Optimization of Carbonated Cellulose Fiber-Cement Composites

Won, Jong-Pil* **Bae, Dong-In****

ABSTRACT

This research developed an accelerated curing processe for cellulose fiber reinforced cement composites using vigorous reaction between carbon dioxide and cement paste. A wet-processed cellulose fiber reinforced cement system was considered. Carbonation curing was used to complement conventional accelerated curing.

The parametric study followed by optimization investigation indicated that the carbonation curing can enhance the productivity and energy efficiency of manufacturing cellulose fiber reinforced cement composites. This also adds environmental benefits to the technical and economical advantages of the technology.

Keywords : carbonation, cellulose fiber-reinforced cement composite, flexural strength, initial stiffness, specific gravity, toughness, water absorption

---

* KCI Member, Assistant Prof., Dept. of Agricultural Engineering, Konkuk University, Korea

** KCI Member, Graduate student, Dept. of Agricultural Engineering, Konkuk University, Korea

1. Introduction

Cement and concrete products are notable for their weakness in tension, and lack of toughness which gives rise to early cracking under mechanical or thermal loads. The use of reinforcing fibers, such as polypropylene, steel and glass fiber, to overcome such deficiencies is now well established.

The cellulose fibers derived from softwood or hardwoods present highly cost effective means of reinforcement for thin cement products. Cellulose fibers provide adequate stiffness, strength, and bonding capacity to cement-based matrices for substantial enhancement in flexural strength , toughness, and impact resistance. The improvements are achieved through the stopping and deflecting cracks propagating in brittle cement matrices by cellulose fiber. The processing of cellulose fiber-reinforced cement composites are sensitive to the specific wood species and cellulose fiber types. The setting and hardening of cellulose fiber-reinforced cement composites is a slow process. Wet processed cellulose fiber reinforced cement needs to be autoclaved for several hours before it gains sufficient strength. Efforts to substantially increase the setting and hardening rate of cement should still provide sufficient "open time" during which the cellulose fiber-cement-water furnish stays plastic to be mixed, formed and pressed. Tremendous reductions in the setting time of cement-based binders can be achieved by the addition of carbon dioxide (CO_2). The predominant chemical reaction occurring with carbonation involves $Ca(OH_2)$ resulting from the hydration of cement, which reacts with CO_2 to produce $CaCO_3$ (limestone). The rapid carbonation reaction may also help reduce the cure time in wet processing of cellulose fiber-reinforced cement.

The goal of this study is to develop a rapid, cost-effective, and energy-efficient processing technique which enables the production of cellulose fiber-cement composites with improved engineering properties. The study relies on CO_2-cure of cement-based materials was use to accomplish the objective.

2. Experimental Program

2.1 Identification of Influential Processing Variables

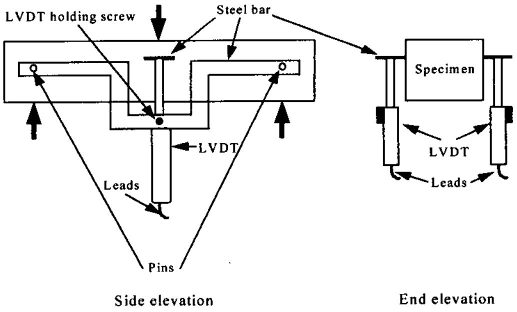
The objective of this part was to determine variables which influence proportioning and processing in manufacturing cellulose fiber-cement composites. The cellulose fiber selected for this investigation was Southern Softwood Kraft (SSK) pulp. Average length of the fiber was 3.0mm. The cellulose fiber-reinforced cement composites were manufactured as followes: (1) a weighted dry lap of cellulose fiber was soaked in water for minimum 4 hours; (2) a laboratory-scaled pulp disintegrator (TMI refiner) was used at a speed of 3,000 rpm. The beating time used in this study was 10 minutes; (3) The fibers, sand, flocculating agent, and cement were proportioned; (4) The ingredients were mixed in water to produce a slurry of 20% solids by weight; a high-speed mixer was used to achieve a uniform dispersion of cellulose fibers and other mix ingredients in the slurry. Flocculating agent was the last solid constituent to be added, which improved the binding of cement particles to cellulose fibers and prevented the cement particles from escaping during vaccum-dewatering of the slurry; (5) Vaccum dewatering of the slurry to extract the excessive water. The effects of following variables was investigated through a full factorial experimental design: oven temperature, oven duration, CO_2-chamber duration, and autoclave duration. Each factor was considered at two levels as shown in Table 1 which summarizes the experimental program.

2.2 Experimental Set-Up

Flexural tests were performed in accordance with the ASTM C 1185. The flexural test samples had a clear span of 254 mm, a width of 152.4 mm, and thickness of 10 mm. Fig. 1 shows the 3-point flexural test set-up used. The flexural performance was evaluated in wet condition. Specimens were immersed in water at a temperature of 23±4℃ for a period of minimum 48 hours. The specimens were tested immediately upon removal from the water.

3. Test Results and Statistical Analysis

The flexural test results are shown in Table 2 and Fig. 2. Based on the statical analyses, oven duration had statistically significant effects at 95% level of confidence on flexural strength. In the case of toughness, oven temperature, oven duration, oven duration-CO_2 chamber duration interaction, and CO_2 chamber duration-autoclaved duration interaction were statistically significant effects at 95% level of confidence. In the case of stiffness, oven duration, oven duration-CO_2 chamber duration interaction, and CO_2 chamber duration-autoclave duration interaction were statistically significant effects at 95% level of confidence (Figs. 3, 4, and 5).



Side elevation

End elevation

Fig. 1 Flexural test set-up for cellulose fiber-cement composite

These effects had to be considered in the optimization process at next phase of the study. However, in order to reduce the size of optimization test program, we decided to keep the less in flexural factor of oven temperature constant. In order to select this temperature, a series of tests were conducted at different

Table 1 Disign of experiment

Factor	Code	Factor Level		Control 1	Control 2
		-	+		
Oven temperature (℃)	A	50	100	50	
Duration in oven (hours)	B	1	2	1	
Duration in CO_2 chamber: @20% CO_2 concentration (hours) (50℃ and 95% RH)	C	1	4	1 (0% CO_2)	
Autoclaved duration (hour)	D	4	8	4	8

Table 2 Flexural performance of cellulose fiber-reinforced cement composite

Run order	A	B	C	D	Strength (Mpa)	Mean (St.Dev.)	Toughness (N-mm)	Mean (St. Dev.)	Stiffness (N/mm)	Mean (St. Dev)
1	-	-	-	-	5.135 5.866 5.538	5.513 (0.366)	3002.119 2329.511 2629.431	2653.687 (336.959)	137.632 141.769 140.167	139.856 (2.086)
2	+	-	-	-	5.432 5.918 4.918	5.423 (0.5)	1999.263 2007.884 1922.771	1976.639 (46.850)	157.213 176.426 107.288	146.976 (35.688)
3	-	+	-	-	5.723 5.331 4.935	5.330 (0.394)	1966.347 1843.331 1808.799	1872.826 (82.812)	81.642 80.342 79.621	80.535 (1.024)
4	+	+	-	-	3.084 5.461 4.321	4.289 (1.189)	670.425 1180.216 869.211	906.617 (256.946)	31.089 45.042 40.568	38.900 (7.125)
5	-	-	+	-	7.079 7.035 7.055	7.056 (0.022)	2554.452 2401.637 2429.364	2461.818 (81.413)	116.016 111.432 113.214	113.554 (2.311)
6	+	-	+	-	7.406 5.785 6.993	6.728 (0.842)	2793.201 2624.798 2683.703	2700.567 (85.459)	166.653 159.653 163.213	162.983 (3.791)
7	-	+	+	-	2.529 2.556 2.490	2.525 (0.033)	838.152 829.555 814.732	827.480 (11.847)	51.678 53.216 59.598	54.831 (4.200)
8	+	+	+	-	2.437 2.005 2.490	2.226 (0.216)	954.226 609.203 782.214	781.881 (172.512)	54.432 20.490 40.316	38.413 (17.051)
9	-	-	-	+	5.327 5.183 5.695	5.402 (0.264)	2183.562 1966.189 2314.418	2154.723 (175.897)	105.221 104.215 108.184	105.873 (2.063)
10	+	-	-	+	3.106 3.187 3.099	3.131 (0.049)	885.823 737.394 799.921	807.713 (74.521)	46.102 39.990 43.210	43.101 (3.057)
11	-	+	-	+	3.963 3.503 3.713	3.726 (0.230)	1916.395 1545.357 1721.334	1727.695 (185.601)	120.150 110.003 115.412	115.188 (5.077)
12	+	+	-	+	3.821 4.260 3.578	3.886 (0.346)	1510.313 1419.553 1642.057	1523.974 (111.879)	89.216 84.958 92.232	88.802 (3.655)
13	-	-	+	+	4.678 5.216 6.125	5.340 (0.731)	2515.911 2843.524 3223.308	2860.914 (354.019)	141.786 146.219 154.340	147.448 (6.367)
14	+	-	+	+	6.326 6.471 6.080	6.292 (0.198)	2721.226 2632.058 2972.258	2775.181 (176.401)	139.213 165.021 133.073	145.769 (16.953)
15	-	+	+	+	5.810 4.323 4.826	5.320 (±0.863)	3204.346 2217.397 2823.421	2748.388 (497.734)	206.071 135.995 170.221	170.762 (35.041)
16	+	+	+	+	3.820 3.530 4.486	3.945 (0.490)	1526.931 1380.532 1813.403	1573.622 (220.180)	92.624 71.170 113.059	92.284 (20.947)
Control 1					3.200 4.438 3.284	3.641 (0.692)	1596.002 2173.015 1349.216	1706.078 (422.787)	105.400 108.416 96.915	103.577 (5.963)
Control 2					4.225 5.499 5.392	5.039 (0.707)	1956.227 2345.003 2004.321	2101.850 (211.945)	159.526 142.926 150.338	150.930 (8.316)

oven temperatures (20, 35, 50, and 100℃). The fixed variables used were oven duration of 1 hour, CO_2 chamber duration of 1 hour, and autoclave duration of 8 hour which yielded the highest flexural strength. Fig. 6 shows the flexural performance obtained at different oven temperatures for cellulose fiber reinforced cement composites. 50℃ yielded a balanced properties among the four temperatures considered.

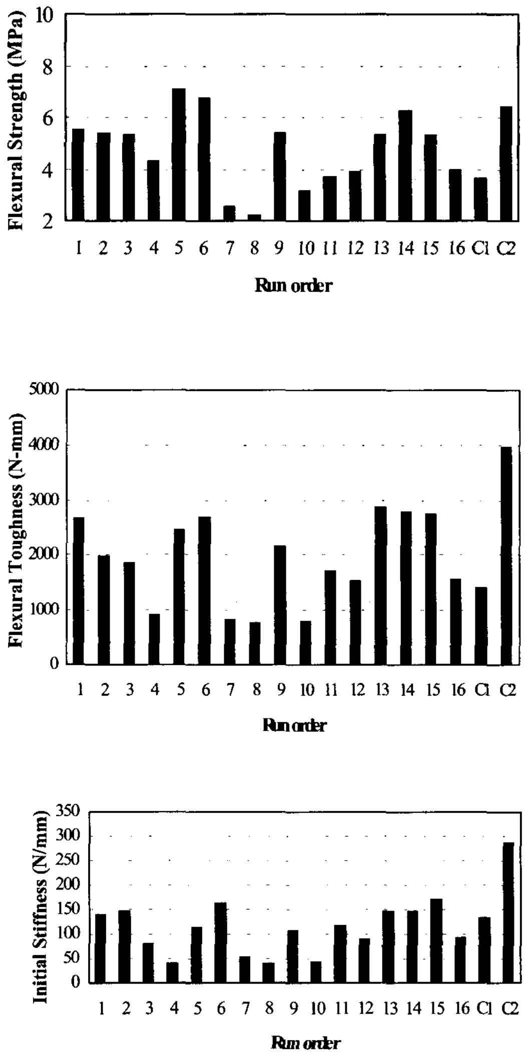


Fig. 2 Flexural test results: c1=control 1; c2=control 2

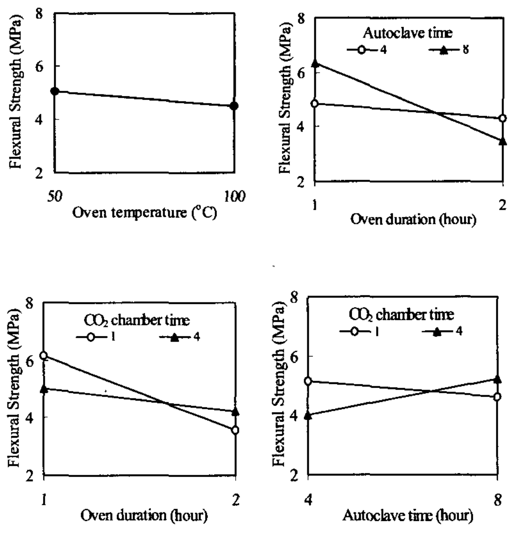


Fig. 3 Trends in flexural strength

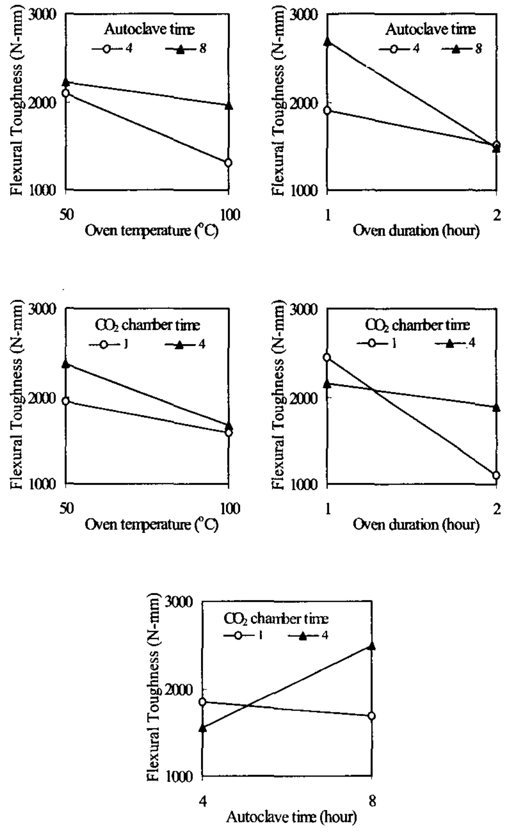


Fig. 4 Trends in flexural toughness

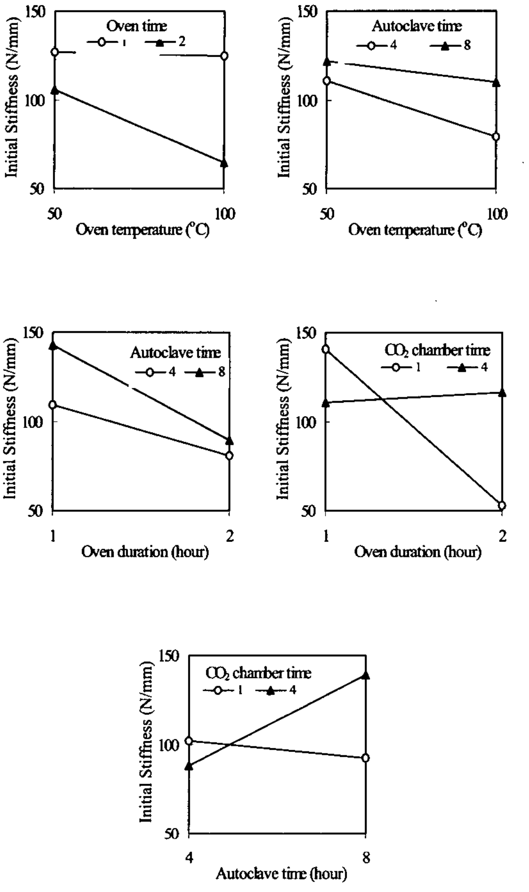


Fig. 5 The trends in initial stiffness

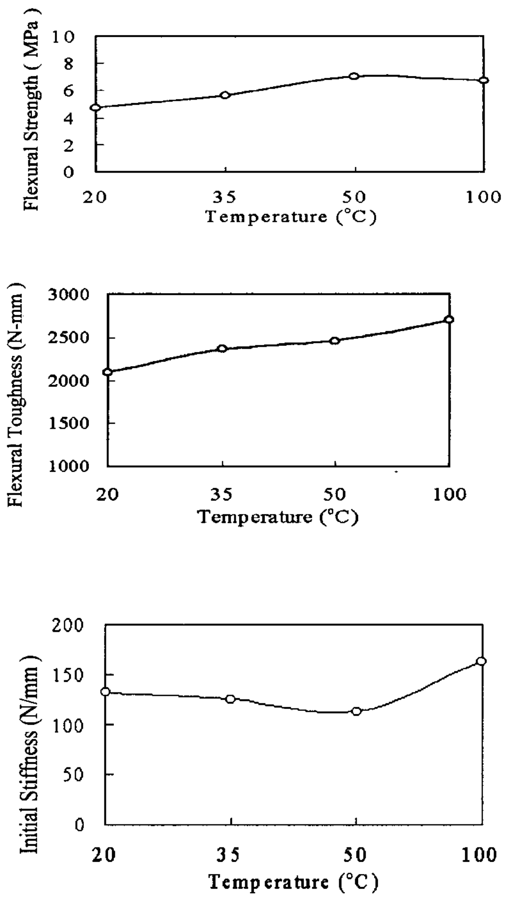


Fig. 6 Flexural performance at the different oven temperatures

4. Optimization of Cellulose Fiber-Reinforced Cement Composites by CO_2 Curing

Three influential variables identified in previous phase of the study (oven duration, CO_2 chamber duration, and autoclave duration) were optimized based on performance and cost consideration. The optimization for experimental design was formulated based on the statistical theory of response surface analysis. The composites were optimized considering their flexural performance (strength, toughness, and stiffness) and material cost. Table 3 shows the experimental program for optimization through response surface analysis. Various combinations of the three statistically influential variables were considered in the experimental program. A summary of all flexural test results is listed in Table 3. The optimum level of statistically influential variables, which was derived from the response surface analysis of the flexural test results and cost analysis were as follows: 50℃ oven temperature, followed by oven duration of 1 hour, CO_2 chamber duration of 1 hour, and autoclaved duration for 6 hours.

The flexural performance of the optimized cellulose fiber-reinforced cement composite produced through CO_2-cure is compared in Fig. 7 and Table 4 with one of two control boards manufactured without the CO_2 cure. All tests were conducted in equilibrium condition. Strength of CO_2 cured composite was higher than those of control boards at 95% level of confidence.

Table 5 shows percentile difference in flexural properties of the CO_2-cured composites versus those of the control boards.

Table 3 Optimization of experimental design and test results

Run order	A	B	C	Strength (MPa)	Toughness (N-mm)	Stiffness (N/mm)
1	0.5	4	4	4.246 4.031 3.797	1425.917 1621.500 1779.600	112.134 120.376 118.542
	Mean (St. Dev.)			4.019 (0.225)	1609.006 (177.172)	117.017 (4.327)
2	1	2.5	6	5.653 5.522 5.511	2435.489 1885.827 1921.531	157.210 72.698 121.214
	Mean (St. Dev.)			5.562 (0.079)	2080.949 (307.559)	117.041 (42.410)
3	1.5	1	4	3.889 3.660 4.397	2001.224 1963.115 1841.763	77.645 85.065 69.452
	Mean (St. Dev.)			3.982 (0.377)	1935.367 (83.273)	77.387 (7.810)
4	1	2.5	6	5.159 5.037 5.110	1799.249 1505.522 1621.554	96.546 191.886 132.621
	Mean (St. Dev.)			5.102 (0.062)	1642.108 (147.938)	140.351 (48.138)
5	0.5	1	4	5.145 5.200 5.250	2462.260 1924.521 1725.402	213.402 153.211 95.638
	Mean (St. Dev.)			5.198 (0.053)	2037.313 (132.637)	154.084 (58.887)
6	0.5	1	8	5.214 5.896 4.107	2019.346 2219.677 1968.917	142.169 134.985 153.683
	Mean (St. Dev.)			5.072 (0.903)	2069.313 (132.637)	143.612 (9.432)
7	1.5	1	8	6.769 6.772 6.774	2928.741 2543.263 2315.729	158.996 139.211 124.919
	Mean (St. Dev.)			6.772 (0.003)	2595.911 (309.879)	141.042 (17.112)
8	1	2.5	6	5.650 5.421 5.368	2399.291 1898.541 1821.699	150.624 129.553 132.626
	Mean (St. Dev.)			5.480 (0.150)	2039.844 (313.653)	137.601 (11.382)
9	1.5	4	4	4.736 4.686 4.589	1824.316 1644.419 1743.216	110.624 104.421 108.523
	Mean (St. Dev.)			4.670 (0.075)	1737.317 (90.093)	107.856 (3.155)
10	1.5	4	8	4.899 4.767 5.207	2100.913 2098.820 2260.309	159.523 155.791 168.041
	Mean (St. Dev.)			4.898 (0.130)	2153.347 (92.637)	161.118 (6.279)
11	1	2.5	6	5.160 5.308 5.529	1821.215 1621.213 1892.321	100.548 188.521 100.629
	Mean (St. Dev.)			5.242 (0.256)	1778.250 (140.568)	129.899 (50.768)
12	0.5	4	8	4.826 4.709 4.920	1621.523 1502.582 1770.075	116.435 110.928 119.919
	Mean (St. Dev.)			4.818 (0.106)	1631.393 (134.019)	115.761 (4.533)

*A : Oven duration B: CO_2 Chamber duration
C : Autoclaved duration, Numbers for A, B, and C represent hours

Table 4 Flexural performance of optimized CO_2-cured cellulose fiber-reinforced cement composites vs. controls

Type of Composite	Strength (MPa)	Toughness (N-mm)	Stiffness (N/mm)
CO_2-cured (1-1-6)*	11.72 11.61 11.64 11.14	2281.792 1660.080 1721.514 1834.543	191.313 194.831 187.215 190.216
Mean (St. Dev.)	11.528 (0.262)	1874.543 (280.989)	190.894 (3.145)
Control 1 (1-1-6)**	7.66 7.60 7.58 7.68	1767.638 1984.112 1824.312 1722.558	122.327. 141.628 131.219 129.214
Mean (St. Dev.)	7.63 (0.048)	1824.655 (114.166)	161.348 (19.755)
Control 2 (1-1-8)**	8.92 8.39 8.42 8.36	2622.431 2100.280 2522.521 2213.216	177.170 132.792 149.215 139.228
Mean (St. Dev.)	8.523 (0.266)	2364.612 (247.789)	149.601 (19.582)

* : 1-1-6 means 1 hour in oven at 50℃, 1 hour in CO_2 chamber and 6 hours in autoclave

** : 1-1-6 or 1-1-8 means 1 hour in oven at 50℃, 1 hour in CO_2 chamber but with 0% CO_2 and 6 or 8 hours in autoclave

Table 5 Comparison of flexural performances: CO_2-cured boards vs. controls

	CO_2-cure/Control 1	CO_2-cure/Control 2
Flexural Strength	+51%	+35.1%
Flexural Toughness	+2.7%	-20.7%
Initial Stiffness	+18.3%	+29.4%

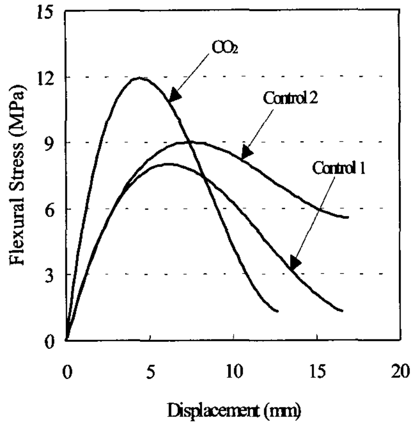
Table 6 Water absorption and specific gravity test results

	Control 1	Control 2	CO_2
Specific Gravity	1.11	1.18	1.11
Water Absorption(%)	51.42	48.39	45.04

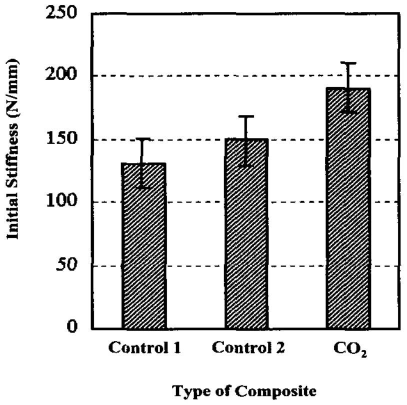
CO_2-cure yielded better matrix and board qualities, but it had some negative effects on initial toughness.

Density, water absorption, and dimensional stability tests were carried out on the optimized composites in accordance with ASTM C 1185 and C 1186. Water absorption and specific gravity are indirectly related to density in that both are dependent upon the void volume of the sample. The water absorption capacities are shown in Table 6 and Fig. 8 for CO_2-cured and control boards. The CO_2-cure on cellulose fiber-reinforced cement composites in Fig. 8 shows reduced water absorption when compared with control. A denser structure could be responsible for this phenomenon.

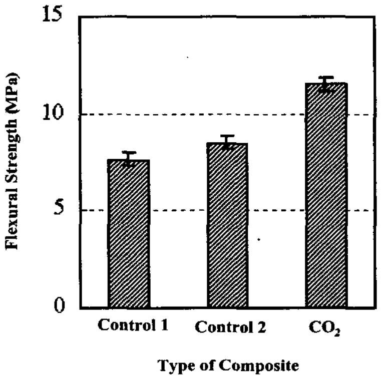
A cellulose fiber consists of long chains of glucose molecules attracted to each other by hydrogen bonds. It contains voids in excess of 50% of the total volume. When these voids are filled with moisture, due to expansion of cellulose fibers, the dimensional stability of the material is affected. The diameter change of cellulose is large, and can affect the bonding of the fiber to the matrix. Dimensional stability is measured in terms of dimensional movements expressed as the



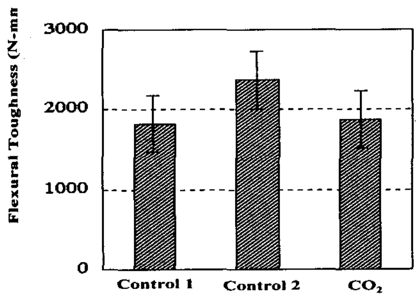
(a) Load-Deflection Behavior



(b) Flexural Strength



(c) Flexural Toughness



(d) Initial Stiffness

Fig. 7 Flexural performance of optimized CO_2 cured cellulose fiber reinforced cement composite (equilibrium condition) vs. controls: control (1-1-6); control 2 (1-1-8); CO_2 (1-1-6)

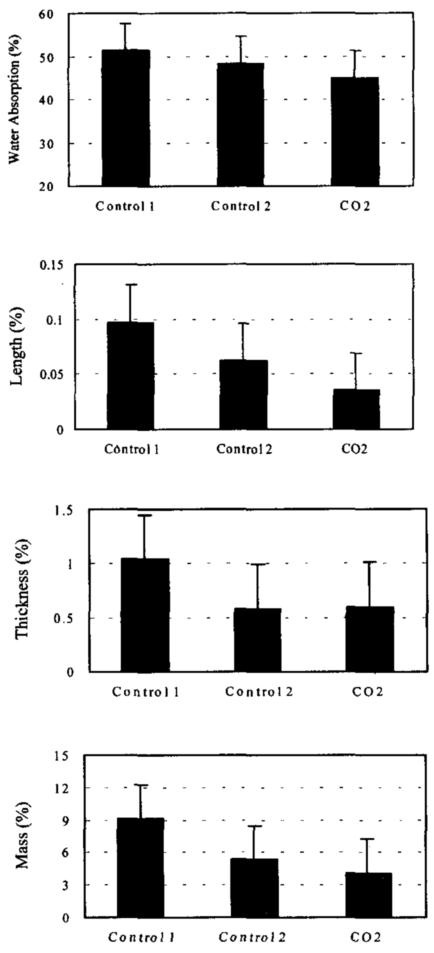


Fig. 8 Water absorption and dimensional stability test results

percentile changes in length, thickness, and mass as relative humidity is increased from 30% to 90%. Fig. 8 shows the results of tests on dimensional stability of CO_2-cured and conventional composites. CO_2-cure is observed to yield major improvements in the dimensional stability of the composites.

5. Summary and Conclusions

The main objective of this study was to optimize the production process of cellulose fiber-reinforced cement composites for achieving the best performance characteristics at reasonable cost. The influential variables in the processing of fabricating cellulose fiber-reinforced cement composites were optimized based on response surface analysis technique. Optimization was based on flexural strength, toughness, and stiffness of composites. Due consideration was also given in the optimization process to the cost of raw materials. The optimized composites were then technically evaluated and compared with two control boards manufactured without the CO_2-cure. The conclusions deriven can be summarized as follows:

(1) The flexural test results revealed that oven duration had statistically significant effects at 95% level of confidence on flexural strength. In case of toughness, oven temperature, oven duration, oven duration-CO_2 chamber duration interaction, and CO_2 chamber duration-autoclaved duration interaction had statistically significant effects at 95% level of confidence. In case of stiffness, oven duration, oven duration-CO_2 chamber duration interaction, and CO_2 chamber duration-autoclave duration interaction had statistically significant effects at 95% level of confidence.

(2) Optimum composites were obtained using 50℃ oven temperature, 1-hour oven duration, 1-hour CO_2 chamber duration, and 6-hours autoclaved duration.

(3) CO_2-cure can reduce time and cost in manufacturing cellulose fiber-reinforced cement composites, and improves flexural strength, stiffness, dimensional stability, and water absorption of composite. Flexural toughness tends to be somewhat reduced with CO_2-cure. However, the expected improvements in resistance to aging effects (carbonation) may yield improvements even in toughness after aging.

References

1. ASTM C 1185-92, "Standard Test Methods for Sampling and Testing Non-Asbestos Fiber-Cement Flat Sheet, Roofing, and Siding Shingles, and Clapboards."
2. Coutts, R.S.P., "Sticks and Stones," Forest Products Newsletter, CSIRO Division of Chemical and Wood Technology (Australia), Vol.2, No.1, January, 1986, pp.1-4.
3. Cook, D.J., "Concrete and Cement Composites Reinforced with Natural Fibers," Concrete International, 1980: Symposium Proceeding on Fibrous Concrete in London, Construction Press, New York, 1980, pp.99-114.
4. Harper, S., "Developing Asbestos Free Calcium Silicate Building Boards," Composites, 13(2), 1980, pp.123-128.
5. Andonian, R., Mai, Y.W., and Cotterell, B., "Strength and Fracture Properties of Cellulose Fiber Reinforced Cement Composites," The International Journal of Cement Composites, Vol.1, No.3, 1979, pp.157-168
6. Sanderman, W., "Research Results of a Wood-Cement System," Translation from

Chemie und Chemische Technol. Mineralgebundener Holzwerkstaff, Internationale Arbeitstagung, 1969, pp.5-21; also cited in Forest Abstract 31:5559

7. Geimer, Robert L., "Method of Pressing Reconstituted Lignocellulosic Materials," US Patent No.4393019, July 12, 1983.
8. Suchsland, Otto and Woodson, Geoge E., "Fiberboard Manufacturing Practices in the United States," Forest Products Research Society, 1991, pp. 263.