Interface Strengthening Mechanisms in Polymeric Fiber Reinforced Cementitious Composites

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ABSTRACT

This paper reviews several techniques reported in the literature and their effectiveness for improving bond properties of polymeric fibers in cementitious matrices, with special focus on some recent results of a plasma treatment process. Some discussions of the unique bonding behavior of polymeric fiber in a cementitious matrix is included.

INTRODUCTION

The interfacial bond strength governs many important composite properties. These include all composite strengths, fracture energy, ductility, and energy absorption capacities [1, 2]. In short, all composite properties beyond the linear elastic range of deformation may be expected to activate the failure of the fiber/matrix interface. Hence techniques to enhance interfacial bond strength has been a major research activity.

One of the early pioneers in devising practical and effective techniques in enhancing the interface bond of polypropylene fiber reinforced cementitious composites is Professor Herbert Krenchel. This paper is dedicated to Professor Krenchel, in honor of his seventieth anniversary and his major contributions to the science and technology of fiber reinforced cement based composites, over a career span of almost fifty years.

CHARACTERISTICS OF POLYMER FIBER/CEMENT MATRIX INTERFACE

A variety of polymer fibers, including polypropylene, polyethylene, polyester, Nylon, Aramid, acrylic and PVA have been used in cement and concrete reinforcements. While polymer fibers have certain advantage over other fiber types, they also have their limitations. One of these limitations is the poor adhesion and wettability to a cementitious matrix, as a result of their chemical inertness and low surface energy [3]. Typical bond strength for polymeric fibers without any surface treatment appears to be under 1 MPa, in comparison to several MPa for steel fibers, glass, or carbon fibers [4, 5, 6, 7, 8, 9]. Consequently, it is usually necessary to develop special techniques to enhance the interfacial bond of polymer fibers in order to assure advantageous composite properties.

Apart from weak interfacial bond, another unique characteristics of polymer fibers in cementitious matrix is the abrasion effect as the fiber is drawn out from the matrix after full debonding [10]. The abrasion effect is a result of fiber surface damage due to the low strength of most polymeric fibers in the lateral direction. Interestingly, the fiber surface abrasion leads to an improvement in the pull-out load and enhance energy absorption in the process. Figure 1 shows the abraded surface of three segments of a Nylon fiber after being pulled from a cement matrix. The embedded end which went through the largest amount of slippage s (equal to the embedded length) suffers the largest amount of abrasion. It can be expected that the stripped fibrils cause the fiber to jam, leading to the increase in pull-out load. Figure 2 shows typical pull-out load vs. separation curves for Nylon and Polypropylene fibers. Unlike steel fibers, the pull-out curve after full debonding shows a hardening rather than a softening behavior.
The slip hardening process has been modeled by Wang et al. [11] who found that a polynomial representation of the bond strength to local slip displacement provides a good fit to experimental load-displacement curves:

\[ \tau = \tau_0 + as + bs^2 \]  

(1)

The initial bond strength \( \tau_0 \) and coefficients \( a \) and \( b \) are expected to depend on the specific fiber type and manufacturing process. In addition, the nature of the surrounding matrix material, such as the amount of surface contact with the fiber surface, can also be expected to alter the extent of surface abrasion and therefore the magnitude of \( a \) and \( b \). As a result of this slip-dependence, the pull-out branch of the load-displacement curve becomes highly non-linear. For small amount of abrasion, the pull-out curve descends but with a concave downwards shape. For more severe abrasion, the pull-out curve actually rises with the amount of slip. This hardening behavior appears to be a hallmark of surface abrasion in polymer fibers pulled out from a cementitious matrix.

**STRENGTHENING MECHANISMS**

A variety of interface bond strengthening mechanisms have been proposed and utilized. Some of these make use of macroscopic processes such as fiber deformation. Others utilize microscope changes such as fiber surface and/or transition zone modifications. Because of
the weak adhesion of polymer fibers to cementitious matrix, it is unlikely that techniques such as transition zone densification would be effective. This concept is schematically illustrated in Figure 3. Based on microhardness test, Wei et al [12] determined that the hardness reaches a minimum in a transition zone with a thickness of approximately 50 \mu m. If it is assumed that the strength scales with this microhardness, then it can be inferred that failure during fiber debonding or pull-out can occur in this weak transition zone. This is the case if the adhesion strength (e.g. point A) is higher than the minimum strength (point C) in the transition zone. However, if the adhesion strength is lower (e.g. point B), then failure can occur directly along the fiber surface. This latter case appears to describe most polymeric fibers in a cementitious matrix. For such material system, techniques aimed at strengthening the transition zone, such as by reducing its porosity through use of microfillers [e.g. 13], tends not to be effective. Figure 4 shows the bond strength from pull-out tests of two types of polyethylene fibers from two matrix types. The matrix with silica fume has a densified transition zone. Yet no improvement on the bond strength can be observed. However, if the use of microfillers lead also to increasing the contact surface of the transition zone material with the fiber surface, then the adhesion strength may increase, resulting in an increase in interface bond strength.

![Diagram](image_url)

Figure 3: Schematic Illustration of Adhesion Strength, Strength in Transition Zone and in Bulk Matrix

**Deformed Fibers**

Most fiber deformation processes result in increase in surface area of contact with the cement matrix per unit fiber length. Mechanical interactions between fiber and matrix may occur on the mm scale. Hence the effective bond property must be interpreted in this broader context.

One of the earliest concepts in modifying the interfacial bond between polymeric fiber and cementitious matrix by means of fiber fibrillation was proposed by Krenchel and co-workers [14, 15, 16]. The manufacturing process, controlled by mechanically splitting the extruded polypropylene tape, achieves two aims: It increases the specific surface area of contact and enhances mechanical bond to the cement matrix by means of fibrillation, and it also improves the fiber modulus by molecular chain alignment. Figure 5 shows a fibrillated fiber. Krenchel [16] indicated that a factor of 3 or more can be achieved in bond
strength improvement when the fiber has been fibrillated and also surface treated by chemical additives.

![Graph showing bond strength comparison between Spectra fiber and Snia fiber](image1)

Plain matrix: w/c=0.40
SF matrix: Silica fume/c=0.20

**Figure 4:** Bond Strength of Two Polyethylene Fiber Types Showing No Effect From Transition Zone Densification.

![Photograph of fibrillated polypropylene fiber](image2)

**Figure 5:** Photograph of Fibrillated Polypropylene Fiber
A crimping process, by running a fiber under a loose set of gears, can also be utilized to deform polymeric fibers. Li et al [17] showed that the pull-out process can be significantly altered by mechanically crimping a Nylon fiber (Figure 6a). Significant improvement of the effective bond strength can be achieved (Figure 6b). The fluorocarbon is added so that the fiber can be pulled out instead of breaking due to the mechanical locking effect.

Figure 6a: Photograph of a Crimped Nylon Fiber [17].

Figure 6b: Pull-out Test Curves For Nylon Fiber With Different Surface Treatments: 1, No Treatment; 2, Coated With Fluorocarbon Dry Lubricant; 3, Crimped And Coated With Fluorocarbon Dry Lubricant [17]

Another technique of improving mechanical anchorage and therefore bonding to a cementitious matrix is by twisting the fibers. Naaman et al [18] reported that a factor of 7 in interfacial bond can be achieved by this method. Figure 7a shows a photograph of two ply twisted polypropylene fibers. The effect of fiber twisting on the pull-out curve can be observed in Figure 7b.

Surface Modification
Cold gas plasma has been in commercial use to enhance adhesion between plastic parts. This technology was recently applied to alter interfacial bond characteristics in polymeric fibers/cementitious matrix systems [19]. In the presence of a gas plasma, hydrogen atoms
are removed from the polymer backbone and replaced by polar groups. The specific polar group is related to the type of gas used. The presence of polar functional chemical groups on the fiber surface enhances reactivity and thus improve the adhesion between fiber and cement. In addition, wettability of the fiber surface by cement can be enhanced, leading to increased contact between fiber and the surrounding cementitious matrix. As a result, it may be expected that the fiber surface abrasion effect described earlier becomes more significant.

Figure 7a: Photograph of Twisted Polypropylene Fibers.

Figure 7b: Pull-out Test Curves For Twisted Polypropylene Fibers [18].

Figure 8 shows the pull-out test result of a polyethylene fiber treated with an Argon plasma gas, with a flow rate of 40 ml/min, 100 Watt. of power, and 5 minutes of treatment
duration. A cement paste with w/c ratio of 0.4 serves as the matrix material. For comparison, pull-out test result of an identical fiber, untreated, is also shown. In both cases, the pull-out curves show the characteristics of that of a polymeric fiber. A steep rise (appearing as a linear line on this scale) associated with interface debonding is followed by the 'hardening' behavior of surface abrasion. Surface abrasion was observed for both fibers after pull-out.

![Pull-out Load vs. Pull-out Distance](image)

Figure 8: Comparison of Pull-Out Test Results of Treated and Untreated Polyethylene Fiber

It is clear that the load at the end of debonding is almost doubled, reflecting a bond strength increase from 0.55 MPa to 1.06 MPa due to plasma treatment. Indeed, for the case of the plasma-treated fiber, the load drop at the end of debonding indicates that the adhesion bond seems to have exceeded the initial frictional bond. Thereafter, the effect of abrasion on the plasma-treated fiber appears to be more significant than that on the untreated fiber, indicated by the more rapid rise in load with pull-out distance.

Various polar groups have been found present after plasma treatment [20]. The actual groups depend on the specific gas used. Argon plasmas introduce oxygen groups (carbonyls, carboxyls, and hydroxyls), whereas nitrogen and oxygen plasmas introduce oxygen groups and nitrogen groups (amides, imides, and nitriles) onto polyethylene surface. Table 1 summarizes the relative elemental compositions obtained for each of the gases and controls from ESCA measurements. Similar results are also obtained from our own measurements.

The enhanced effect of surface abrasion due to plasma treatment is even stronger in a polypropylene fiber treated under the same conditions as for the polyethylene fiber. For the polypropylene fiber, the amount of slip-hardening is significantly enhanced over that of the untreated fiber (Figure 9). Microstructural observations under an Environmental Scanning Electron Microscope (ESEM) reveal different interfacial microstructure on the contact surface after the fibers are carefully peeled off. In Figure 10a, the matrix surface in contact with untreated fiber shows more pores and less compact structure. On the other hand, the matrix surface in contact with plasma treated fiber (Figure 10b) is smooth and well hydrated. The less compact interfacial microstructure may indicate low wettability of the pp-fiber to cement solution. Plasma treatment may have improved the wettability of the pp-fiber to cement solution, which can be checked with a contact angle measurement device. For the pp-fiber, the significantly enhanced abrasion effect may be associated with
the alteration of the physical nature of the contact surface. As can be seen in Figure 9, the initial bond value $\tau_o$ appears to be only marginally improved (about 20%). Plasma treatment seems to have a stronger effect on the coefficients $a$ and $b$ in Eqn. (1) for the pp-fibers.

Table 1: Elemental compositions obtained from ESCA measurements of plasma-treated and untreated polyethylene fibers [20].

<table>
<thead>
<tr>
<th>Gas</th>
<th>Elemental composition</th>
<th>Composition ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Si (%)</td>
<td>C (%)</td>
</tr>
<tr>
<td>Control</td>
<td>5.33</td>
<td>86.41</td>
</tr>
<tr>
<td>Argon</td>
<td>2.63</td>
<td>78.43</td>
</tr>
<tr>
<td>Nitrogen</td>
<td>4.41</td>
<td>78.49</td>
</tr>
<tr>
<td>Carbon dioxide</td>
<td>3.24</td>
<td>77.14</td>
</tr>
</tbody>
</table>

Figure 9: Comparison of Pull-out Curves of Treated and Untreated pp-Fibers.

CONCLUSIONS AND FURTHER DISCUSSIONS

Polymer fibers have the unique characteristics of poor interfacial bond strength with cementitious matrices, and weak lateral strength resulting in surface abrasion. The poor bonding characteristics is a severe limitation to the effective use of polymer fibers in high performance cement based composites. The need for enhancing interface bond properties is especially important for higher modulus higher strength polymeric fibers, which are being introduced commercially at increasingly attractive prices. For these fibers, better interfacial bond strength is necessary to exploit the improved fiber property in the performance of the composite.
Figure 10: Fiber/Matrix Contact Surface for (a) Untreated pp-Fiber Showing Less Compact Interface Microstructure, and (b) Treated pp-Fiber Showing Smooth and Dense Microstructure.

Two classes of techniques are described in this paper for enhancing bonding of polymeric fibers to a cementitious matrix. The first class involves fiber deformation which includes fibrillation, crimping, and twisting. The second class involves fiber surface modification. A plasma treatment process is described in this paper. It is shown that for polyethylene fiber, interfacial bond strength can be enhanced by 100% over untreated fiber. For polypropylene fiber, the effect of plasma treatment appears to result in significant change in the pull-out characteristics associated with the surface abrasion of the fiber. The two classes of techniques are not exclusive of one another. It is likely that a combination of mechanical deformation technique and fiber surface treatment may produce synergistic results.

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