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The degradation of glass fiber reinforced concrete and the potential for its improvement: A review

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ABSTRACT

One of the most widely used products is fiber-reinforced concrete. Glass fibers are used for reinforcement. The strength of the interaction between the fibers and the cement matrix is the most important factor. The degradation of glass fibers can occur for several reasons. One cause may be chemical processes, another cause may be the effect of hydration products formed at the interface between the fibers and the matrix. Another cause is minor defects in the manufacturing process, which can lead to fracture under static loading. The use of active mineral additives and special coatings can be used to improve the protection of the fiberglass surface and to prolong the durability of fiberglass reinforced concrete.

KEYWORDS;- *alkali-resistant glass fiber, glass fiber reinforced concrete, active mineral admixtures*

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I. INTRODUCTION

Fiber has been used as reinforcement since ancient times. In the 1950s, the concept of composite materials emerged, and one of the most interesting topics was fiber-reinforced concrete. Research into new fiber-reinforced concretes continues today [1]. The idea of using fibers as reinforcement is not new. Fiber-reinforced concrete is a composite material consisting of a cementitious matrix in which various fibers are dispersed. The two components have different chemical and physical properties, but when combined, the resulting composite has properties that cannot be achieved when they are used separately, while also retaining their unique properties [2].

Glass fiber reinforced material is quite versatile and is the most commonly used material in the concrete industry [3,4]. Glass fiber reinforced concrete is lightweight, strong, and resistant to environmental influences. It is environmentally friendly and poses no health risk, as fiberglass-reinforced concrete is an inorganic material. On the one hand, glass fiber is used to produce small, simple, and unsophisticated products [5]. On the other hand, leading international architects are using it for high-tech projects, meeting the highest demands in terms of structural complexity, size of elements, freedom of form, and impressive appearance, while maintaining excellent durability and quality. The use of fiberglass-reinforced concrete has given architects greater freedom of form combined with high environmental standards [6]. The use of fiberglass-reinforced concrete has been recommended in the construction industry to achieve sustainability [7].

This material has been used for more than 30 years for non-structural elements such as façade panels (about 80% of GRC production), sewerage pipe network systems, decorative items, and other products [6,8]. The influence of glass fiber on the various properties of concrete compared to plain concrete is presented. The mechanical advantages of GFRC over fiber-free concrete include an increase in tensile strength by up to 50%, allowing for the production of smaller cross-sections of elements. Additionally, the reduction of crack widths and the reduction of the total crack area by approximately 50% create GFRC elements with better protection against the ingress of harmful substances and adverse environmental conditions than conventional concrete [9].

Many researchers have investigated various properties of glass fiber reinforced concrete or cement mortars [3, 10-12]. In 1931, glass fibers (GF) were first used as reinforcement in mortar and concrete [9,13]. It should be mentioned that the type and geometry of the fibers affect the mechanical properties, fracture toughness, durability of the concrete, and pore structure [14]. Glass fiber reinforced concrete was commercially used by Pilkington Bros. in 1956. Since then, a significant amount of research work has been carried out over the last 60 years [15].

Glass fiber has excellent strength [16,17], thermal performance, durability, and interface attachment to the matrix [18]. Glass fibers improve the strength and stiffness of concrete. They are resistant to high temperatures, non-flammable, and resistant to corrosion, while also possessing high tensile strength and good thermal, acoustic, and electrical insulation properties. The bulk density of glass fibers is about 2.5-2.7 g/cm³. They are characterized by high tensile strength, ranging from 1200 to even 4800 MPa. Additionally, glass fibers have a much higher modulus of elasticity than synthetic fibers, though lower than steel and carbon fibers. They are resistant to high temperatures and start to soften around 700-900°C [19]. Glass fibers can be added to concrete for various functions, such as crack prevention [20, 21].

The chemical composition of E and S glass fibers is given in Table 1. As in conventional glass (window and container glass), the main constituent of all glass fibers is silica (SiO_2). Other oxides, such as B_2O_3 and Al_2O_3, are added to modify the structure of the SiO_2 network and improve the properties of the glass. Unlike conventional glass, the Na_2O and K_2O content of E and S glass fibers is relatively low, making them more resistant to alkaline corrosion, though not sufficiently so, and they also have higher surface resistance. The internal structure of glass fibers is a three-dimensional, long network of silicon, oxygen, and other atoms arranged in a random pattern. Thus, glass fibers are amorphous (non-crystalline) and isotropic (having uniform properties in all directions) [22].

Different types of glass are used in various industries:

E glass: used for reinforcement where high strength and high electrical resistivity are required, with a maximum alkali content of 1% by weight; **A glass**: used where strength and electrical resistivity requirements are lower than those of E glass; **C glass**: used where resistance to chemical attack and corrosion is required; **D glass**: used in the electromagnetic industry due to its low dielectric constant; **R and S glass**: used for high strength, structural reinforcement of textiles or composites, and special applications; **AR glass**: used as concrete reinforcement due to its high strength and resistance to alkali [22, 23].

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Type	SiO_2	Al_2O_3	CaO	MgO	B_2O_3	Na_2O
E-glass	54.5	14.5	17	4.5	8.5	0.5
S-glass	64	26	-	10	-	-

Table 1. Typical Composition of Glass Fibers (in (wt%) [22]

The first glass fibers used were not resistant to alkaline media, so conventional Portland cement could not be used as a bonding material. Initially, alkali-resistant glass (ARG) fibers were developed to reinforce cementitious matrices, which were widely used in construction and civil engineering. It is well known that the alkali resistance of glass is due to the high content of zirconium oxide (ZrO_2 > 15 wt% Figure 1) in the glass. It was only after the development of alkali-resistant glass fibers by English researchers and the introduction of high ZrO_2 content by Japanese manufacturers that the applications of glass-fiber-reinforced concrete increased. However, tensile strength has been a concern, and the impact strength of glass-fiber-reinforced cement products decreases due to possible corrosion reactions between the fiber surface and the cement matrix during aging [24,25].

The chemical composition of alkali-resistant glass commonly used for fiber production is given in Table 2.

Type	SiO_2	ZrO_2	Al_2O_3	CaO	Na_2O	K_2O
AR-glass	60 -62	16 -20	0.3 -0.8	0.5 -5.6	14 -17	0 -2

Table 2. Chemical composition of alkali-resistant glass [6]

Figure 1. Glass fibers: A) roving, B) chopped [37]

Yin [9, 26] concluded that the inclusion of glass fibers increased the adhesive strength between fibers and the matrix exposed to freeze-thaw cycles (in the presence of sodium chloride) compared to normal concrete. The maximum pull-out force was measured, and then the average adhesion strength was calculated. According to the research presented in [27], glass fiber (GF) filled the voids and reduced the permeability of the concrete. It was also found that a hybrid mix with glass fiber (GF) and polypropylene fiber (PPF) gave the best results in terms of void size. The smaller diameter PPF was able to bridge the smaller voids, while the thicker GF was able to bridge the larger voids. In summary, the addition of GF can produce a more brittle, dense, and impermeable material.

The fibrous material reinforces the FRC structure. It consists of thousands of small individual fibers that are randomly oriented and dispersed. Cement and alkali-resistant glass fibers are used in the production of glass-fiber reinforced concrete (GFRC), which allows the production of structural concrete products such as wall panels that are strong and lightweight. GFRC can also be used to create beautiful concrete products such as facade wall panels and concrete work surfaces [28]. Studies show that short fiber lengths primarily control the propagation of micro-cracks and improve ultimate strength, while long fiber lengths contain macro-cracks and improve post-cracking deformation of concrete [29]. Specimens with short fiber lengths (3 mm and 6 mm) have a higher tensile strength than specimens with long fiber lengths (12 mm and 20 mm). The specimens with long fiber lengths (12 mm and 20 mm) had a higher strain than the specimens with short fiber lengths (3 mm and 6 mm). Fiber dispersion, fiber orientation, and embedded fiber length affect the strength and deformation of GFRC specimens [29].

II. CAUSES OF DESTRUCTION

In glass fiber reinforced concrete, a very good distribution of glass fibers is important. The addition of fibers reduces the size of voids as well as the width of shrinkage cracks in concrete (Figure 2) [27]. Glass fiber reinforced concrete (GFRC) can experience strength losses when exposed to a humid environment [30, 31]. This reduction in the properties of GFRC is influenced by two main phenomena. The first is alkali attack on the glass fiber by hydroxyl ions, which form due to hydration of the cement matrix. In this case, chemical changes occur, leading to a decrease in strength. The second phenomenon involves changes at the fiber-matrix interface, which densify the matrix around the glass fiber. The final phenomenon is related to the deposition of hydration products (mainly $Ca(OH)_2$) inside the glass fiber, resulting in improved adhesion of the individual fibers [32,33].

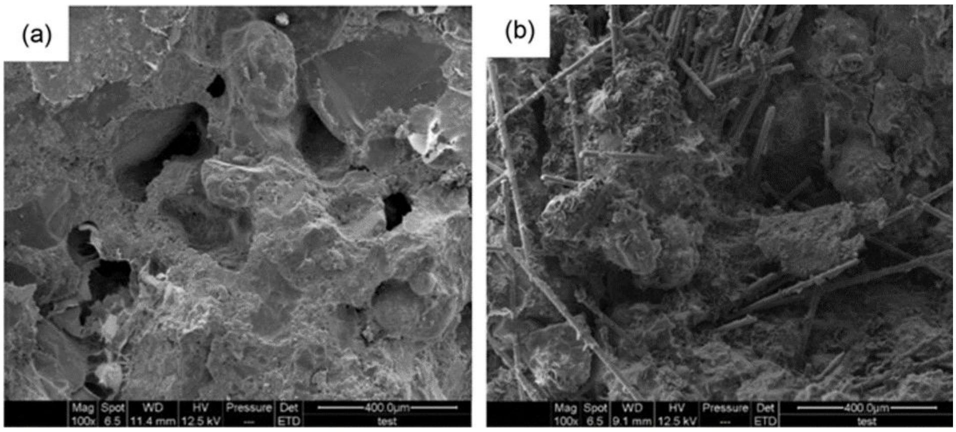

Figure 2. SEM image of OPC concrete and fiber reinforced concrete; (a) SEM image of OPC concrete, (b) SEM image of GFRC [27]

To improve the strength and flexibility of GFRC over time, enhancements have been made in two directions: the use of alkaline-resistant (AR) fibers with a high zirconium oxide content and the modification of the Portland cement-based matrix with various additives such as silica micro-particles, slag, fly ash, metakaolin, acrylic polymer, or sulfoaluminate. The purpose of these additives is to reduce alkalinity and remove CH (calcium hydroxide) around the filaments.

The exact mechanisms of the degradation process are still debated, but it is generally accepted that it involves corrosion of the glass fibers caused by hydroxyl ions in the pore solution [34, 35], and significant calcium hydroxide (CH) deposits between and around the fibers, leading to a loss of flexibility [36].

When studying the behavior of concrete after cracking, it is important to analyze the fracture energy (Gf), which is the energy required to fracture the test specimen, and the fracture toughness (KIC), defined as the ability of the test specimen to resist brittle decay or crack expansion [17]. The fracture energy increased by 40%, 46%, and 64% for samples with 0.50%, 0.75%, and 1.00% glass fiber content, respectively, compared to plain concrete

[17]. Additionally, the fracture toughness was approximately 1.37 and 1.75 times higher for GFRC with 0.50% and 1.00% glass fiber content, respectively, than for PC (plain concrete). The finding that glass fiber has a remarkably positive effect on the ductility of concrete [37].

Calcium sulfoaluminate (CSA) cement is a low-carbon cement with high early strength, good permeability, and improved durability [38]. Its pore solution is less alkaline than that of Portland cement, and calcium hydroxide (CH) is absent in the hydration products [39]. Therefore, it has the potential to produce high-performance GFRCs with improved durability properties. Many studies have been carried out to investigate the microstructural properties and durability of CSA cement modified GFRC (CSA/GFRC).

Yin [40] concluded that the use of glass fibers increased the adhesive strength between the fibers and the matrix subjected to chloride freeze-thaw cycles as well as chloride dry-wet cycles compared to conventional concrete. The maximum pull-out force was measured, and then the average adhesion strength was calculated.

Several main mechanisms of glass fiber degradation have been proposed in the literature: in the first case, the Si-O-Si glass network is affected by hydroxyl ions; in the second case, hydration products affect the filament/concrete and increase the transversal shear of the filament; and, in the third case, fracture under static loading due to minor defects in manufacturing processes.

In the second case, Bentur [31,41,47] assumes that embrittlement due to the growth of hydration products around the glass filaments occurs at the beginning of the cure. In this case, it is the loss of the filament strength rather than the chemical effect that is important.

In the third case, the fracture may be caused by minor defects in the glass [42,43]. There will be stress concentrations near the defects that have occurred during the manufacturing process when the glass is subjected to a continuous load. The growth of the damage will then increase and eventually lead to a delayed fracture (also known as static fatigue). This delayed fracture depends on the mechanical load and the environmental conditions. A third case has been studied extensively by the researcher Purnell [35,44].

To make glass fittings more stable in alkaline environments, alkaline-resistant glass (AR-glass) contains 15-20% zirconium. This does not solve the problem of degradation definitively, although the durability of the reinforcement may be improved [45- 47].

The properties of hardened fiberglass concrete depend on many factors. The main component of fiberglass-reinforced concrete is the cementitious matrix, whose properties change over time. Time is one of the primary factors influencing the properties of hardened fiberglass-reinforced concrete. Currently, accelerated aging tests are used, where long-term natural exposure is replaced by a shorter exposure of the same magnitude in more aggressive warm or hot solutions.

Initial corrosion is caused by sodium leaching and disruption of the Si-O-Si network. The stable ZrO_2 surface slows down the diffusion of other ions. In this case, this area is called the inner layer.

In addition, the recondensation of silanol groups in a weak surface layer with a destroyed glass network can also slow down the entry of OH into the glass structure [48]. The process is controlled by diffusion. This area is called the outer layer.

III. ADDITIVES AND INNOVATIONS FOR THE IMPROVEMENT OF FIBERGLASS

Use of micro fillers and additives

Active mineral admixtures are often used as concrete components and are characterized by pozzolanic reactions with portlandite phases. These reactions reduce the concentration of $Ca(OH)_2$ crystals, which, as they grow, damage the fiber surface and degrade the long-term mechanical properties of the composite. Due to their extreme fineness, these additives compact the cementitious stone and improve the bond between the cement and aggregate particles, thus enhancing the mechanical properties of the cementitious matrix. This can be explained by their effect on the thickness of the intermediate phase between the aggregate particles and the cement paste, as well as on the orientation of the crystals of the CH phase. The reduction of pure silica to silicon at temperatures up to 2000 ^{0} C produces SiO_2 vapor, which oxidizes and condenses into small particles of non-crystalline silica in the low-temperature range [49].

This material is also known as microsilica. It is about 100 times smaller than the average cement particle. Until the 1960s, almost all microsilica was released into the atmosphere. Environmental requirements to capture and bury silica have made it economically important to use, especially in the production of high-quality concrete. Due to its extreme fineness and high silica content, microsilica is added to Portland cement concrete to improve its properties, particularly its compressive strength, adhesive strength, and abrasion resistance. Attempts have been made to modify the matrix of fiberglass-reinforced concrete by replacing up to 10% of the cement with microsilica. This modified matrix results in higher strength and durability of the composite. A study of a glass-fiber reinforced (fiberglass) element in the presence of microsilica showed that the latter prevented the crystallization of $CaCO_3$ between the fibers. Silica improved the net increase in compressive, splitting tensile, and flexural strength due to the incorporation of the fiber. This was attributed to the strengthening and stiffening of the cement paste, which improved the bond between the matrix and the fibers [50].

Amin et al. [51,52] concluded that the use of silicon microspheres (SF) significantly improved the microstructure as the silicon microspheres reacted rapidly with crystalline $Ca(OH)_2$ to form C-S-H gels. This resulted in a reduction in the number and size of $Ca(OH)_2$ crystals and a denser microstructure [53]. The use of SF as a cement replacement can reduce the porosity, the CH content, and the calcium-to-silica ratio, thus improving the performance of the concrete microstructure [54]. The pozzolanic reactions between SiO_2 and calcium hydroxide lead to the formation of additional calcium hydrosilicates (C-S-H), which grow into voids around the hydrated cement particles.

Increasing the density of the interfacial zone and improving the bond between cement and aggregate particles by using a SiO_2 micro-particle additive has been repeatedly demonstrated in scientific studies [55]. The use of SiO_2 microparticle additive in concrete significantly reduces the diffusion of chloride ions. This can be attributed to the fact that the hydration of the cement is accompanied by the splitting of large pores into smaller ones through pozzolanic reactions. SiO_2 microparticle improves the long-term resistance of concrete to alkali corrosion but increases the depth of carbonation [56]. Studies on silica-fiber reinforced composite by Bartos [57] showed that the addition of silica microparticle improves the long-term mechanical properties of this material.

Metakaolin particles are smaller than cement but larger than microsilica. Advantages include increased compressive and flexural strength, reduced permeability, increased resistance to chemical attack, enhanced durability, decreased exposure to alkaline environments, improved concrete workability, and reduced particle shrinkage and compaction. This aggregate is used to produce high-quality, high-strength lightweight concrete, glass fiber reinforced concrete, mortar, and plaster.

Metakaolin (MK), or $Al_2Si_2O_7$, is produced by calcination of kaolinite clay at 700-800°C. Dehydroxylation occurs after kaolinite has been exposed to high temperatures for a specific period. The pozzolanic activity of metakaolin (MK) in cementitious materials has been extensively studied, characterized by the formation of new calcium hydrosilicate phases (C_2ASH_8, C_4AH_{13}, and C_3AH_6). Researchers' studies on the substitution of metakaolin (10% to 25%) for part of cement have shown that this additive improves the mechanical properties and durability of cementitious materials [58, 59].

The main challenge in using metakaolin in cementitious mixtures is the increased water demand caused by the high specific surface area of the particles (17 m^2/g) and their irregular morphological structure (metakaolin particles are plate-like) [60 ,61]. In studies on glass fiber-reinforced composites, Brandt [62] found that control samples without additives lost 85% of their initial flexural strength after soaking in water at 50°C for 84 days. However, specimens in which 40% of the cement was replaced by a metakaolin additive showed almost no loss of the original mechanical properties under similar conditions. SEM-EDX analysis revealed no inclusions of portlandite crystals between the glass fibers, which explained the positive effect of metakaolin on the durability of the glass fiber-reinforced composite.

Marikunte et al. [63] also reported an improvement in the durability of GFRC exposed to accelerated aging with the addition of metakaolin. The addition of 10% metakaolin resulted in a 10% decrease in flexural strength after aging, whereas without metakaolin the loss was 50%.

Similar behavior in the aging of GFRC with metakaolin and the addition of metakaolin and acrylic polymers has also been reported by other researchers [64]. Several studies [65, 66] have reported an improvement in the durability of GFRC with fly ash partially replacing cement. Zhang and co-authors [65] observed that when up to 60% of the cement was replaced by fly ash in the samples, the flexural strength improved with time. Ageing tests were carried out in a hot bath at 80 °C. Peled et al [67] also highlighted the improvement of GFRC with the addition of fly ash and the ageing process. GFRC with fly ash retained its flexural strength when ageing was carried out in a hot bath at 80 °C. GFRC without fly ash showed a 40% reduction in flexural strength.

Fiber surface improvement

The concept of polymeric retrofitting of glass fiber reinforced concrete was first introduced by Forton BV in 1979 at GRCA London. In 1983, a report was published on the usefulness of adding 5% by volume of polymer solids to a Portland cement matrix reinforced with alkali-resistant glass fiber. To explore an alternative to the then-required 7-day wet curing regime to achieve maximum matrix strength, further investigation was conducted into the full properties of this mixture. A sand-cement mix, with a minimum of 6-7% polymer solids added to the Forton polymer cement paste and reinforced with alkali-resistant glass fiber, produces the required composite [68].

Direct tests on ten glass fibers showed an increase in tensile strength, attributed to the treatment of defects in the organic tape coating and the higher fracture energy of the modified fibers [69]. Further durability tests of silane coatings demonstrated a significant reduction in NaOH exposure at ambient and higher temperatures.

Among the polymeric coatings tested, styrene-butadiene dispersions were found to be the most resistant [70]. Recent studies indicate that the mechanical properties and durability of ARG fibers reinforced with carbon nanotube-reinforced nano-clay coatings have significantly improved. Recent research has also shown that carbon nanotube-modified coatings have a positive effect on fiber properties The tensile strength of such fibers was found to be 50% higher after seven days of treatment with a 5 wt.% NaOH aqueous solution under ambient conditions [71]. The researchers used a "surface defects healed with super materials" approach (Figure 3) using traditional alkali-resistant glass fibers (ARG).

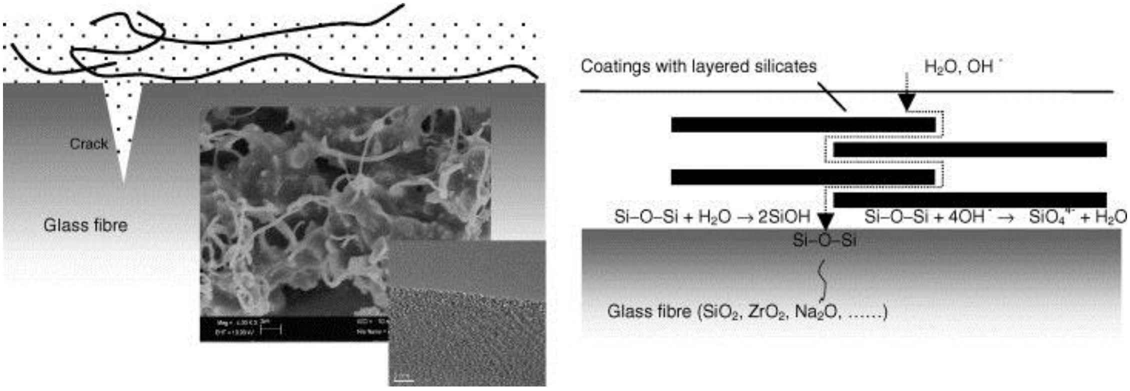

Figure 3. Schematics of nanostructured coatings with nanotube/layered silicate polymer network on glass fiber surface to enhance flaw healing effect and corrosion resistance [71]

The modification of the fiber surface structure with various impregnates and coatings is a common method used by fiber producers to improve the long-term performance of fibers in cementitious systems and to increase the resistance of fibers to various mechanical stresses during transport and mixing. The latest generation of ZrO_2-containing glass fibers is coated with organic silane polymer coatings several nanometers thick.

Various researchers have studied the effect of these coatings on the properties of fiberglass. Silane polymers form spatial networks on the surface of the fiber, which increase the adhesion between the fiber and the matrix, protect the fiber from the effects of water, and improve the mechanical properties of the fiber. In direct tensile tests on fibers, silane polymers have been shown to cover damage on the fiber surface and increase the strain energy of the fibers [69, 70].

The use of acrylic polymers as concrete admixtures was introduced in the 1980s, increasing the early strength of GRC slabs and allowing the elimination of long-term wet curing in production. Nowadays, ether-based polycarboxylates or superplasticizers allow the use of lower W/C ratios to achieve adequate workability of concrete. Another application of acrylic polymer is in the field of improving the durability of GRC. The acrylic polymer slurry is typically added at 7-10% of the cement weight. Ball H. reviewed durability data obtained from GRC coupons naturally retained for 19 years in wet-dry climates. There was a decrease in flexibility and ductility of up to 40% in composites made without polymer after 19 years. On the other hand, GRC with 7% acrylic polymer retained most of its stiffness over the entire period. Studies have also shown that wet-dry accelerated ageing is more consistent with real weather data for polymer-modified composites [68].

Qian X. investigated the effect of PVA (polyvinyl alcohol) powder on the durability of GRC. The immersion of 6 mm thick samples in 20 °C water for ageing was examined. Direct tensile tests showed that PVA powder increases flexibility and tensile strength during ageing. Microstructural analysis explained the positive effect of PVA powder, which created protective thin polymer films on the surface of the glass fiber and prevented the accumulation of brittle calcium hydroxide in the interfacial zone [72].

Due to surface defects in brittle materials, the actual tensile strength is much lower than the maximum theoretical strength. Coatings can be used to 'cure' surface defects and modify surface properties. The process involves the application of a nanometre-scale hybrid coating layer consisting of a styrene-butadiene copolymer with single or multi-walled carbon nanotubes (SWCNT, MWCNT) and/or nanosheets as a mechanical enhancement and environmental barrier layer on alkali-resistant glass (ARG) and E-glass fiber. Data show that nanostructured and functionalized conventional glass fibers exhibit significantly improved mechanical properties and resistance to environmental corrosion. The tensile and flexural strength of the degraded glass fiber increases enormously when a small fraction of the nanotubes is broken. There is no variation in the strength of the nanoclay-coated fiber when subjected to alkaline treatment. In addition, nanocomposite coatings increase the adhesion of the fiber/matrix interface [71].

IV. CONCLUSION

The optimum fiber dosage is influenced by the ratio of water to binder. This is because an increase in the water/binder ratio results in a sudden increase in pore size and volume in the concrete matrix. The effect of the water/binder ratio should be considered in relation to the effect of fibers on mechanical strength or microstructure. Higher water/binder ratios may compromise the structural integrity of the concrete and therefore a balance between these factors is necessary for optimum performance.

The degradation of glass fiber reinforced concrete is related to the compressive effects of mechanical loading and environmental conditions. The strength of the interaction between the fibers and the cement matrix is the most important factor. The coupled action of mechanical load and environmental conditions accelerates the environmental degradation process.

Although the concrete is made using alkali-resistant AR-type glass fibers, they still lose much of their original flexural and tensile strength under natural conditions. Different main damage mechanisms are recognized for glass fiber reinforced cementitious matrices:

1) Hydration products, in particular hydroxyl ions, chemically attack the glass fiber, breaking the Si-O-Si glass network. This chemical degradation weakens the fibers and compromises the integrity of the concrete.

2) Hydration products are formed at the interface between the fibers and the concrete, which can disrupt the bond between the fibers and the cement matrix.

3) Minor defects in the manufacturing process can cause fracture under static loading. These fractures propagate over time and further weaken the concrete.

The strength of the interface transition zone between the fiber and the cement matrix is mainly determined by chemical interactions. The main cause of the loss of strength is the appearance and growth of hydration products on the surface of the glass fiber, a process known as static fatigue, which is not yet fully understood by scientists.

The strength of the interface transition zone between the fiber and the cement matrix is mainly determined by chemical interactions. The main cause of loss of strength is the formation and growth of hydration products on the surface of the glass fibers. This process is known as static fatigue and is not yet fully understood by scientists.

Active micro-fillers can improve the chemical stability of the cement matrix and reduce the destruction rate of glass fibers. Reducing the portlandite content of the cement matrix can reduce the chemical attack on the glass fibers. The use of acrylics and PVA polymers can increase the flexibility and strength of the concrete and improve its resistance to cracking and spalling. Nano-coatings can protect fibers from chemical attack by modifying the fiber surface. Taking these factors into account, the durability and performance of glass fiber reinforced concrete can be significantly improved, resulting in more durable and resilient building materials.

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