Interfacial Characteristics of Engineered Pulp Fibre Reinforced Concretes Containing Various Pozzolans

Emad Booya1*, Adeyemi Adesina2, Karla Gorospe3 and Sreekanta Das4

1Department of Civil and Environmental Engineering, University of Windsor, Canada
E-mail: booya@uwindsor.ca
2Department of Civil and Environmental Engineering, University of Windsor, Canada
E-mail: adesina1@uwindsor.ca
3Department of Civil and Environmental Engineering, University of Windsor, Canada
E-mail: gorospek@uwindsor.ca
4Department of Civil and Environmental Engineering, University of Windsor, Canada
E-mail: sdas@uwindsor.ca
* Corresponding author: Department of Civil and Environmental Engineering, University of Windsor, Canada. Tel: (1) (519) 992 1086

Abstract. Increasing durability threats on existing structures made with cementitious composites has prompted a need to find ways to improve the durability of new structures. One of the effective ways to improve the durability properties of cementitious composites is by the incorporation of fibres. A type of modified Kraft pulp fibres was employed in this study to improve the durability properties of cementitious composites. A microstructural investigation was also conducted to study the fibre morphology and its effect on the performance of the cementitious composites. Moreover, the durability tests in terms of water absorption and rapid chloride penetration were carried out. Overall results showed higher improvement of the durability of the cementitious composites with composites incorporated metakaolin, silica fume, and slag. This was further confirmed by microstructural analysis showing that these supplementary cementitious materials densified the cementitious matrix.

Keywords: Engineered Fibres; Pulp; SEM; Strength; Durability.

1. Introduction

More sustainability awareness in the construction industry in recent times has led to the use of various environmentally friendly materials as components in concrete. Of such eco-friendly materials that can be incorporated into concrete to improve the performance are natural fibres such as cellulose fibre. These fibres are sustainable alternatives to the conventional petrochemical-based and synthetic fibres as they
exist naturally in the environment and does not require and complex production process (Ardanuy et al. 2015). Similarly, the natural fibres are available worldwide, renewable and do not pose any hazardous threat to the environment. Hence, these materials have several promising potentials to be used in producing environmentally friendly concrete.

The penetration of deleterious materials into concrete is known to pose a durability threat and a corresponding degradation of mechanical performance (Booya et al. 2018 and Booya et al. 2020). These deleterious materials get into concrete can occur by diffusion or by using water as a pathway. On the other hand, concrete could also experience higher shrinkage, low ductility, etc. which might hinder the performance of the composite. The incorporation of cellulose fibres in concrete can be used to improve some of the detrimental properties by enhancing the microstructural permeable pores. The study by Banthia et al. (2012) showed that the use of virgin, fully purified softwood cellulose fibre at dosages of 0.1% and 0.3% by volume of the mixture resulted in a reduction in the pore volumes of cement. In addition, the cryoporometry and mercury intrusion porosimetry (MIP) observations showed that higher dosages of cellulose fibre in cement paste decreased the pore size to approximately 5 nm from approximately 70 nm. Hence, it was concluded from the study that cellulose fibres can the incorporation of cellulose fibres can be used to mitigate the penetration of deleterious materials into the concrete. In a similar study, Sappakittipakorn and Banthia (2012) evaluated the effect of virgin and fully purified softwood cellulose fibre with a length of 2.3 mm on the chloride transport characteristics of fibre reinforced concretes. The findings from the study showed a reduction in the effective chloride (free) diffusion coefficients can be achieved due to the ability of the cellulose fibres to chemically bind the chlorides. Another study carried out at the University of Florida also explored the effect of cellulose fibres on the permeability and durability properties of concrete (Roque 2009). In the study, an unmodified cellulose fibre with a length and aspect ratio of 2.1 mm and 117, respectively was used. Results from the study showed that the incorporation of the cellulose fibres also resulted in an improvement in the durability performance of concrete.

Despite the promising advantage of incorporation of cellulose fibres as evident in the laboratory results, the use of these fibres on large scale is limited as a result of some major challenges. One of the challenges with the use of cellulose fibres in concrete is the high alkalinity of the concrete pore solution which might result in degradation of the fibres in the long-term (Savastano 2009). Another major challenge is the possible migration of hydration products into the lumen of the fibres which can result in the mineralization of the fibre and consequently weaken the fibres.

The degradation of natural fibres in concrete can be prevented or minimized by modifying the fibre or modifying the composition of the concrete (Mohr et al. 2007, Toledo Filho 2003, Mohr et al. 2006). The fibres can be modified chemically or my physical methods (Mohr et al. 2007). These methods have been found to be effective in enhancing the bonding of fibre in the composite and reducing the moisture transfer around and through the fibres. Claramunt et al. (2011) studied the effect of hornificated Kraft pulp fibres on the durability properties of cement mortar composites. It was concluded from the study that the hornification of the fibres enhanced the performance of the composites. On the other hand, composites made with Kraft pulp without any modification exhibited lower performance. Several studies have shown that these natural fibres can be modified by processes such as fibre beating, initial drying age, bleaching, and pulp treatments (thermomechanical or Kraft) (Mohr et al. 2006 and Mohr et al. 2005). It was also concluded from these studies that the drying state and beating did not result in any significant influence on the mechanical performance of the composites when subjected to wet/dry cycles.

The performance of concrete reinforced with natural fibres can also be improved with the use of materials such as supplementary cementitious materials (SCMs). Some studies have explored the use of SCMs as partial replacement of cement in order to refine the pore structure and reduce the alkalinity of the cement matrix. Mohr et al. (2005) evaluated the effect of SCMs in mitigating the fibre degradation resulting from wet/dry cycles. Results from the study showed that the use of 30% silica fume, 90% slag and 30% metakaolin as replacement of cement prevented the degradation of the concrete samples subjected to wet and dry cycles. Similarly, the study by Toledo Filho et al. (2003) showed that the pore solution alkalinity reduced to 12.9 from 13.2 when silica fume was used as a 17% replacement of the cement. Findings from Silva et al. (2010) study also showed that the ultimate bending strength of sisal-
fibre composites made with metakaolin and calcined waste as replacement of the cement is four times more than those made with only cement as the binder.

Hence, this current study aims to investigate the influence of highly refined pulp fibre and SCMs on the interfacial characteristics and durability properties of concretes. An experimental program was carried out to assess the influence of engineered pulp fibre, namely mechanically modified fibre (MMF). MMF is a modified softwood fibre developed by Domtar Inc and the surface of MMF is modified externally. The compressive strength, chloride permeability, absorption alongside the interfacial properties of concrete with MMF and different types of SCM was evaluated.

2. Experimental Program

2.1 Materials

A Kraft pulp fibre (fully bleached) type like the one used by Booya et al. (2018) was used as reinforcement. The fibre used is a type of softwood fibres that is mechanically modified and engineered to have increased fibril counts and increased surface area. The average fibre length is 1.8 mm, and the average width (or diameter) of the fibre is 18 µm. The fibre had a density of 0.746 gm/cc and negligible lignin content.

General use limestone type cement (GULC) was used in all mixtures. Silica fume (SF), type C Fly ash (FA), metakaolin (MK) and grade 80 slag (SG), which are commercially available, were utilized as supplementary cementitious materials (SCMs). The chemical and physical characteristics of the cement and SCMs satisfies the requirements and recommendations of CSA A3001, ASTM C1240, ASTM C989, and ASTM C618 standards. Oven dried sand with maximum particle size of 5 mm and coarse aggregate that have a maximum particle size of 19 mm were used in all mixtures. A water reducer agent (high range super plasticizer) conforming to ASTM C494, was used to attain workability in the ECC mixtures.

2.2 Mixture Design and Proportioning

In order to determine the effect of SCMs on the fibre reinforced concretes, five mixtures were designed and proportioned with 1.3% by volume fibre content. One of the mixtures had no SCM in it and hence it was considered as a control mixture. The other four mixtures included binary blends in which a 10 percent of cement by weight was replaced with SCM. Varied amounts of HRWRA (superplasticizer) was added to all mixtures, to achieve required workability (slump range between of from 70 to 100 mm). Table 1 lists the mixtures and testing matrix of the tested concretes.

A specific mixing procedure was followed in this study to fabricate the concrete specimens. Moreover, the fibre preparations and conditioning followed similar procedure used by Booya et al. (2019). From each mixture, cylinders having a diameter of Ø100 and height of 200 mm, were cast and compacted. After 24 hours, the specimens were demoulded. Then, the specimens were let to cure in limewater until the time of testing (28 or 90 days). The mixture IDs are designated to reflect the type of SCM used in the concrete for example, mixture M-SF refers to a concrete mixture that included silica fume bends, while M-SG refers to concrete mixtures that included slag blends.
2.3 Testing Method

Scanning Electron Microscopy (SEM) technique was used to study the interfacial properties and the fibre fracture properties of the composites. The specimens for SEM testing were made of mortar-fibre composite that is reinforced with 2% MMF fibres by the weight of cement. The mortars also included ten percent cement replacement by a SCM type (i.e., fly ash). The chemical reaction was accelerated by subjecting the specimens to heat at 50°C for 90 days.

ASTM C39 was followed to determine the compressive strength of the produced mixtures. Three cylinders of Ø100 × 200 mm were tested, and the average value was reported at each testing age of 28, and 90 days.

The resistance chloride ion penetration of the mixtures was assessed in accordance with ASTM C1202, and the charge passed through the disk specimen was monitored and calculated. For each mixture, two 50 mm thick disks (cut from the mid-portion of a Ø100 mm diameter by 200 mm high cylinder) were prepared for testing ages of 28 and 90 days. After following the conditioning procedure specified in ASTM C1202, the disk specimens were attached to the testing cell with one face in contact with 3% NaCl solution and the other face in contact with 0.3N sodium hydroxide (NaOH) solution. A data acquisition system (DAQ) was employed to log the current (DC) of 60 ± 0.1 volts, and time over a period of six hour. The passed charge through the specimen was then calculated. The chloride permeability rating of concrete was then determined (ranging from “negligible” to “high”) per ASTM C1202.

ASTM C642 recommendations were followed to determine the water absorption percentages. Two 50 mm thick cylindrical specimens were cut from Ø100 × 200 mm cylinder specimens. The specimens were conditioned, and oven dried for 24 hours, and then cooled down to room temperature. The disk specimens were then totally immersed in 20°C water for a period of two days. The change in specimen mass was calculated. The two-day immersion water absorption was selected for each testing age of 28, and 90 days.

3. Results and Discussions

3.1 Interfacial properties

Composites containing MMF fibres were examined and investigated for interfacial characteristics. Scanning electron microscope (SEM) image of the control mixture specimen is shown in Figure 1. From this figure, it is obvious that a few fibres were pull-out from the composite matrix. The SEM examinations also showed empty pockets that indicates fibre de-bonding failure mode. Moreover, a gap in the fibre-matrix zone from specimen drying was observed. The existence of such gap contributes to increasing the matrix porosity. Hence, these observations can be associated to increasing permeability.

Table 1: Mixture proportions and testing matrix

| Property | Mixture ID | Control | M-SF | M-FA | M-MK | M-SG |
|----------|------------|---------|------|------|------|------|
| Cement (Kg/m³) | | 485 | 436.5 | 436.5 | 436.5 | 436.5 |
| Gravel (Kg/m³) | | 935 | 935 | 935 | 935 | 935 |
| Sand (Kg/m³) | | 759 | 759 | 759 | 759 | 759 |
| Water/binder | | 0.35 | 0.35 | 0.35 | 0.35 | 0.35 |
| SCM (Kg/m³) | | N/A | 48.5 | 48.5 | 48.5 | 48.5 |
| Fibre volume (%) | | 1.3 | 1.3 | 1.3 | 1.3 | 1.3 |
| Fibre mass per cubic meters | | N/A | 9.7 | 9.7 | 9.7 | 9.7 |
| Cement/SCM type | | General use lime | Silica fume | Type C Fly Ash | Metakaolin | Grade 80 Slag |
| Cement/SCM Supplier Name | | St Marys GUL | Euclid Chemicals | Lafarge Canada | Whitemud Resource | Lafarge Canada |
values reported for the control concrete mixture.

Figure 2 shows SEM images of silica fume (SF) containing specimen. The figure shows that the hydration products are precipitated on the outside surfaces of the fibres. Moreover, the composite matrix seems to have rough texture. This suggests improved bonding and reduced moisture transfer. Mohr et al. (Mohr et al. 2005) reported similar observations on the effect of including SCMs in composites reinforced with pulp fibre and exposed to wet/dry cycle.

Figure 3 shows an image for fly ash (FA) containing specimen. The SEM image shows a breakage in the fibre tip which suggests a brittle failure mode. Moreover, the image revealed that the composite exhibited fibre detachment from its matrix. Linking these observations with water permeability characteristics of the composites, it can be concluded that the fibre debonding is a predominant failure mode.

**Figure 1** Control SEM image at x500

**Figure 2** M-SF mixture SEM image at x500
On the other hand, mortar composites with 10% metakaolin blends (Figure 4) had the fibre tips fractured. The improved fibre-matrix bond was enough to break the fibres. SEM image also revealed that the matrix has dense surface which contributes to the lowered water and chloride permeations. No spaces were visible at the fibre-matrix interfacial zones.

Composites containing slag seemed to enhance the fibre-matrix bond and hence had fibre fractured as shown in Figure 5. The behaviour of slag and metakaolin, in terms of permeability and absorption, seems to be similar. However, metakaolin blends are more effective at lower replacement levels.

In all mixtures, SEM investigation did not have any evidence of ettringite formation around the fibre or near the interfacial zone. The research work herein concluded that the MK, SF, and SG blends, are beneficial in modifying the cementitious matrix which resulted in enhanced durability. However, FA blends had insignificant effect on permeability reduction.
3.2 Compressive Strength

The compressive strength values of all mixtures are shown in Figure 6. From this figure, it is obvious that mixtures with fly ash blends had the lowest compressive strength (M-FA), regardless of the testing age. Metakaolin blends have higher specific surface area than cement which optimizes the particle distribution of the mixtures. These fine metakaolin particles reduce the pore sizes that result in reducing the permeability of the concretes (Booya et al. 2019). Mixture M-MK had the highest compressive strength of 53.8 MPa, at 90 days. This is a 32% increase in strength when compared to the control mixture (Control). Silica fume and slag blends (M-SF, and M-SG) slightly increased the compressive strength in comparison to the control mixture. However, the mixture incorporated fly ash (M-FA) blends had 21.2%, and 9.6 % reduction in strength at 28, and 90, respectively, as compared to the control mixture.

![Figure 5: M-SG mixture SEM image (Booya et al. 2019)](image)

![Figure 6: Compressive strength of the mixtures at 28 and 90 days](image)
3.3 Rapid Chloride Penetration

Chloride transfer through cementitious materials often led to major defects, particularly when chloride ions reach embedded steel reinforcements. The penetration of chloride ions causes the formation of rust products, resulting in reduced durability and service life of cement-based structures (Booya et al. 2018 and Booya et al. 2019). The rapid chloride ion permeability test (RCPT) is a way of assessing and evaluating the long-term durability of concrete (Booya et al. 2020). Table 2 lists the chloride penetration charges in coulombs that passed through the specimen after a period of 6 hours. Incorporating SCM materials in the mixtures decreased the charge passed through the specimens and hence had reduced ion permeability ratings as compared to the control mixture, regardless of testing age. The reduced permeability reported for SCM containing concretes may be attributed to pore refinement or transformation of large pores into fine pores (Güneyisi et al. 2015, Chia and Zhang 2002).

Table 2: RCPT Charges and ratings as per ASTM C1202

| Age  | Mixture ID | Control | M-SF | M-FA | M-MK | M-SG |
|------|------------|---------|------|------|------|------|
| 28 day | <5500       | <3100   | <4700 | <2600 | <4100 |
| Rating | High        | Moderate | High | Moderate | high |
| 90 day | <3500       | <1600   | <2400 | <1700 | <2800 |
| Rating | Moderate     | Low     | Moderate | Low | Moderate |

3.4 Immersion Absorption

The immersion water absorption test is a way of assessing and estimating the total interconnected pores that water can reach in cementitious composites. The seventh days absorption test results are shown in Figure 7. Mixture M-FA had the largest absorption of 4.2% at 28.

It is evident from this figure that metakaolin and slag blends reduce water absorption, regardless the testing age. Hence, M-MK had the highest reduction in water absorption among all mixtures, at both testing ages.

Figure 7 Immersion Absorption of the mixtures at 28 and 90 days
4. Conclusions

Below are the main conclusions that can be drawn based on the experimental results presented in this paper. The conclusions made herein are limited to the specimens, and conditions used in the study.

- Microstructural investigation showed that few fibres were de-bonded from the matrix and that fibres had spaces at the fibre interfacial zones. The presence of these voids at the interfacial zone is expected to increase the permeability of the composites. In contrast, the use of SF, MK and SG resulted in a very dense microstructure.
- The use of MK as a replacement of cement resulted in better enhancement of the compressive strength compared to FA used.
- All concrete mixtures incorporating SCMs exhibited better durability performance compared to the control mixtures.

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