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DEVELOPMENT OF HIGH TENACITY POLYPROPYLENE FIBRES FOR CEMENTITIOUS COMPOSITES.

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ABSTRACT: This study outlines the major findings of a research program aimed at developing high tenacity polypropylene fibres for cementitious composites. In the first part, it is shown by micromechanical modelling that the use of ordinary high tenacity PP fibres leads to unstable crack propagation and large crack opening at MOR of the composites. The same model indicates that by increasing the level of interfacial frictional and chemical bonding, large improvements of the fibre-cement properties (strength, cracking behavior) are to be expected. In a second part, we explain how we developed such fibres having improved bonding to cement by optimizing the composition of the sheath of high tenacity bicomponent core/sheath fibres as well as by applying surface coatings. The obtained improvements in the properties of fibre-cement and fibre-concrete composites in comparison with ordinary high tenacity PP fibres are described.

KEYWORDS: Polypropylene fibres, bicomponent fibres, fibre-cement, fibre-concrete

1 INTRODUCTION

The progressive substitution of asbestos in the European fibre-cement industry was initiated in the early 1980's. Two main fibre types were successfully used for this application, i.e., wood pulp fibres and polyvinylalcohol fibres. While the first ones were used as reinforcement of steamcured products destined to both external (cladding) and internal (partitioning, ceiling, etc...) applications, a combination of both fibre types was used in air cured products mainly destined to roofing applications (slates and corrugated sheets).

Although many other fibre types such as polyacrylonitrile fibres (PAN), polyethylene fibres (PE), alkali resistant glass fibres (ARG), polypropylene (PP) nets and fibres, carbon fibres, (aromatic) polyamide fibres etc... are or were also used to some extent in this industry, none of them were used to the same extent as the above-mentioned fibres.

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As the asbestos substitution further progressed worldwide, particularly in low income countries, the need for cheap asbestos substitutes nevertheless continued to increase, particularly for roofing applications.

While ordinary polypropylene fibres are extensively used in the field of crack prevention of concrete, they could not be used for the reinforcement of fibre-cement products as their properties were unsatisfactory. However, as polypropylene is a relatively inexpensive polymer, worldwide available, and can be processed by classical melt spinning technologies, it remains an attractive raw material for this purpose. This is the reason why we carried out the present research program which was composed of two major parts:

1.1 On the one hand, a mathematical model of the mechanical performances and of the cracking behavior of fibre-cement was needed in order to
   a) better understand the mechanism of cement reinforcement by polymer fibres,
   b) to identify the reasons for the unsatisfactory performances of existing commercial PP fibres and, finally,
   c) to suggest ways to improve them. This was done by the Laboratory of Advanced Civil Engineering Materials of the University of Michigan, using micromechanical models.

1.2 On the other hand, based on the conclusions from this first part, the production of improved fibres was investigated using a pilot fibre spinning plant as well as a pilot fibre-cement manufacturing facility at Redco.

The present paper outlines major findings from this program and shows that it is possible to produce polypropylene based fibres having improved interfacial bonding to cement and, hence, to produce fibre-cement and fibre-concrete having improved properties in comparison with those containing ordinary PP fibres.

2 COMPOSITE MODELLING

The behavior of reinforced fibre-cement composites can be simulated using 13 micromechanical properties of the fibre-matrix system that govern composite performance.

These are basic fibre properties (stiffness E_f, length L_f, diameter d_f, and tensile strength σ_{fu}), matrix properties (stiffness E_m, toughness K_m, initial flaw size c), fibre-matrix interaction parameters (frictional bond τ_0, chemical bond (fracture energy) G_d, slip hardening coeficient β, fibre snubbing coeficient f, and fibre strength reduction coefficient due to angle effect f') and fibre volume fraction (V_f).

Fibre-matrix interaction parameters were determined from single fiber pull-out tests from cement blocs carried out as described by Katz & Li [1] and analysed according to Lin et al. [2]. K_m was was estimated from cement paste data having a water/cement ratio similar to real fibre-cement composites (0.2 MPa$\sqrt{m}$). The initial flaw size was chosen to be 60 μm and unbridged by fibres and the fibre volume fraction was set at 4.9%.

The crack bridging law (crack bridging stress vs crack opening) is obtained by averaging the contributions of only those fibres that cross the matrix crack plane as the crack opens (Li et al.[3], Lin et al. [2]).

The modelling of the flexural properties (LOP, MOR, work of fracture at MOR and crack width), uses this bridging law as well as specimen geometry, material property, load, crack length and a bridging stress distribution at each equilibrium step (Kim et al. [4]).

2.1. FIBRE-CEMENT MODELLING USING A REFERENCE ORDINARY HIGH TENACITY PP FIBRE

Based on the pull out test results obained with an ordinary high tenacity PP fibre, the following fibre and fibre/matrix parameters were chosen for the modelling.

E_f (GPa) = 11.6 τ_0 (MPa) = 0.34 f = 0.39
σ_{fu} (MPa) = 928 G_d (J/m^2) = 0 f'= 0.1
d_f (μm) = 16.6 β = 0.005

As can be seen from these data, although fibre strength is quite high, the interfacial bond strength values are low: no chemical bond, low frictional bond.

The computed flexural stress vs crack length using these data is shown in Fig. 1. When no wood pulp is present, a deep "well"appears, i.e., crack propagation occurs with a decreasing flexural load due to inadequate crack closing stress from fibre bridging (Fig. 1A). In other words, composites reinforced with such fibres show unstable crack propagation. A sudden 'pop-in' of the initial flaw into the interior of the plate can be expected to occur during the flexural test. This 'pop-in' reaches to approximately 4.6 mm of the 5.4 mm thick sample at a flexural level of 15 MPa.

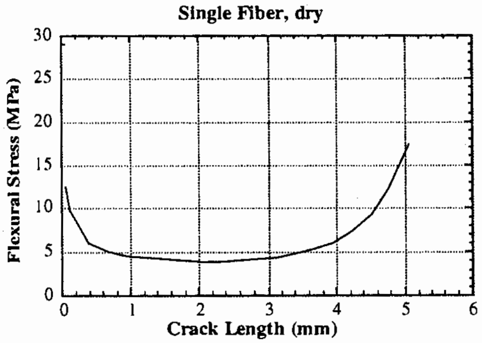


Figure 1A. Computed flexural stress vs. crack length of fibre-cement. (PP fibres only)

When wood pulp is present, such "well" is no longer present, but a phase of unstable crack propagation can still be seen (Fig. 1B).

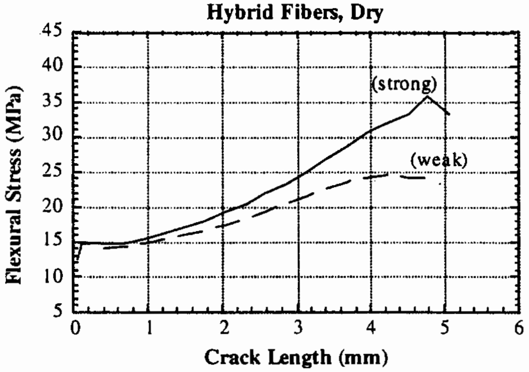


Figure 1B. Computed flexural stress vs. crack length of fibre-cement (PP + wood pulp fibres). "Strong" = machine direction; "weak" = cross direction

Fig. 2 further shows the computed crack opening profile at MOR based on the same fibre and matrix parameters. The predicted maximum crack width (crack mouth opening) is quite significant at 158 μm. Calculated crack widths in the case of PVA fibres using the same model were only in the range of 20 μm.

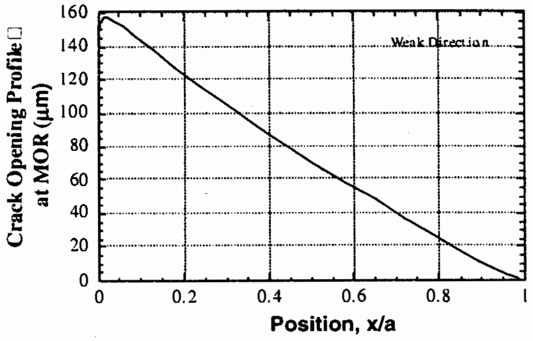


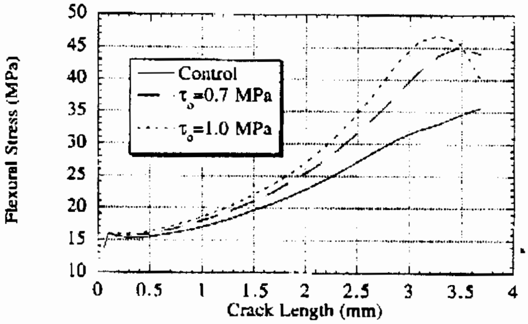
Figure 2. Computed crack opening profile at MOR (ordinary high tenacity PP fibre)

From these results, it appears that using such PP fibres for this application leads to unstable cracking of the products and large crack opening.

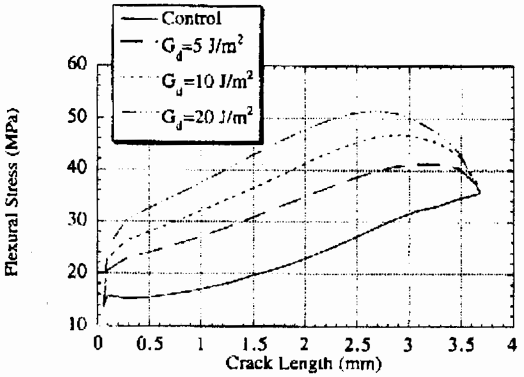
2.2. COMPOSITE MODELLING USING HIGHER BOND STRENGTH VALUES

In a second step, the model was run using higher values of frictional and/or chemical bonding, while keeping other parameters constant.

As can be seen Fig. 3A, as τ_0 increases from the control value of 0.34 MPa, the area of unstable cracking gradually disappears. The MOR also increases from the control 35.7 MPa to 45 Mpa at τ_0= 0.7 MPa, to 47 MPa at τ_0= 1.0 MPa. The size of the crack opening decreases by 59% when τ_0 increases to 1.0 MPa. Increasing the frictional bond means that it is harder for the fibres to pull-out so the springs are stiffer causing the MOR to increase and the crack opening to decrease. As crack length and corresponding crack opening increase, the fibre strength will be exceeded and the fibres will start to break which is the reason for the down turn in the top two curves. The higher frictional bond leads to a higher MOR, but also increases fiber rupture at a crack length that gradually decreases from the control of 3.7 mm.



A.



B.

Figure 3. Influence of the frictional bond (A) and of the interface fracture energy (or chemical bond) (B) on the computed Flexural stress vs crack length curves.

As shown in Fig. 3B, increasing the chemical bond causes the region of unstable cracking to disappear and the MOR to increase from 35.7 MPa to maximum 52 MPa at 20 J/m^2. The LOP similarly increases from the control 16 MPa to 25 MPa. Increasing G_d means increasing the flexural stress that must be applied to the fibres to make them debond from the matrix. This is before the fibres start to pull-out. The flexural stress required to break the chemical bonds is higher than without such bonds, and the peak load will occur at a more shallow crack length which also means a smaller crack opening. An 80% reduction in crack opening is obtained for the highest interfacial chemical bond.

3 FIBRE DEVELOPMENT PROGRAM

As the preceding modelling work shows, for PP fibers, increasing the frictional and/or the chemical interfacial bond quite effectively improves the performances of fibre-cement.

The poor bonding of ordinary high tenacity PP fibres can be easily explained based on their low surface energy (hydrophobic character) and their low roughness.

The objective of the present experimental fibre development program was thus to produce fibres having on the one hand a high tensile strength and the other hand improved surface properties leading to higher fibre-cement bonds.
In order to produce such fibres, it was decided to build a pilot bicomponent fibre spinning plant, able to produce sheath/core types fibres. A view of this pilot line comprising a spinning stage, a drawing stage, a surface treatment and coating unit, a cutter and a bagging unit is shown in Fig. 4.

Figure 4. View of the pilot fibre line at Redco NV, Belgium

The structure of the fibres produced by this line is described in Fig.5.

This technique allows to produce fibres whose sheath layer composition, and thus surface properties, differs from their core. By using this technique, one can on the one hand optimize the sheath layer for interfacial bond strength to cement, while minimizing the loss of strength and the cost increase usually associated with the presence of additives, on the other hand.

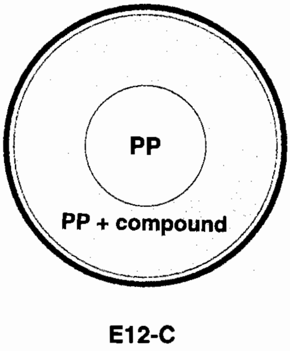


Figure 5. Structure of a sheath/core bicomponent fibre (E12C is a code name of one experimental fibres)

The first company to use this technology for the production fibres for cementitious composites was, to our knowledge, the company Daiwabo of Japan (e.g. Mitsuo et al.[5]) which used fine fillers in the sheath.

In this line, we also included a surface treatment unit, as preliminary research had indicated that a coating treatment was already effective for improving fibre-cement properties and easy to implement (Vidts & de Lhoneux [6],Vidts & de Lhoneux [7]).

In the course of this program we investigated the use of a broad range of additives in view of their contribution to increasing the interfacial bond strength and improving the fibre-cement properties, with or without additional surface treatment.
This was based on a careful analysis of the raw materials properties and processing conditions. Patents were filed about several aspects of this development.

Although fibre properties depended on many parameters, the strength of the spinnable bicomponent fibres reached a rather high level, at around 800 to 850 MPa. Table 1 shows the range of interfacial bond strength values, as measured by the pull-out tests, reached with different fibres from this production line.

Table 1. Influence of the sheath composition and surface coating on the interfacial frictional and chemical bonds.

Additive in the sheath	Surface coating	τ_0 (MPa)	G_d (J/m^2)
None	No	0.22	-
None	Yes	0.4 to 0.72	-
Mineral filler	No	0.64	-
Copolymer A	No	0.82	-
Copolymer B	No	1.51	1.4
Copolymer C	Yes	1.02	-
Copolymer D	Yes	0.76 to 1.1	0.07 to 0.10

Different mechanisms can explain the observed improvements of interfacial bonding, such as the presence of functional groups able to chemically bind with cement (coating and copolymers), an increased roughness of the surface (fillers, coating), a reduced surface stiffness, etc...

4 PROPERTIES OF FIBRE-CEMENT PRODUCTS

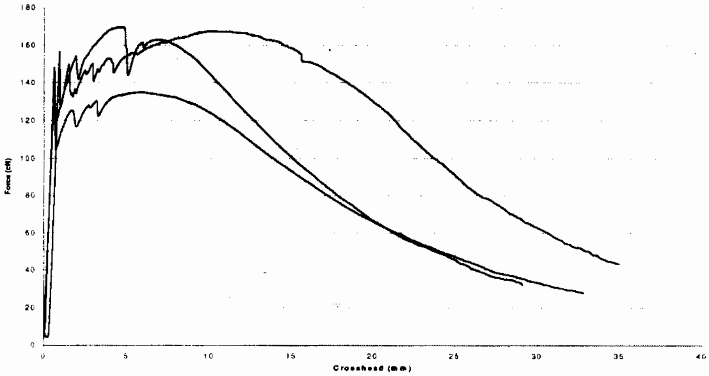
Mechanical properties of laboratory composites manufactured using 1.7 wt% of different PP fibres are shown in Table 2.

Table 2. Bending test results of laboratory fibre-cement samples containing different fibres (air dry testing, span = 146 mm, speed = 20 mm/min). A = reference ordinary high tenacity PP fibre; B = coated fibres (no additives in the sheath); C = coated fibre with fine filler in the sheath; D = coated fibre with copolymer D in the sheath.

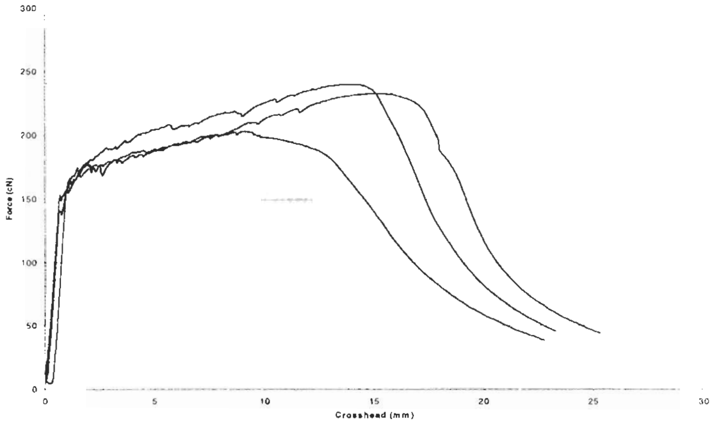
	A	B	C	D
MOR (MPa)	10.1	13.1	14.0	16.1
Work of fracture at MOR (J/m2)	317	3228	3471	4193
Density (g/cm3)	1.47	1.48	1.46	1.49

These results indicate a large influence of the surface treatment as well as of the sheath composition on the strength and toughness of the fibre-cement composites, as was expected based on the increase of the interfacial bonds observed in the pull-out tests.

In addition, as Figure 6 shows, the stress-strain curves of the composites made with the bicomponent fibres containing the copolymer D do no longer show typical load drops of ordinary PP fibres which can be attributed to crack extensions during the loading.



A.



B.

Figure 6. Stress-strain curves in three points bending test of laboratory composites in air-dry conditions containing 1.5 wt % fibre.
A = ordinary high tenacity PP fibre;
B = Coated fibre with polyolefinic copolymer D in the sheath.

In view of the achieved fibre-cement properties improvements, industrial production of fibre-cement roofing was initiated using some of these new fibres in different countries. The most important productions were made in Latin America (Peru, Colombia, Argentina, Chile) where about 300.000 M2 roofing products (corrugated sheets) were sold on the market (Fig. 7).

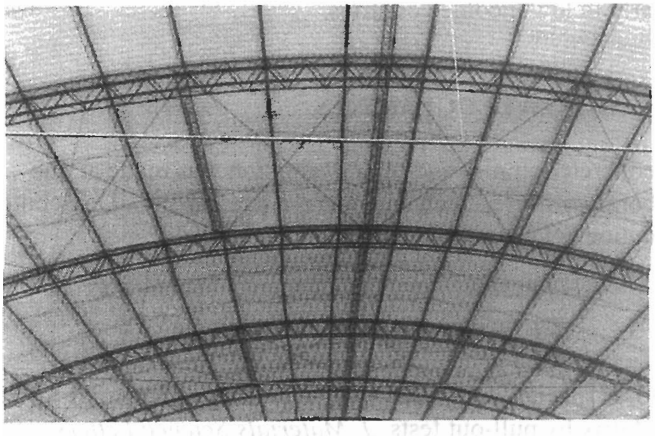


Figure 7. Fibre-cement roof made in Peru using bicomponent PP fibres from Redco pilot line.

5 FIBRE-CONCRETE

Properties of concrete containing 0.9 kg/m³ of (A) an (low tenacity) PP fibre for concrete and (B) a coated fibre (without additive in the sheath) from the pilot line are shown in Table 3.

Table 3. Properties of fibre-concrete at 0.9 kg of fibres per m^3 tested according to NBN B15-238 & B15-239 (Belgian Standards) (bending strength) and according to ACI commission-544 (J. Amer. concr. Instit. 85, 583-93)

	Bending strength (max. value) N/mm^2	Impact Number of impacts before failure
Ordinary PP fibre for concrete	1.6	11
Coated PP fibre from pilot line	1.8	75

Although no significant difference between both fibres can be seen as far as the bending strength is concerned, the improved PP fibres provide higher impact strength.

More experimentation is nevertheless needed, as significant scatter in the individual impact test results were observed.

6 CONCLUSIONS

This study has shown that the interfacial bond strength is a key factor in the development of PP fibres for fibre-cement and fibre-concrete.

By optimizing the surface properties of high tenacity fibres, it is possible to reach satisfactory fibre-cement properties.

The commercial availability of such fibres in the future will encourage the production of asbestos-free products, particularly in low income countries.

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