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THE INFLUENCE OF TYPE AND REFINEMENT OF THE CELLULOSE PULP IN THE BEHAVIOR OF FIBER CEMENT WITH HYBRID REINFORCEMENT – A REGRESSION ANALYSIS APPLICATION

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ABSTRACT

The objective of this work was to evaluate six different variables related to formulation and processing and their effect on mechanical and physical behavior of non-conventional fiber cements. The variables under analysis were (1) synthetic fiber supplier, (2) content of synthetic fiber, (3) softwood cellulose pulp supplier, (4) refinement degree of softwood cellulose pulp, (5) content of softwood cellulose pulp and (6) refinement of hardwood cellulose pulp. The fiber cement was produced in laboratory scale by the slurry de-watering process and compaction by pressing. The combination of the six variables generated 32 compositions that were submitted to the regression analysis in relation to mechanical parameters of the ordinary Portland cement based composites under flexural test at 28 days. The increase of synthetic fibers concentration and the refinement degree of the softwood cellulose pulp contributed to higher values of the modulus of rupture. The combination of softwood and hardwood pulp fibers generated the improvement of mechanical behavior of the composite regardless of the refinement of the hardwood pulp.

Keywords: PVA fiber, Cellulose pulp, Cement matrix, Fiber-cement.

INTRODUCTION

The general objective of the regression analysis is to establish a useful inter-relation between a dependent variable Y and one or more independent predictors x. The simple linear regression model can be represented in the form of one equation $Y = \alpha + \beta x$. However, in many situations there will not be a strong inter-relation between Y and any single predictor variable, and the values determination of several independent variables may considerably reduce uncertainty concerning the associated Y value. The objective of this article is to apply the regression methodology to evaluate the influence of six different predictors related to formulation and processing on mechanical and physical behavior of non-conventional fiber cements. Much

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of the research in recent years on the use of natural fiber reinforced cement (NFRC) materials has been motivated by the ready availability of such high-strength fibers. Single cell cellulose fibers can be derived from wood by the pulping process. The individual fibers present aspect ratios of about 50, with diameters in the range of 20-60 μm for hardwood, and 30-120 μm for softwood. The lengths are in the range of 0.5-3.0 mm for the hardwood fibers and 2.0-4.5 mm for the softwood fibers (Campbell and Coutts, 1980). The cellulose fibers are particularly sensitive to alkali attack (Singh, 1985). The most important reactions responsible for the loss of polysaccharides and reduction of the chain length of cellulose in alkaline pulping are peeling and hydrolytic reactions. Cellulose fibers are intrinsically strong, and refinement (or mechanical beating) of the cellulose fibers greatly improves their ability for processing. It is necessary if the composite is to be manufactured successfully using the Hatschek production method (Kim *et al*., 1999). The cellulose fiber-matrix interfacial bond, reflected in the changed mode of cellulose fiber failure at the composite fracture surface, varies with environment (dry, wet and aged). The bond of the cellulose fiber to the cementitious matrix is frictional and chemical in nature, quantified by frictional bond (τ_0) and chemical debonding energy (G_d). Loss of cellulose cement toughness with aging is attributed to the increase of interfacial bonding of the cellulose fiber to the cement matrix, which causes increased fiber rupture. Polymeric (particularly hydrophobic) fibers have the unique characteristics of poor interfacial bond strength with the cementitious matrix and weak lateral strength resulting in surface abrasion during fiber pullout from the matrix. The poor bonding characteristic is a severe limitation to the effective use of polymeric fibers in high performance cementitious composites (Wu and Li, 1999). The polyvinyl alcohol (PVA) fibers are typically produced with diameter of 10-20 μm and a tensile strength of 2,000-2,500 MPa (Kanda and Li, 1998). They are expected to present a strong bond with cementitious matrix due to their hydrophilic nature and alkalis resistance. This high bond strength is attributed to hydroxyl groups on the carbon backbone and resulting strong hydrophilic characteristics of PVA fibers (Akers *et al*., 1989). Fiber rupture has been associated to fiber strength, embedment length, frictional bond strength and angle between the fiber direction and the crack surface (Maalej *et al*., 1995). The fibers influence the mechanical behavior of composites ahead of the crack tip in the frontal process zone (FPZ) and behind the crack tip in the crack bridging length (Nelson *et al*., 2002).

EXPERIMENTAL WORK

The matrix of the composite was composed by ordinary Portland cement CPIIF type (Brazilian Standards NBR 11578, specific surface area of 0.60 m^2/g), carbonate filler (specific surface area of 0.45 m^2/g) and amorphous silica (specific surface area of 22.5 m^2/g and pozzolanic activity equal to 814 mg/g). PVA fibers were used as reinforcement with 6 mm of length and average diameter equal to 15,4 μm. Three types of cellulose pulp were used to assist with filtering in the fiber cement production and with reinforcement in the hardened composite: Brazilian *Pinus taeda* unbleached kraft pulp (approximate Kappa number 45), Chilean *Pinus radiata* unbleached kraft pulp (Kappa number 25) and bleached eucalyptus (mix of *Eucalyptus saligna* and *Eucalyptus grandis*) kraft pulp. The Kappa number (Appita P201 m-86) is an indirect measurement of lignin content. It is of particular interest in the characterization of unbleached kraft pulps. The major properties of cellulose fibers are described in Table 1. The Schopper-Riegler (°SR) number is a measurement of the drainability of a suspension of pulp in water, determined and expressed as specified in SCAN-C19: 65 (Scandinavian Pulp, Paper and Board Testing Committee, 1964). The pads of cement composite were produced in laboratory scale by

slurring the raw material in water solution (20% of solids) followed by vacuum drainage of the excess of water and pressing. It was an attempt to simulate the Hatschek method for sheeting fabrication. Hardened pads were wet diamond sawn with dimensions of 40 x 160 mm. Test specimen depth was the thickness of the pad, which was in the region of 5 mm. Twenty specimens for each formulation were submitted to wet curing for seven days and then allowed to air cure until the execution of mechanical tests which were conducted in the same environment (Eusebio *et al*., 1998).

TABLE 1. Properties of Cellulose Fibers and Pulps

Sample	Length (mm)	Coarseness (mg/100m)	Drainability (°SR)	Fines (%) [b]
Chilean softwood pulp 0 [a]	1.85	32.73	13.0	9.11
Chilean softwood pulp 8000	1.22	11.30	66.0	48.94
Brazilian softwood pulp 0	1.72	42.80	13.0	8.06
Brazilian softwood pulp 7000	1.24	11.56	70.0	25.28
Hardwood pulp 0	0.70	6.92	19.0	10.97
Hardwood pulp 4500	0.68	5.77	69.0	10.95

[a] The number after the fiber description indicates the number of the revolutions in the PFI refiner.
[b] Percentage by mass of fibers with length under 0.07 mm.

The mechanical and physical properties evaluated at 28 days of total age were modulus of rupture, toughness and water absorption. The mechanical behavior based on the Rilem recommendations (49 TFR) was employed using a four point bending configuration. A span of 135 mm and a deflection rate of 1.5 mm/min were used for all tests in a EMIC DL30000 universal testing machine equipped with load cell of 1 kN. Physical characterization followed the specifications of the Brazilian Standard NBR-9778. The microstructure of the fracture surface of the composite was evaluated by scanning electron microscope (SEM) using backscattering electron image.

The combination of the six predictors generated compositions that were submitted to the regression analysis in relation to the variables: modulus of rupture (Y_1), toughness (Y_2) and water absorption (Y_3) of the fiber cement composites at the total age of 28 days. The predictors under analysis were (x_1) synthetic fiber supplier (Japanese: 0, Chinese: 1); (x_2) content of synthetic fiber (1.2% by mass of dry raw-materials: -1, 2.4% by mass of dry raw-materials: +1); (x_3) supplier of the softwood cellulose pulp (Brazilian: -1, Chilean: +1); (x_4) refinement degree of softwood cellulose pulp (no refinement: -1, maximum refinement: +1); (x_5) content of softwood cellulose pulp (1.2% by mass of dry raw-materials plus 2.8% of short fiber: -1, 4% of long fiber and no short fiber in the formulation: +1); and (x_6) refinement degree of hardwood cellulose pulp (no refinement: -1, maximum refinement: +1, and no short fiber in the formulation: 0). The experiment was designed with half of the factorial adding thirty-two different compositions. This model was adjusted for each answer as a multiple regression model (Devore and Peck, 1996) containing six factors plus all interactions between the major factors two by two, as in the Eq. (1).

$$Y_j = \beta_0 + \beta_1 x_1 + \beta_2 x_2 + \beta_3 x_3 + \beta_4 x_4 + \beta_5 x_5 + \beta_6 x_6 + \beta_7 x_2 x_3 + \beta_8 x_2 x_4 + \beta_9 x_2 x_5 + \beta_{10} x_2 x_6 + \beta_{11} x_3 x_4 + \beta_{12} x_3 x_5 + \beta_{13} x_3 x_6 + \beta_{14} x_4 x_5 + \beta_{15} x_4 x_6 + \beta_{16} x_5 x_6 + ERROR_j \quad (1)$$

Where the β´s are called population regression coefficients. Each β_i can be interpreted as the true average change in Y_j when the predictor x_i increases by one unit and the values of all other predictors remain fixed. β_0 = average of the variable Y_j.

RESULTS

The presence of higher concentration of PVA fibers associated with the effect of the refinement of the cellulose fibers contributed with the increase of the modulus of rupture of the composites. Figure 1 shows the refined pulp after pullout from the cement matrix and with hydration products incrusted at its surface. The strong adhesion of the fibers with the matrix reduced the pullout of fibers and increased the modulus of rupture (MOR) of the composites at 28 days. Figure 2 depicts the variation of the modulus of rupture with the content of synthetic fiber associated with the refinement of the softwood cellulose pulp.

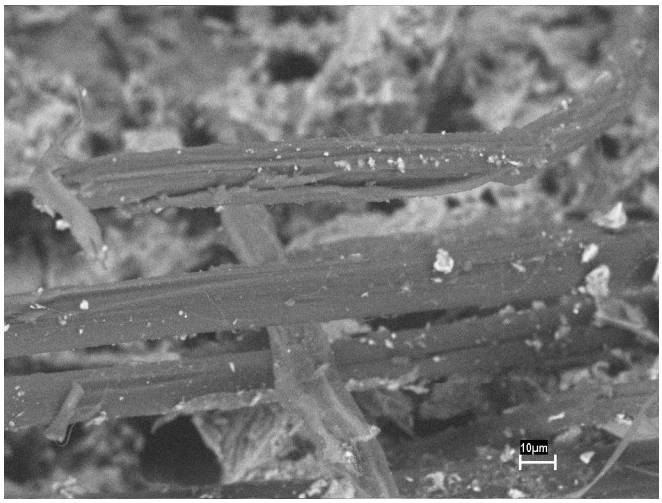

FIG. 1. SEM of fracture surface of the composite. Detail of pulled out cellulose fibers (scale bar = 10 µm)

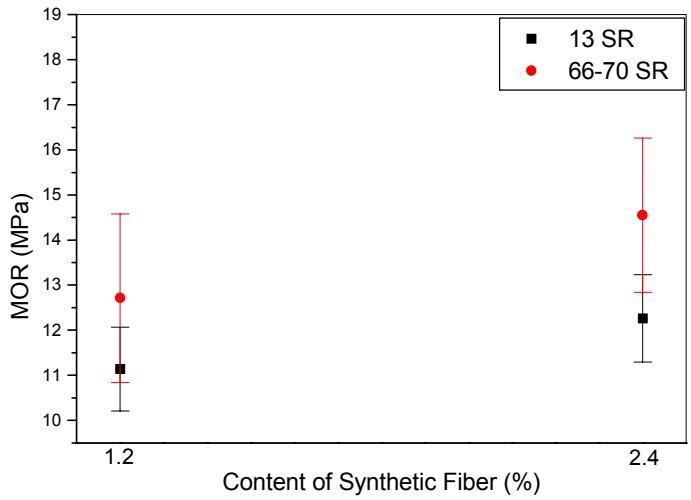

FIG. 2. Modulus of rupture x content of synthetic fiber associated with refinement degree of softwood cellulose pulp

Table 2 presents the results of the variable Y_1 obtained by the application of the regression analysis. According to this model the Eq. (2) explains 87.3% of variability by Y_1 (s = 0.9587, R-Sq = 87.3% and R-Sq(adj) = 75.3%). The highest percentage by mass of softwood cellulose pulp (x_5) conducted to a negative effect in relation to the modulus of rupture (Y_1) of the composites. The Japanese synthetic fiber (x_1), the higher concentration of PVA fiber (x_2) and the maximum refinement of the softwood cellulose pulp (x_4) caused the more significant ($P < 0.05$) increase of the Y_1 variable. The positive interactions x_2*x_4 and x_4*x_5 were considered the more significant ($P < 0.05$) ones.

TABLE 2. Modulus of rupture (Y_1)

Predictor	Coef	StDev	P	Predictor	Coef	StDev	P
Constant	12.0438	0.2399	0.000	β_8	0.4774	0.1695	0.012
β_1	-1.1787	0.3390	0.003	β_9	-0.1783	0.1695	0.308
β_2	0.6017	0.1695	0.003	β_{10}	0.5599	0.2397	0.033
β_3	0.1660	0.1695	0.342	β_{11}	0.1652	0.1695	0.344
β_4	0.6975	0.1695	0.001	β_{12}	0.0203	0.1695	0.906
β_5	-1.0285	0.1695	0.000	β_{13}	0.0573	0.2397	0.814
β_6	-0.2922	0.2397	0.240	β_{14}	0.5108	0.1695	0.008
β_7	-0.1414	0.1695	0.416	β_{15}	-0.4459	0.2397	0.081

$$Y_1 = 12.04 - 1.18x_1 + 0.60x_2 + 0.17x_3 + 0.70x_4 - 1.03x_5 - 0.29x_6 - 0.14x_2x_3 + 0.48x_2x_4 - 0.18x_2x_5 + 0.56x_2x_6 + 0.17x_3x_4 + 0.02x_3x_5 + 0.06x_3x_6 + 0.51x_4x_5 - 0.45x_4x_6 \quad (2)$$

Table 3 presents the results of variable Y_2 obtained by application of the regression analysis. According to this model the Eq. (3) explains 92.7% of variability by Y_2 (s = 0.3099, R-Sq = 92.7% and R-Sq (adj) = 85.9%). The Japanese synthetic fiber (x_1), the increase of the content of synthetic fiber (x_2), the refinement of softwood cellulose pulp (x_4) and the addition of hardwood cellulose pulp (x_5) without refinement (x_6) contributed to the increase of the composite toughness ($P < 0.05$). The combination of long and short cellulose fibers probably conducted to more homogeneous distribution of fibers inside the composites with the favorable formation of multiple secondary cracks during the flexural tests. The synthetic fiber content (x_2) presented a positive interaction with the refinement of the softwood pulp (x_4) ($P < 0.05$). Other significant interactions ($P < 0.05$) for the variable Y_2: x_2*x_6, x_4*x_5 and x_4*x_6.

$$\begin{aligned} Y_2 = {} & 3.71 - 0.28x_1 + 0.49x_2 + 0.05x_3 + 0.05x_4 - 0.27x_5 - 0.29x_6 + 0.30x_2x_4 \\ & - 0.10x_2x_5 + 0.36x_2x_6 + 0.02x_3x_4 + 0.01x_3x_5 + 0.02x_3x_6 + 0.16x_4x_5 - 0.31x_4x_6 \end{aligned} \quad (3)$$

TABLE 3. Toughness (Y_2)

Predictor	Coef	StDev	P	Predictor	Coef	StDev	P
Constant	3.70501	0.07748	0.000	β_8	0.29629	0.05479	0.000
β_1	-0.2825	0.1096	0.020	β_9	-0.09884	0.05479	0.090
β_2	0.48690	0.05479	0.000	β_{10}	0.35798	0.07748	0.000
β_3	0.04691	0.05479	0.405	β_{11}	0.01770	0.05479	0.751
β_4	0.04634	0.05479	0.410	β_{12}	0.01234	0.05479	0.825
β_5	-0.26571	0.05479	0.000	β_{13}	0.02367	0.07748	0.764
β_6	-0.29359	0.07748	0.002	β_{14}	0.15827	0.05479	0.011
β_7	-0.00098	0.05479	0.986	β_{15}	-0.30728	0.07748	0.001

Table 4 presents the results of variable Y_3 obtained by the application of the regression analysis. According to this model the Eq. (4) explains 87.8% of variability by Y_3 (s = 0.8255, R-Sq = 87.8% and R-Sq (adj) = 76.4%).

TABLE 4. Water absorption (Y_3)

Predictor	Coef	StDev	P	Predictor	Coef	StDev	P
Constant	21.8756	0.2064	0.000	β_8	-0.0007	0.1459	0.996
β_1	0.0805	0.2919	0.786	β_9	-0.2339	0.1459	0.129
β_2	0.7504	0.1459	0.000	β_{10}	-0.1383	0.2064	0.512
β_3	-0.2909	0.1459	0.064	β_{11}	0.3399	0.1459	0.033
β_4	-0.9206	0.1459	0.000	β_{12}	0.2036	0.1459	0.182
β_5	0.2204	0.1459	0.150	β_{13}	0.4261	0.2064	0.056
β_6	-0.2481	0.2064	0.247	β_{14}	-0.7502	0.1459	0.000
β_7	-0.0313	0.1459	0.833	β_{15}	-0.0353	0.2064	0.866

$$Y_3 = 21.90 + 0.08x_1 + 0.75x_2 - 0.29x_3 - 0.92x_4 + 0.22x_5 - 0.25x_6 - 0.03x_2x_3 - 0.23x_2x_5 - 0.14x_2x_6 + 0.34x_3x_4 + 0.20x_3x_5 + 0.43x_3x_6 - 0.75x_4x_5 - 0.04x_4x_6 \quad (4)$$

Figure 3 shows that the values of water absorption reduced for composites reinforced with refined softwood as a consequence of the improved packing of the material. The increase of synthetic fiber content (x_2) led to the higher water absorption. The significant interactions ($P < 0.05$) for the variable Y_3 were x_3*x_4 (positive) and x_4*x_5 (negative).

The interaction between the content of softwood pulp (x_5) and refinement of hardwood pulp (x_6) was found to be highly correlated with other x predictors for the Y variables under analysis. For this reason the interaction x_5*x_6 was removed from all the regressions equations (2) to (4).

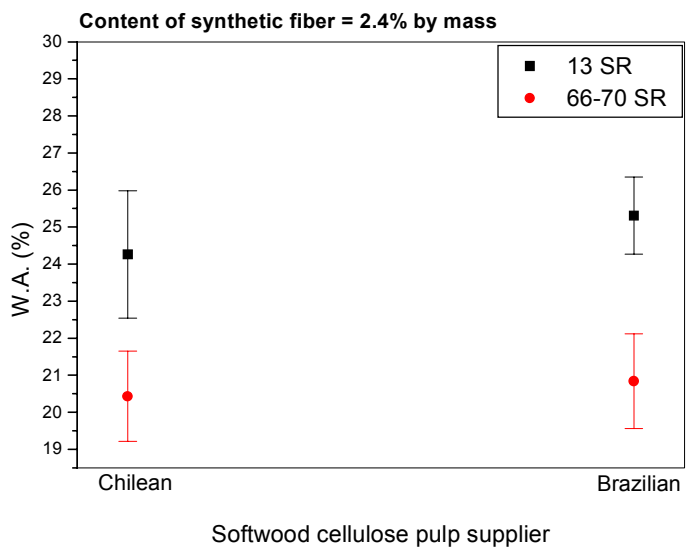

FIG. 3. Water absorption x softwood cellulose pulp supplier associated with refinement degree of softwood cellulose pulp.

CONCLUSIONS

The variation of the factors "supplier of softwood cellulose fiber" (x_3) and "refinement of hardwood cellulose fiber" (x_6) did not cause significant changes ($P < 0.05$) in the modulus of rupture of the composites containing synthetic fibers. The refinement degree of the softwood cellulose pulp was one of the most important factors in mechanical strength and water absorption of the composites. The reinforcement with 1.2% by mass of softwood pulp combined with 2.8% by mass of hardwood pulp conducted to better mechanical behavior than 4% by mass of sole softwood pulp. The reinforcement with Japanese PVA fiber resulted in composites with higher modulus of rupture and tenacity. The higher content of synthetic fiber was also associated with a sensible improvement of the tenacity and of the modulus of rupture of the composites. Intermediate levels between 1.2-2.4% by mass of synthetic fiber should be tested in future works.

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