Pergamon

Available online at www.sciencedirect.com

SCIENCE DIRECT®

Cement and Concrete Research 33 (2003) 1889–1896

CEMENT AND CONCRETE RESEARCH

Influence of iron oxide pigments on the properties of concrete interlocking blocks

Hyun-Soo Lee[a], Jae-Yong Lee[b,*], Myoung-Youl Yu[a]

[a]*Department of Architecture, Seoul National University, Seoul 151-742, Republic of Korea*
[b]*Division of Architecture, Pukyong National University, Busan 608-739, Republic of Korea*

Received 17 December 2002; accepted 25 June 2003

Abstract

Concrete interlocking blocks (CIBs) are utilized in a variety of commercial, municipal, and industrial applications. Superior engineering properties, low maintenance, ease of placement and removal, reuse of original blocks, aesthetic appeal, and immediate availability are the primary reasons for choosing concrete block pavement over other paving surfaces. It is a common practice to pigment building materials, such as mortar, concrete pavers, concrete roof tiles, and prefabricated concrete products; CIBs are colored using iron oxide pigments. This article presents experimental results detailing the properties of CIBs dyed with pigments. The results of these experiments are as follows: Because the particles of iron oxide pigments are finer than those of brown iron oxide, interlocking blocks mixed with the former acquired higher color strength than with the latter. Additional analysis determined a definite relationship between the flexural strength and the absorption ratio of pigment-dyed blocks; the correlation coefficient (R^2) of interlocking blocks at 91 days was .90. It is suggested that if iron oxide pigments are to be used to color CIBs, the pigment-to-cement ratio should be below 4%.

Keywords: Admixture; Physical properties; SEM; Pigment; Iron oxide

1. Introduction

The use of concrete interlocking block (CIB) pavements has increased markedly in recent years. Concrete block pavement has many advantages, including resistance to freeze–thaw cycles and deicing salts, ease of maintenance and repair, access to utilities, low maintenance costs, and the availability of various shapes and colors that are both functional and aesthetically pleasing [1]. The durability of these blocks is determined by the concrete mix design used to manufacture the blocks.

Nowadays, it is common practice to pigment building materials, such as mortar, concrete pavers, concrete roof tiles, and prefabricated concrete products. Pigments are used for permanent coloring of concrete, that is to dye it a color different from the natural color of the cement or the aggregates [2]. Beginning with the pigmentation of standard building blocks, an ever-increasing variety of colored concrete products are being offered by industry, such as colored split blocks, slump stones, patio stones, paving blocks, screen blocks, fluted blocks, and roof tiles [3].

Pigment powders are significantly finer than cement, therefore, the water-to-pigment ratio is likely to be high; the variations between pigments depend on particle size and shape. The water demand of the pigment, in conjunction with the added rate required to achieve satisfactory color saturation and color durability, defines the influence each pigment has on the block mix design [4]. Pigments used for the integral coloring of concrete are either organic or inorganic compounds and are generally insoluble in water. These pigments are composed of very small particles so they distribute uniformly and permanently within the mass of concrete [5]. Despite the widespread use of these pigments, little research has been conducted on the physical properties of pigment-dyed CIBs [6]. The quality of pigment itself and its effect on the physical properties of pigment-dyed CIB need further study.

This study examines the material properties of pigment-dyed CIBs, such as the flexural strength, absorption, and color strength, with the ultimate goal of providing optimum manufacturing, proper mixing pigment ratios, and usage criteria for pigment-dyed CIBs. The coloring agents used in

* Corresponding author. Tel.: +82-51-620-1393; fax: +82-51-622-8340.

E-mail address: jylee@pknu.ac.kr (J.-Y. Lee).

0008-8846/$ – see front matter
doi:10.1016/S0008-8846(03)00209-6

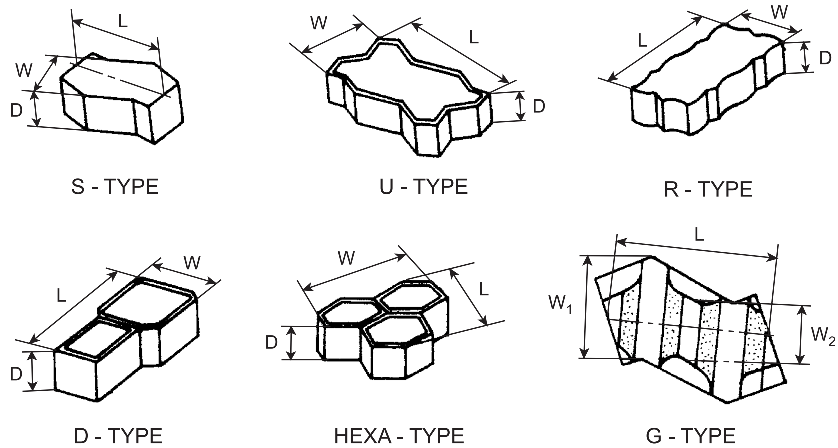

Fig. 1. Shape of modification CIBs.

this experiment were synthetic iron oxide pigment and brown iron oxide, a byproduct from steel manufacturing.

2. Concrete interlocking block

The Korean Industrial Standards Committee [7] classifies CIBs by their size and shape. Basic CIBs are categorized as either I or O type, while a modified block (as shown in Fig. 1) is categorized as S, U, or R type by its intended usage (sidewalks or roads). This experiment described below tested the commonly used U-type blocks, which measured 8.86 × 4.43 × 2.36 in. (see Table 1).

Ordinary Portland cement, sand, stone dust, and pigment are used to manufacture CIBs [8]. A diagram detailing the production process of CIBs is shown in Fig. 2. CIB pavement has several features that differentiate it from asphalt or concrete pavement. CIB can be easily matched

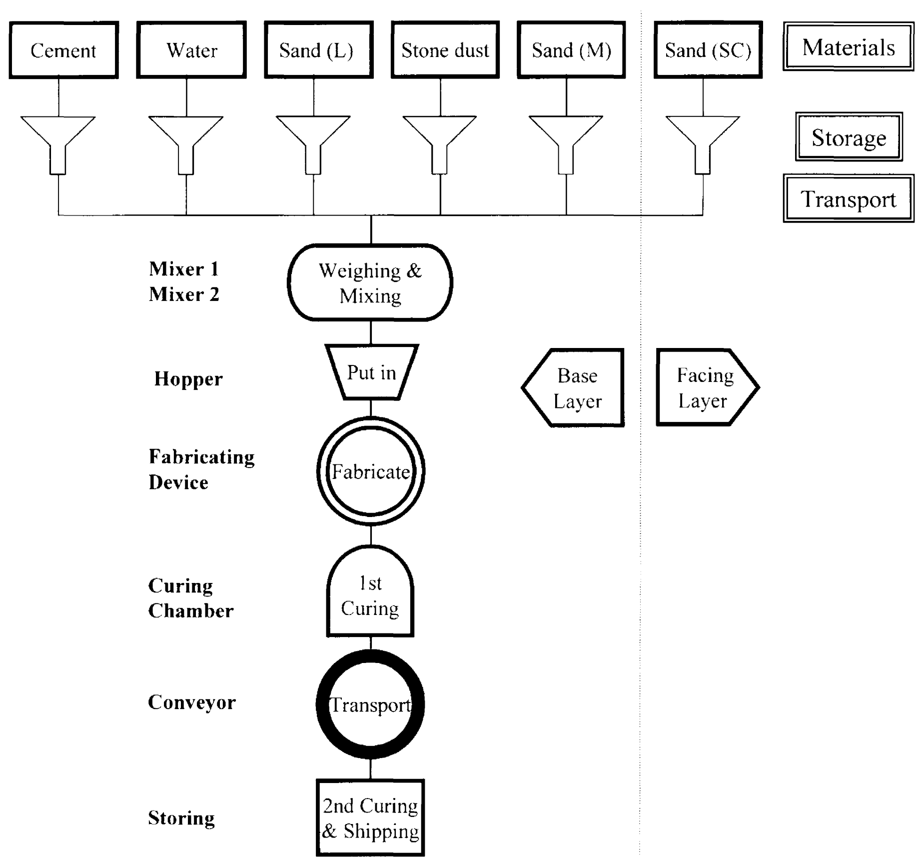

Fig. 2. Production process of CIBs.

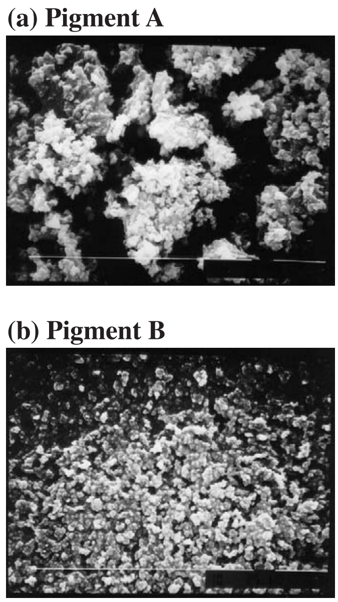

Fig. 3. SEM picture of Pigments A and B (magnifying power × 7500).

to its surroundings and, in most cases, it can be installed without any special skill or heavy equipment. Roads or sidewalks paved with CIB can be use immediately after placement.

3. Experimental procedure

To measure the flexural strength, absorption, and coloring strength of CIB after being mixed with iron oxide, a base layer fabricated from first mixing and a facing layer fabricated from second mixing were used. Colored blocks were made with pigment on the facing layer only, and whole colored layer blocks were made with pigment on both the base and facing layers. A total of 80 blocks were fabricated and manufactured for this experiment, 40 partially colored blocks and 40 whole colored layer blocks.

3.1. Materials

Ordinary Portland cement (ASTM Type I) was used for this experiment. Table 2 details the physical properties of the mix. Large size sand (sand L), stone dust, and medium size sand (sand M) were used for the base layer block. Size-controlled sand (sand SC), able to pass through a No. 4, 4.75-mm sieve, was used for the colored surface layer. The physical properties of aggregate used are shown in Table 3. The iron oxides used in this experiment consisted of brown iron oxide (designated Pigment A) and iron oxide pigment (designated Pigment B). Quality standards for iron oxide (KS M 5102, "Iron Oxide") and required physical properties (in accordance with KS M 5131, "Testing Methods for Pigments") are shown in Table 4.

3.2. Mixing design, fabrication, and curing of test blocks

To ensure that the material properties of the experimental interlocking block were identical to that of an untreated interlocking block, the standard mixing proportion table according to the manufacturer of untreated interlocking block was examined. The water-to-cement ratio and air content was under 25% and 20%, respectively. Therefore, it was determined that Pigment A would consist of 3%, 6%, 8%, and 10% and that of Pigment B would consist of 3%, 4%, and 5%, respectively, depending on the pigment mixing ratio (pigment-to-cement ratio = P/C). The mixing of the base and colored layers was prepared according to the weight method, as shown in Tables 5 and 6). The base layer was mixed with an aggregate weight ratio of 1:6. In case of aggregate, the weight ratio of sand L/sand M/stone dust was 1:2.7:2.3, respectively. The weight ratio of cement to sand SC in mixing the colored layer was 1:5.

Using a MASA 6000VB, the fabrication time of the base layer for the first mix was 0.3 s and that of colored layer for second mix was 0.2 s. These layers were automatically pressured with a total main frequency of 7 s until the block reached a height of 2.36 in. (60 mm). The CIB concrete was stream curing for 3 h in steam chamber, where maximum temperature was controlled under 65 °C (149 °F) after fabrication in accordance with KS F 4419. After 3 h of curing, the specimens were exposed to the environment until the testing date.

3.3. Flexural strength and absorption test

Three blocks from each mixing ratio were tested for flexural strength and absorption according to KS F 4419. The specimen took the span at 5.5 in. (140 mm) and applied the load at the center. The breaking load was defined as the maximum load indicated by the testing machine. Two test specimens underwent the flexural strength test. The test pieces were placed into the air dryer at a constant temperature of 110 ± 5 °C (230 ± 41 °F). Once the specimens had

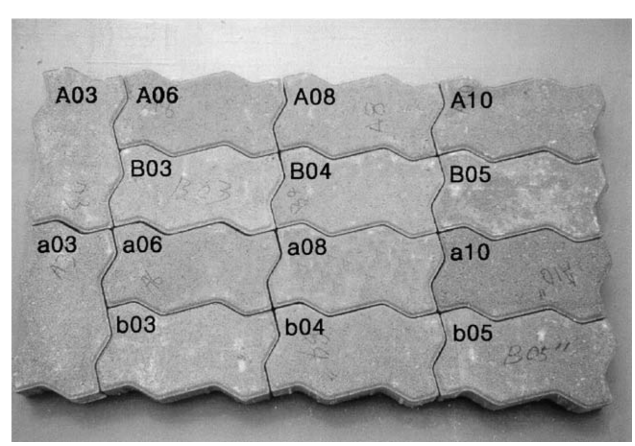

Fig. 4. Tinting strength comparison of Pigments A and B.

been dried for more than 24 h, they were removed from the dryer, weighed down to 10 g (designated as the dry weight, or W_1). Next, the specimens were submerged in water for 24 h, removed, and then weighed (designated as the wet weight, or W_2). The absorption percent was obtained using the following formula: $[(W_2 - W_1)/W_1] \times 100$.

4. Results and analysis

4.1. Mixing effect of pigments

Fig. 3 shows a scanning electron micrograph (SEM) taken of the coloring agents to be studied in this experi-

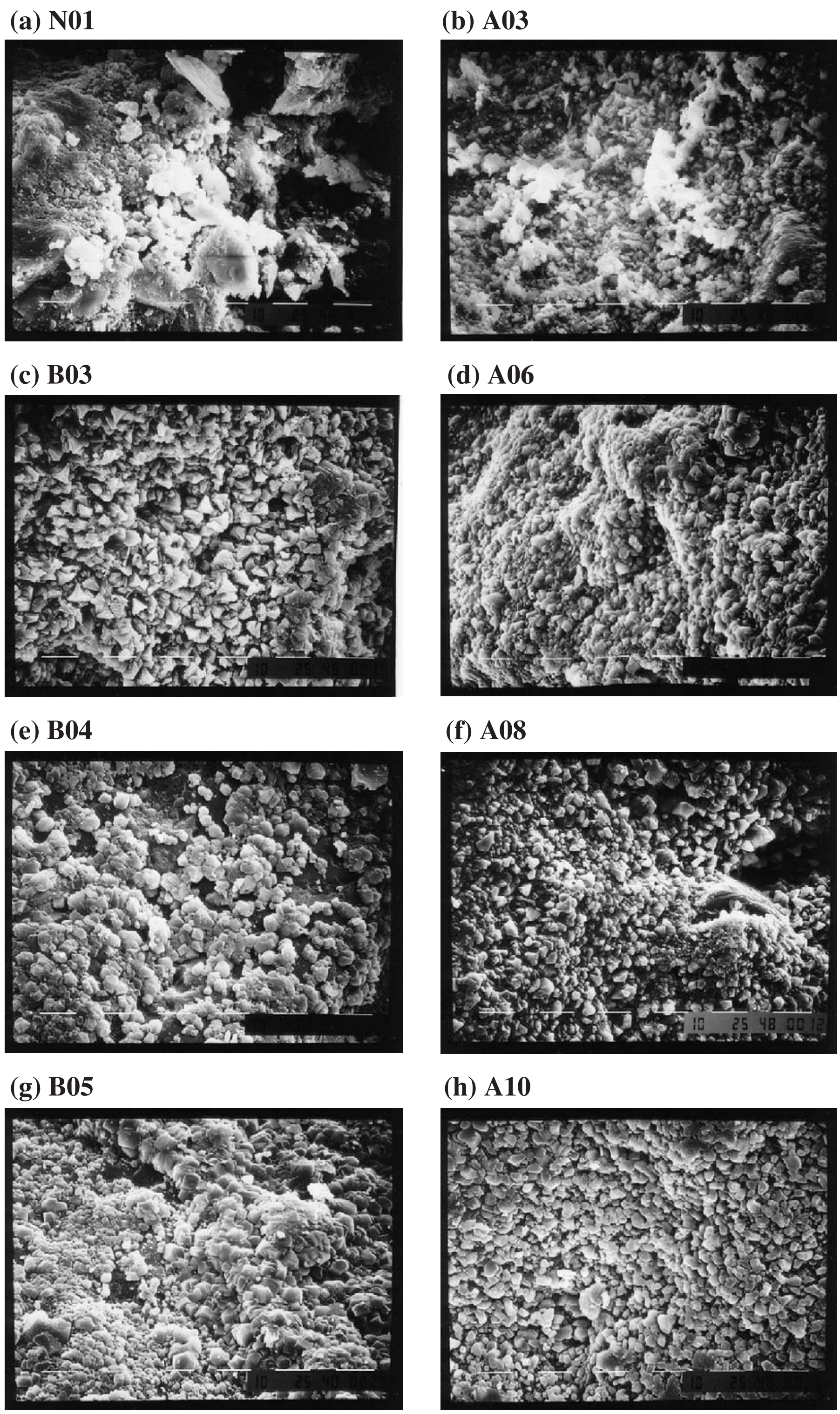

Fig. 5. Internal structure per type of block (magnifying power × 1000).

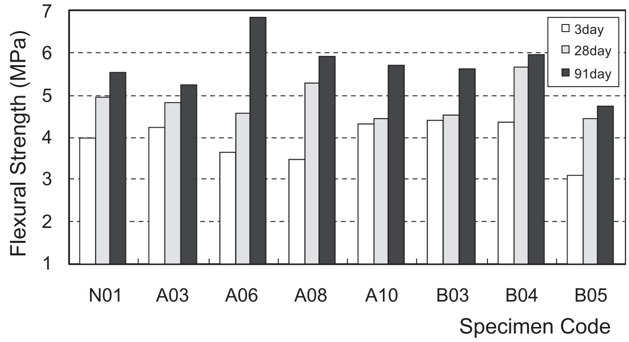

Fig. 6. Flexural strength of partially colored block.

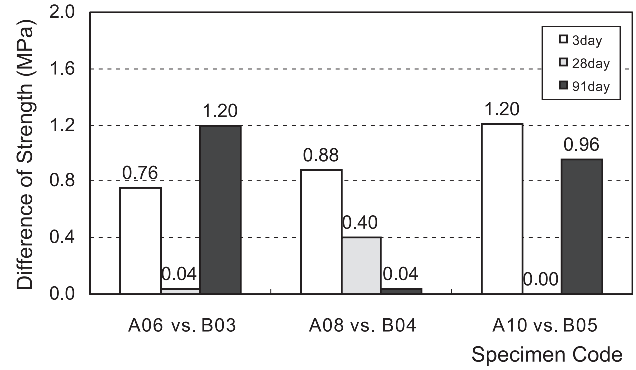

Fig. 8. Strength of mixing ratio and type of pigment.

ment: brown iron oxide (Pigment A) and iron oxide pigment (Pigment B). The particle size and shape of Pigment B was found to be more spherical and finer than those of Pigment A, although the amount of particles in the two specimens that could not pass through a No. 325 sieve were almost similar. Based on these results, it was determined that pigments finer in diameter than Portland cement provide excellent color in the cement matrix. In addition to being preferable to Pigment A in terms of size and shape, Pigment B had no adverse effect on the chemical and physical process of manufacturing concrete and may play a role as filler in concrete.

As for the pH level, Pigment A did not comply with KS M 5102. This pigment is an iron processing byproduct, produced from manufacturing steel products that use hydrochloric acid. The high pH (3.2) clearly shows that the purification process was not properly conducted in the course of processing. Accordingly, removal of hydrochloric acid is necessary to comply with KS M 5102 if brown iron oxide is to be used for dyeing CIBs.

With regard to quantifying the performance of the pigments used, tinting power and color intensity depended on the purity and particle size of the pigment. Relative tinting power was judged by comparing the amount of the two pigments necessary to achieve more or less similar color intensity. Fig. 4 compares CIBs with different pigment ratios. The color intensity of the blocks was compared visually. It proved too difficult to compare the mixed color of CIBs with the standard color chip, which is generally used as the color mark standard.

As shown in Fig. 4, A06, containing 6% Pigment A, attained a similar color to that of B03. Blocks A08 and B04 and A10 and B05 were also similar in color. Clearly, Pigment B colors more effectively than Pigment A. As for the amount of pigment needed to achieve the same color, the amount of synthetic iron oxide pigment was approximately half that of the brown iron oxide. This result supports the current practice of block manufacturers, whereby only half the amount of iron oxide pigment is needed to produce similarly colored blocks as compared to blocks manufactured with brown iron oxide.

Fig. 5 shows the internal structure of different pigment mixing ratios of pigments. Note that the internal structure of blocks mixed with pigment is denser than those blocks without pigment (N01); the internal structure became denser as the mixing ratio is increased. This is most likely due to what is called the *microfiller effect*, whereby the size and shape of the pigment particle is finer and the specific surface area is larger than cement [9].

4.2. Results of flexural strength test

Fig. 6 shows the average flexural strength of a colored block fabricated by combining the colored and base layers.

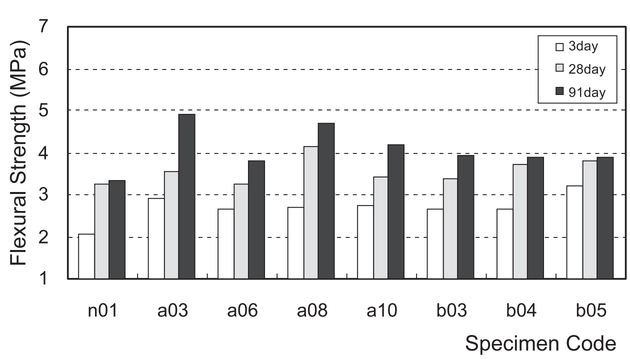

Fig. 7. Flexural strength of whole colored block.

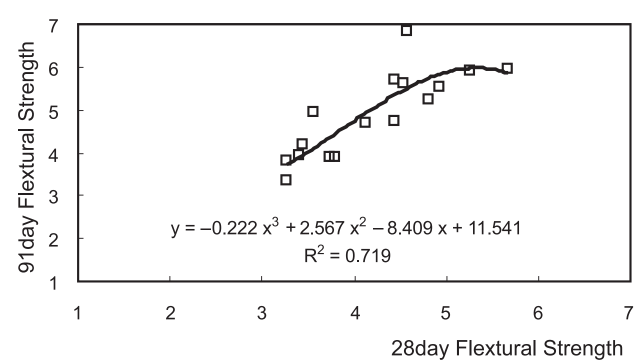

Fig. 9. Correlation of flexural strength at Days 28 and 91.

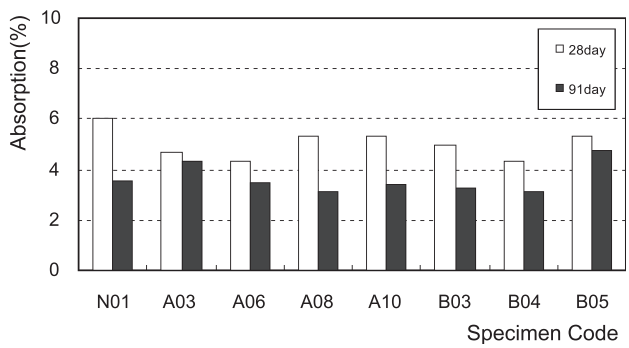

Fig. 10. Change of absorption of colored block per age.

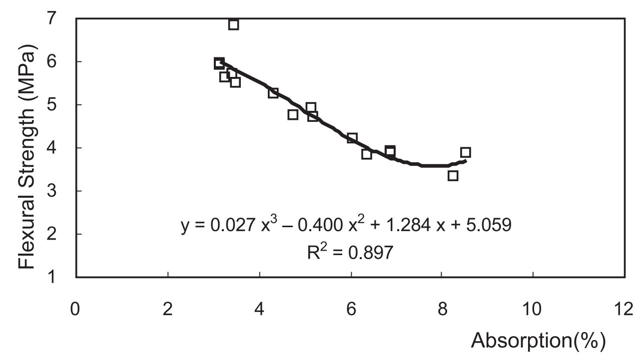

Fig. 12. Absorption of flexural strength at Day 91.

Fig. 7 shows the average of flexural strength of whole colored block. The flexural strength do partially colored blocks at 3 days N01 (without the pigment), Pigment A, and Pigment B were 3.99 MPa 3.48–4.31 MPa, 3.10–4.41 MPa, respectively (see Fig. 6), showing very little difference in flexural strength. By Day 28, N01, Pigment A, and Pigment B were 4.94, 4.44–5.27, and 4.44–5.67 MPa, respectively. A10 and B04 demonstrated a minimum value of 4.44 MPa and a maximum value of 5.67 MPa. Maximum flexural strength of CIBs appeared in A08 and B04. The flexural strength of a CIB specified in KS F 4419 is over 4.9 MPa for sidewalks and 5.9 MPa for roads, thus the flexural strength at 28 days did not comply with the specifications.

The flexural strength at Day 91 of A06 and B05 recorded a maximum value of 6.83 MPa and minimum value of 4.75 MPa, respectively. Therefore, the curing period should be increased or the blocks delivered for installation only after tested for flexural strength. When combining Pigments A and B, a maximum strength was obtained with a mixing ratio of 6–8% and 4%, respectively; it is suggested that for optimum performance, this mixing ratio be used in the case of the red interlocking block.

The increase in strength by Day 28 was 24% on average compared with the age of 3 days. By Day 91, strength improved by 18% on average compared with Day 28. Flexural strength of a whole colored layer block is shown in Fig. 7, which did not meet specification KS F 4419. In case of the colored block, the strength of the 1.97-in. (50-mm)-thick base layer was remarkably higher than that of 0.39-in.-thick colored facing layer. However, in case of whole colored block, as the colored layer with low strength is located at the bottom of block, the flexural strength was considerably declined, compared with the partially colored block.

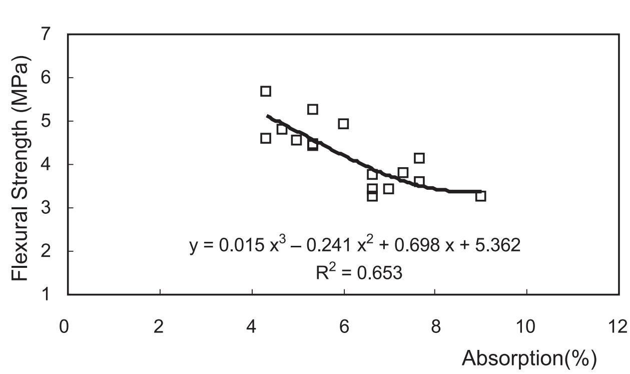

Fig. 11. Absorption of flexural strength at Day 28.

Fig. 8 shows the absolute value of flexural strength of the CIBs difference per mixing ratio and pigment type on similar color intensity. It was observed that there was little difference in the strength of Blocks A08 and B04 as a function of age. In addition, the flexural strength of blocks A10 and B05 was higher in blocks using Pigment A. In sum, the strength difference at Day 28 was − 0.40–0.04 MPa for Blocks A06 and B03, A08 and B04, and A10 and B05, and by Day 91 was − 0.04–1.20 MPa. Therefore, blocks with similar mixing ratios and color effects showed little difference in strength between Pigments A and B. Fig. 9 shows the correlation of flexural strength at Days 28 and 91. Although the correlation coefficient (R^2) at Day 28 is relatively high (.72), there is not sufficient data to estimate long-term strength.

4.3. Results of absorption test

The absorption ratio of concrete continues to increase due to the freezing and thawing phenomenon resulting from weather changes. Because the absorbed water can cause cracking and efflorescence of concrete, control of the

Table 1
Size of CIB (U type)

Position	Length	Width	Depth	Colored layer[a]
Size				
In inches	8.86	4.43	2.36	39
In millimeters	225	112.5	60	10

[a] In case of coloring the facing layer of block only, in colored block, the thickness of colored layer should be + 0.39 in. (10 mm) in principle, + 0.32 in. (8 mm) at the minimum. No cracks should appear after flexural strength test.

Table 2
Physical properties of cement

Specific gravity	Blaine value	Soundness (%)	Compressive strength (MPa)		
			3 days	7 days	28 days
3.15	3346	0.25	22.1	30.1	37.7

Table 3
Physical properties of fine aggregate

Division	Sand L	Stone dust	Sand M	Sand SC
Specific gravity	2.56	2.58	2.57	2.58
Fineness modulus	3.69	4.13	3.16	3.15
Sieve passing percentage (%)				
No. 4	94	72	98	100
No. 8	76	45	88	92
No. 16	42	31	68	71
No. 30	12	18	24	18
No. 50	5	12	4	3
No. 100	2	9	2	1

absorption ratio is critical. Fig. 10 shows the absorption at Days 28 and 91. The absorption rate at Day 28 for Block N01 was 6.0%, Pigment A was 4.3–5.3% (4.9%, on average), and Pigment B was 4.3–5.3% (4.9% on average). The absorption rate at Day 91 for Block N01 was 3.5%, for Pigment A between 3.1% and 4.3% (3.6% on average), and for Pigment B between 3.1% and 4.8% (3.7% on average). All specimens satisfied the range of absorption specified in KS F 4419, that is, under 7% on average and under 10% for each block. It should be noted that ASTM C-936 [10] specifies the maximum absorption value as 5% in locales where freezing and thawing cycles occur. Recent research [11] suggests that it is more advantageous to set the absorption of CIBs under 3.8% to obtain adequate durability against freezing and thawing cycles.

This finding suggests that testing be done on a variety of manufacturer's blocks to determine their durability to freezing and thawing cycles. All blocks in this experiment satisfied ASTM C-936 at Day 91; however, Blocks A03 (4.3%) and B05 (4.8%) failed to satisfy the specified value of under 3.8%. Accordingly, it is suggested that the standard of absorption specified in KS F 4419 should be verified by further research that includes freezing and thawing experiments on blocks manufactured with various mixing ratios.

Table 4
Physical properties of iron oxide pigment

Test item	Quality standards (KS M 5102)	Type of pigment	
		A	B
Fe_2O_3 (%)	over 92.0	99.3	98.7
Ig.loss (%)	under 2.5	0.91	0.94
Water-soluble salts (%)	under 0.8	0.1	0.4
pH	5.0–8.0	3.2	6.6
Sieve residue on No. 325 (%)	under 0.5	0.28	0.26

Table 5
Mixture proportion of base layer

W/C (%)	Mixture content (kg/m^3)				
	Cement	Sand L	Sand M	Stone dust	Water
25	276	276	734	644	69

Table 6
Mixture proportion of colored layer

Specimen code	W/C (%)	P/C (%)	Mixture content (kg/m^3)			
			Cement	Sand SC	Pigment	Water
N01	20	0	325	1625	0	65
A03	20	3	325	1625	9.75	65
A06	20	6	325	1625	19.5	65
A08	20	8	325	1625	26.0	65
A10	20	10	325	1625	32.5	65
B03	20	3	325	1625	9.75	65
B04	20	4	325	1625	10.6	65
B05	20	5	325	1625	16.25	65

Based on the colored layer to use second mixing, the pigments are classified as A and B depending on the type of pigment used. Specimens without pigment are marked as N. This code also shows the pigment ratio (P/C). If the whole colored layer block is manufactured only with second mixing, it was marked as small letter (e.g., specimen name of whole colored layer with 3% of brown iron oxide 3% was marked as A03).

In case of the whole colored/layered block, the absorption ratio of Block N01 at Day 28 was 9.0% and that containing mixing pigment was between 6.7% and 7.7%. At Day 91, N01 recorded an absorption ratio of 8.3% and that with pigment recorded a ratio of 5.1–8.5%; these values are slightly higher than that specified by the Korean Standard. In brief, although the whole colored/layered block met the specification for the absorption ratio, it does not meet the specification for flexural strength.

4.4. Relationship between flexural strength and absorption

At present, KS F 4419 specifies the flexural strength and absorption ratio only in terms of CIBs. The correlation between absorption and flexural strength in this experiment is shown in Figs. 11 and 12. The correlation coefficient (R^2) between the absorption ratio and flexural strength at Day 28 was .65; at Day 91, however, it was considerably higher—.90.

5. Conclusions

This study measured the flexural strength, absorption capability, and color saturation of CIBs used for sidewalks and roads after incorporating synthetic iron dioxide pigment and brown iron oxide. The following results were obtained:

1. Synthetic iron oxide pigment (Pigment B) was found to perform in a superior manner in terms of shape and size of particle to that of brown iron oxide (Pigment A). If

brown iron oxide is used, it should have a pH between 5.0 and 8.0, as specified in KS M 5102.

2. Comparing the color saturation of the CIBs with the Pigment A versus Pigment B, the iron oxide pigment blocks were found to have two times the tinting power than that of brown iron oxide (depending on the mixing ratio). It is suggested, therefore, that the proper amount of iron oxide pigment to be used should be half that of brown iron oxide.
3. The flexural strength of colored blocks at Day 28 containing brown iron oxide and iron oxide pigment were 4.44–5.27 and 4.44–5.67 MPa, respectively. The blocks with brown iron oxide showed a low value of 4.44 MPa at a mix ratio of 10%, and the blocks with iron oxide pigment showed a high value of 4.75 MPa at a mix ratio of 4%. The flexural strength at Day 91 for the blocks with brown iron oxide and iron oxide pigment had a high value of 6.83 MPa and a low value of 4.75 MPa at 6% and 5%, respectively. The change in strength as a result of the concentration of pigment was as follows: The brown iron oxide and iron oxide pigment showed the highest strength at a mix ratio of 6–8% and 4%, respectively; it is suggested that this mix ratio be used to achieve optimum performance levels.
4. Absorption rate of the CIBs at Day 91 satisfied specifications KS F 4419 (under 7%) and ASTM C-936 (under 5%). It failed, however, to meet the specification of under 3.8% suggested in the recent research by Ghafoori and Mathis [11] to obtain optimum durability against freezing and thawing cycles. The current experiment incorporated brown iron oxide at mix ratio of 3% and iron oxide pigment at a mix ratio of 5%, resulting in an absorption rate of 4.3% and 4.8%, respectively. Additional research is necessary to determine the ideal proportion of pigment concentration and absorption ratios on a variety of manufactured CIBs.
5. The correlation coefficient (R^2) between the flexural strength and absorption ratio of CIB at Day 91 was .90 (quite high), thus, absorption is considered a major factor in influencing the flexural strength of CIBs.

References

[1] R.R. Gonzalo, D.R. Smith, J.S. Miller, M.W. Witczalc, Structural design of concrete block pavements, J. Transp. Eng., ASCE 116 (5) (1990) 615–635.

[2] G.E. Kurz, A.E. Jungk, The use of coloured concrete in creative design, Proc. 3rd Int. Conf. Concr. Block Paving, Rome (1988) 37–40.

[3] T.C. Patton, Pigment Handbook, Wiley, New York, 1973.

[4] S.M. Bruce, G.H. Rowe, The influence of pigments on mix designs for block paving units, Proc. 4th Int. Conf. Concrete Block Paving 2 (1992) 117–124.

[5] P. Bartos, Fresh Concrete Properties and Test, Elsevier, Amsterdam, 1992.

[6] N. Anthony, S. Beaty, G.P. Raymond, Concrete block road paving, Concr. Int. 17 (5) (1995) 36–41.

[7] Korean Industrial Standards Committee, Concrete Interlocking Block for Side Walk and Road, KS F 4419, Korean Standards Association, Yeoksam-dong, Gangnam-gu, Seoul South Korea, 1995.

[8] Construction Material Institute, Construction Material Handbook, Construction Material Institute, Japan, 1996.

[9] G. Von Szadkowski, The effect of pigments on the quality of concrete blocks, Proc. 1st Int. Conf. CBP, Newcastle-Upon-Tyne, England (1980) 155–156.

[10] ASTM Committee, Standard Specification for Solid Interlocking Concrete Paving Units, ASTM C 936-82, 1989 Annual Book of ASTM Standards, ASTM, West Conshohocken, PA, 1989.

[11] N. Ghafoori, R. Mathis, Prediction of freezing and thawing durability of concrete paving blocks, J. Mater. Civil Eng., ASCE 10 (1) (1998) 45–51.