Two types of epoxy were used and characterised by high and low viscosity with good adhesion. A concrete block is bonded with a carbon and glass fibre-reinforced polymer sheet using modified epoxy and was tested under double shear pull test. The failure of neat epoxy is because of debonding. With carbon nanotubes, the cohesion failure was either at the concrete or the interface between the concrete and FRP, showing fairly good adhesion. Depending on epoxy and fibre types, improvements in bond strength and ultimate slippage has been reported using carbon nanotubes epoxy.

7. Future Alternatives for Epoxy Resin

The growth of epoxy resin market has increased as it becomes widely used in the applications of paints and coating, composites, adhesive, electronic encapsulation, grouts and mortar due to its high corrosion resistance and strong adhesion. Building and construction industries are the leading segments in the application of epoxy resin – they are expected to grow to USD 2696.2 million by 2025 due to the increasing infrastructure developments around the world [74]. The use of epoxy resin as an adhesive to FRP is widely used for repair and rehabilitation of deteriorated concrete structures, which will be of high demand in the future. FRP is one of the media that requires epoxy as an adhesive to attach the FRP to the concrete structures. FRP itself contains polymer matrices such as epoxy, vinyl ester, or polyester resin which can hold the fibre [7]. FRP depends on petrochemical-based resin which consequently compromises the sustainability of FRP [75]. The epoxy is produced from non-renewable sources, with some of the resin increasingly present in the ecosystem and can cause human toxicity. For example, the diglycidyl ether of Bisphenol A is derived from dangerous precursors such as Bisphenol A (also known as BPA) and Epychloridrine [76]. The use of such resins in FRP applications thus partially reduces the advantages of using environmentally-friendly reinforcing agents. In this regard, the resin side of composites should be addressed fairly if sustainable alternatives are to be truly developed for the composite industry.

Given that basic epoxy resin is derived from petroleum, a sustainable alternative to current epoxy for FRP strengthening application requires an understanding of all the materials involved as one system [74,78]. New epoxy should have good flexibility or stiffness, thermal expansion, water vapour diffusion, impact-resistant and more importantly, excellent adhesion to the concrete substrate to the extent that is comparable or even better than existing epoxy in the current market [79], [80]. Another potential improvement to epoxy is its durability. The modified epoxy should be able to deliver a lasting repair over its service life with no change in its inefficiency. Its resistance to UV exposure, resistance to alkaline conditions and physical properties that remain stable across a wide temperature range are some of the factors that could contribute to durability. Other important characteristics include low shrinkage, low permeability and strong adhesion. The durability is dependent on the proper bonding between the substrate (concrete) and the repair material (modified epoxy). Low permeability helps to protect against penetration of carbonation and chloride ions [79]. Given that the bulk of repairs are made to older concrete which does not experience drying shrinkage, the repair materials with low shrinkage can help to prevent loss of bond and cracking.

To achieve environmental sustainability, a renewable resource to substitute existing epoxy such as bio-based epoxy has been studied in recent years such as vegetable oil, sucrose and lignin [81]-[84]. These materials are highly available, safe and do not create harm to humans and animals. While many studies have been carried out on bio-resin as a composite matrix in material itself, general application, coating, manufacturing industry, aircraft, automotive and marine [73], [74], [85], [86], but to date, no application has been made on structural repair associated with FRP.

7. Conclusion

Epoxy has been used widely as a bonding material in the FRP strengthening system to repair deteriorated concrete structures. A drawback of this existing resin is the long curing time leading to an incomplete cross-linking reaction and consequently, affects the bond between FRP and the concrete substrate. In addition, the curing condition in the field is inconsistent which can affect the performance and durability on FRP repaired concrete structure, Besides, epoxy is very toxic and can cause health impacts to people who are living and working with them. Cured epoxy is brittle in characteristic, therefore modifications in epoxy are required to improve its toughening and ductility. The types of supplementary and replacement materials for epoxy, as well as composition, mixing method and testing are among the important criteria for epoxy modification. With various materials discussed in this paper, a modified epoxy has the same and even better properties compared to the existing epoxy. However, developing a novel epoxy with sustainable material that is viable for repair application is still an engineering pursuit.

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