

Center for By-Products Utilization

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Report No. CBU-2003-03
REP-496
January 2003

A manuscript submitted to *Cement and Concrete Research*, January 2003.

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Use of pulp and paper mill residual solids in production of cellucrete

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Abstract

Fibrous residuals generated from pulp and paper mills and de-inking mills were included in concrete. Carefully proportioned concrete mixtures containing some of the residuals showed higher compressive and splitting tensile strengths than concrete without the residuals. Overall, a high correlation was observed between density and strength of concrete containing the residuals. By achieving equivalent density, concrete containing the residuals may be produced that is equivalent to concrete without residuals in strength.

Keywords: Compressive strength (C); Fiber reinforcement (E); Paper mill solid residuals; Tensile properties (C); Waste management (E)

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1. Introduction

Pulp and paper mill wastewater treatment plant residuals are the solid residue removed from mill wastewater before the water is discharged into the environment or reused in the mill. Residuals are removed via a two-step process of treating the wastewater [1,2,3,4].

Primary residual is the solids removed from the primary clarifier. Primary clarification is usually carried out by sedimentation and sometimes by dissolved air flotation. Primary residual consists mainly of cellulose fibers, papermaking fillers (kaolinitic clay, calcium carbonate, and/or titanium dioxide), and moisture. In some cases, ash generated at mill and inert solids rejected during chemical recovery processes become part of the primary residual. The water clarified by the primary treatment is passed on to the secondary treatment.

Secondary treatment is usually a biological process in which micro-organisms convert soluble organic matter to carbon dioxide and water while consuming oxygen. Secondary residual is mainly microbial biomass (called also biosolids) grown during this process and removed through clarification.

Many times primary and secondary residuals are combined to facilitate handling. In most cases, the residuals are dewatered before disposal or beneficial use.

In 1995, pulp and paper industry in the U.S. generated about 5.3 million metric tons of mill wastewater treatment residuals (on dry basis), which is equivalent to about 15 million metric tons of dewatered residuals. About half of this was disposed in landfills/lagoons, a quarter was burned,

one-eighth was applied on farmland/forest, one-sixteenth was reused/recycled in mills, and the rest one-sixteenth was used in some other ways [5].

Due to increasing cost of landfilling, increasingly stringent environmental regulations, and potential long-term environmental liabilities, the percentage of the residuals disposed in landfills has decreased considerably over the years [1,2,5]. However, still significant amount of residuals need to be diverted from landfilling.

Numerous projects have been conducted on the use of cellulose fibers as reinforcement in pressed cement and/or mortar sheets. Cellulose fiber reinforced cement-based sheet composites showed considerably higher flexural toughness than plain cement-based sheets [6,7]. Use of cellulose fibers reduced the extent of shrinkage cracking of mortar [8] and improved the resistance of cement-based composites to freezing and thawing [9].

Concrete is weak in tension (3 to 9 MPa). Wood cellulose fiber is strong in tension (300 to 900 MPa) [10]. Therefore, use of wood fibers in concrete should improve the usefulness of concrete. Paper mill residuals could become an economical source of wood fibers for microfiber reinforcement of concrete.

No research results have been reported on the use of pulp and paper mill residuals in structural-grade concrete. This paper reports the results of a research project conducted on the use of the residuals in concrete.

2. Materials

2.1. Cement, Fine and Coarse Aggregates, and Chemical Admixtures

Type I portland cement, fine aggregate (sand), and coarse aggregate (pea-gravel with a 9.5-mm maximum size) supplied by a local ready-mixed concrete producer were used in this research.

The cement and the aggregates met the requirements of ASTM C 150 and C 33, respectively.

High-range water-reducing admixture (HRWRA) used in this research met the requirements of ASTM C 494, Type F (HRWRA).

2.2. Pulp and Paper Mill Residuals

A total of three sources of pulp and paper mill residuals were used (one each from a pulp mill in USA and Canada, and one from a de-ink paper mill in USA). Properties of the residuals are presented in Tables 1 and 2.

2.3. Deflocculation (or “Repulping”) of Residuals

Due to flocculation and dewatering, as-received residuals contained fibrous clumps of wood fibers, kaolin-type clay (if any), and other particulates (if any). These clumps may be considered as weaker spots in concrete compared with well-dispersed individual fibers and particles. Also, in order for the fibers to function efficiently as fibers, they must be separated into individual fibers as much as possible.

Therefore, the residuals were deflocculated, or “repulped”, into separated wood fibers and particulates before their addition to a concrete mixture. The “pulper” used for this purpose in the laboratory consisted of a 19-liter plastic bucket and a high-speed mixer with a rotor positioned above the bottom of the bucket. Mechanical repulping was performed by immersing the residuals

in room-temperature water with a prescribed amount of HRWRA in the bucket and subjecting the mixture to a high-speed rotation by the rotor blades for not less than 20 minutes.

3. Mixture proportions, results, and discussions

3.1. Mixture Proportions

Mixture proportions and fresh properties of concrete mixtures produced in the laboratory are shown in Tables 3 and 4. As-received residuals content by mass of concrete ranged from 0.2 to 1.2 % for Residual N and 0.2 to 0.8 % for Residuals F and K. Amount of wood fibers (on dry basis) in concrete ranged from 1.1 to 6.1 kg/m³ for Residual N, 0.9 to 3.3 kg/m³ for Residuals F, and 1.7 to 6.3 kg/m³ for Residual K.

Overall, water-cementitious materials ratio, slump, air content, and density were in the ranges of 0.40 to 0.52, 65 to 260 mm, 1.1 to over 10 %, and 2000 to 2420 kg/m³, respectively. Control Mixture, C, and Mixtures F1 and F2 showed very high slump (235, 255, and 260 mm, respectively). Concrete mixtures containing Residual N showed very high air contents. In general, density of concrete decreased as the amount of residuals increased. Density values of Residual N mixtures were lower than that of Control Mixture. Density values of most of Residuals F and K mixtures were almost as high as Control Mixture.

3.2. Compressive and Splitting Tensile Strengths

Compressive and splitting tensile strengths of concrete were determined at 28 days by testing three 100 x 200 mm cylinders for each test type. Test results are presented in Fig. 1 and 2. Most of the Residuals F and K mixtures were equivalent to Control concrete in compressive strength and splitting tensile strength. Within each group of Residuals N, F, and K concrete mixtures,

compressive and splitting tensile strengths generally decreased as the amount of residuals increased.

Mixtures F3 and F4 mixtures showed higher compressive strength than Mixtures F1 and F2, respectively. This may be attributed to lower w/cm of F3 and F4 mixtures compared with F1 and F2 mixtures, respectively. Splitting tensile strength values of Mixtures F3 and F4 were higher than that of Mixture F2 mixture and were nearly equivalent to that of Mixture F1. Compressive strength and splitting tensile strength of concrete showed a very good correlation (Fig. 3).

Overall, compressive and splitting tensile strengths of concrete containing the residuals were proportional to density of concrete (Fig. 4 and 5). To produce residuals-containing concrete that is equivalent to concrete without residuals in strength, density of residuals concrete should be adjusted to approximately match that of the concrete without residuals.

Varying levels of relationships between water-cementitious materials ratio and strengths were observed within each group of concrete mixtures (Control, N, F, and K). However, overall, no close relationships between water-cementitious materials ratio and strengths were observed.

4. Conclusions

Based on these initial results presented, the following general conclusions may be drawn:

(1) In general, within each group of concrete mixtures containing pulp and paper mill residual, density, compressive strength, and splitting tensile strength of concrete decreased with the increase of the amount of the residuals in concrete.

(2) Several concrete mixtures containing the residuals showed higher strength than the concrete without the residuals.

(3) A very good correlation was observed between compressive and splitting tensile strengths of concrete containing pulp and paper mill residuals.

(4) A relatively very good correlation was observed between density and strength of concrete containing the residuals.

(5) Overall, a low correlation was observed between water-cementitious materials ratio and strength of concrete containing the residuals.

(6) By achieving equivalent density of concrete, strength of concrete containing the residuals may be made equivalent to that of concrete without the residuals.

Acknowledgements

The writer expresses deep sense of gratitude to Weyerhaeuser Company, Federal Way, WA for providing its laboratory facilities and financial support for this research.

The UWM Center for By-Products Utilization was established in 1988 with a generous grant from the Dairyland Power Cooperative, La Crosse, WI; Madison Gas and Electric Company, Madison, WI; National Minerals Corporation, St. Paul, MN; Northern States Power Company, Eau Claire, WI; We Energies, Milwaukee, WI; Wisconsin Power and Light Company, Madison, WI; and, Wisconsin Public Service Corporation, Green Bay, WI. Their financial support and additional grant and support from Manitowoc Public Utilities, Manitowoc, WI, are gratefully acknowledged.

References

- [1] J. Unwin, Why bury it when you can use it?: NCASI's support of the industry's efforts to find beneficial uses for solid wastes, in: Proceedings of the 2000 NCASI National Meeting, National Council of the Paper Industry for Air and Stream Improvement, Inc. (NCASI), Research Triangle Park, NC, 2000, pp. 57-74.
- [2] Solid waste management and disposal practices in the U.S. paper industry, Tech. Bull. No. 641, NCASI, New York, NY, 1992.
- [3] G. Scott, A. Smith, Sludge characteristics and disposal alternatives for recycled fiber plants, in: 1995 Recycling Symposium, Technical Association of the Pulp and Paper Industry (TAPPI), Atlanta, GA, 1995, pp. 239-249.
- [4] Alternative management of pulp and paper industry solid wastes, Tech. Bull. No. 655, NCASI, New York, NY, 1993.
- [5] Solid waste management practices in the U.S. paper industry - 1995, Tech. Bull. No. 793, NCASI, Research Triangle Park, NC, 1999.

- [6] K.D. Vinson, J. I. Daniel, Specialty cellulose fibers for cement reinforcement, in: Thin Section Fiber Reinforced Concrete and Ferrocement, vol. SP-124, ACI, Detroit, MI, 1990, pp. 1-18.
- [7] P. Soroushian, S. Marikunte, J. Won, Statistical evaluation of mechanical and physical properties of cellulose fiber reinforced cement composites, ACI Mater. J. 92 (2) (1995) 172-180.
- [8] M. Sarigaphuti, S.P. Shah, K.D. Vinson, Shrinkage cracking and durability characteristics of cellulose fiber reinforced concrete, ACI Mater. J. 90 (4) (1993) 309-318.
- [9] P. Soroushian, S. Marikunte, J. Won, Wood fiber reinforced cement composites under wetting-drying and freezing-thawing cycles, J. Mater. Civ. Eng. 6 (4) (1994) 595-611.
- [10] Fiber reinforced concrete, Portland Cement Association, Skokie, IL, 1991.
- [11] T.R. Naik, Paper industry by-products utilization, Rept. No. CBU-1989-02, UWM Center for By-Products Utilization, Dept. of Civ. Eng. and Mech., Univ. of Wisconsin-Milwaukee, 1989.
- [12] T.R. Naik, Use of residuals in production of cellucrete, a report prepared for the Weyerhaeuser Company, Tacoma, WA, 1998.
- [13] T.R. Naik, R.N. Kraus, Development of concrete utilizing paper mill residual solids, Rept. No. CBU-1998-13, UWM Center for By-Products Utilization, Dept. of Civ. Eng. and Mech., Univ. of Wisconsin-Milwaukee, 1998.

Table 1. Types, physical properties, LOI at 590°C, and wood fiber content of residuals

| Residuals | Type of Residuals | Type of Mill | Fiber Origin | Moisture Content (%) [*] | Avg. Fiber Length [‡] (mm) | Loss On Ignition at 590°C (%) [*] | Wood Fiber Content (%) [*] |
|-----------|-------------------|--------------|--------------|-----------------------------------|-------------------------------------|--|-------------------------------------|
| N | Primary | Paper | Recycled | 87 | 1.46 | 42 | 46 |
| F | Primary | Pulp | Virgin | 125 | ... [†] | 41 | 40 |
| K | Primary | Pulp | Virgin | 138 | 1.81 | 78 | 83 |
| Avg. | ... | | ... | 117 | 1.64 | 54 | 56 |

* % of oven-dry (105°C) mass of residuals.

‡ Length weighted average fiber length, $L_L = \frac{\sum_{i=1}^N n_i l_i^2}{\sum_{i=1}^N n_i l_i}$, using Kajaani FS-200.

† Not available. “Could not run... due to shives and/or large particles.”

Table 2. Oxides composition of ash left after ignition of residuals at 1000°C

| Residuals | | N | F | K | Avg. |
|-------------------------|--------------------------------|------|------|------|------|
| Ash (%) [*] | | 36.1 | 24.7 | 17.7 | 26.2 |
| Oxides (%) [†] | SiO ₂ | 47.6 | 33.4 | 53.3 | 44.8 |
| | Al ₂ O ₃ | 37.0 | 2.5 | 9.5 | 16.3 |
| | CaO | 9.4 | 57.5 | 13.2 | 26.7 |
| | MgO | 2.0 | 1.6 | 11.0 | 4.9 |
| | Fe ₂ O ₃ | 0.8 | 1.6 | 4.7 | 2.4 |
| | TiO ₂ | 4.9 | 0.1 | 0.4 | 1.8 |
| | K ₂ O | 0.4 | 0.7 | 4.4 | 1.8 |
| | Na ₂ O | 0.7 | 1.4 | 2.4 | 1.5 |

* % of oven-dry (105°C) mass of residuals.

† % of mass of ash (1000°C).

Table 3. Mixture proportions and fresh concrete properties (Control, N)

| Mixture Name | C | N1 | N2 | N3 | N4 | N5 | N6 | N7 |
|---|------|------|------|------|------|------|------|------|
| Residuals, as-recd (% of conc. by mass) | 0 | 0.2 | 0.4 | 0.5 | 0.6 | 0.7 | 0.8 | 1.2 |
| Wood Fibers from Residuals (kg/m ³)* | 0 | 1.1 | 2.3 | 2.6 | 3.0 | 3.4 | 3.9 | 6.1 |
| Residuals, as-recd (kg/m ³) | 0 | 4.5 | 9.2 | 10.5 | 12.3 | 13.8 | 16.0 | 24.7 |
| HRWRA (L/m ³) | 2.3 | 3.4 | 3.5 | 4.9 | 6.9 | 6.0 | 7.3 | 12.2 |
| Cement (kg/m ³) | 357 | 334 | 340 | 313 | 307 | 295 | 300 | 296 |
| Sand, SSD (kg/m ³) | 848 | 790 | 801 | 743 | 718 | 697 | 710 | 691 |
| Pea Gravel, 9,5-mm max., SSD (kg/m ³) | 1050 | 978 | 994 | 921 | 899 | 866 | 877 | 866 |
| Water (kg/m ³) | 166 | 138 | 149 | 128 | 132 | 122 | 125 | 134 |
| <i>w/cm</i> | 0.46 | 0.41 | 0.44 | 0.41 | 0.43 | 0.41 | 0.42 | 0.45 |
| Slump (mm) | 235 | 185 | 85 | 165 | 125 | 135 | 110 | 70 |
| Air Content (%) | 1.1 | 8.6 | 5.2 | > 10 | > 10 | > 10 | ... | > 10 |
| Density (kg/m ³) | 2420 | 2250 | 2300 | 2120 | 2070 | 2000 | 2030 | 2020 |

* On dry basis.

Table 4. Mixture proportions and fresh concrete properties (F, N)

| Mixture Name | F1 | F2 | F3 | F4 | K1 | K2 | K3 | K4 |
|---|------|------|------|------|------|------|------|------|
| Residuals, as-recd (% of conc. by mass) | 0.2 | 0.4 | 0.6 | 0.8 | 0.2 | 0.4 | 0.6 | 0.8 |
| Wood Fibers from Residuals (kg/m ³)* | 0.9 | 1.7 | 2.5 | 3.3 | 1.7 | 3.3 | 4.8 | 6.3 |
| Residuals, as-recd (kg/m ³) | 4.8 | 9.5 | 14.1 | 18.4 | 4.8 | 9.6 | 13.8 | 18.1 |
| HRWRA (L/m ³) | 3.6 | 3.6 | 3.6 | 3.5 | 5.9 | 5.7 | 6.2 | 6.6 |
| Cement (kg/m ³) | 356 | 353 | 352 | 346 | 359 | 356 | 344 | 339 |
| Sand, SSD (kg/m ³) | 829 | 802 | 822 | 796 | 829 | 824 | 797 | 771 |
| Pea Gravel, 9,5-mm max., SSD (kg/m ³) | 1047 | 1033 | 1033 | 1006 | 1049 | 1040 | 1003 | 994 |
| Water (kg/m ³) | 161 | 183 | 141 | 159 | 169 | 167 | 163 | 172 |
| <i>w/cm</i> | 0.45 | 0.52 | 0.40 | 0.46 | 0.47 | 0.47 | 0.47 | 0.51 |
| Slump (mm) | 255 | 260 | 140 | 145 | 110 | 65 | 100 | 140 |
| Air Content (%) | 1.2 | 1.4 | 3.5 | 4.7 | 2.2 | 2.0 | 3.7 | 3.5 |
| Density (kg/m ³) | 2400 | 2380 | 2360 | 2330 | 2410 | 2400 | 2320 | 2300 |

* On dry basis.

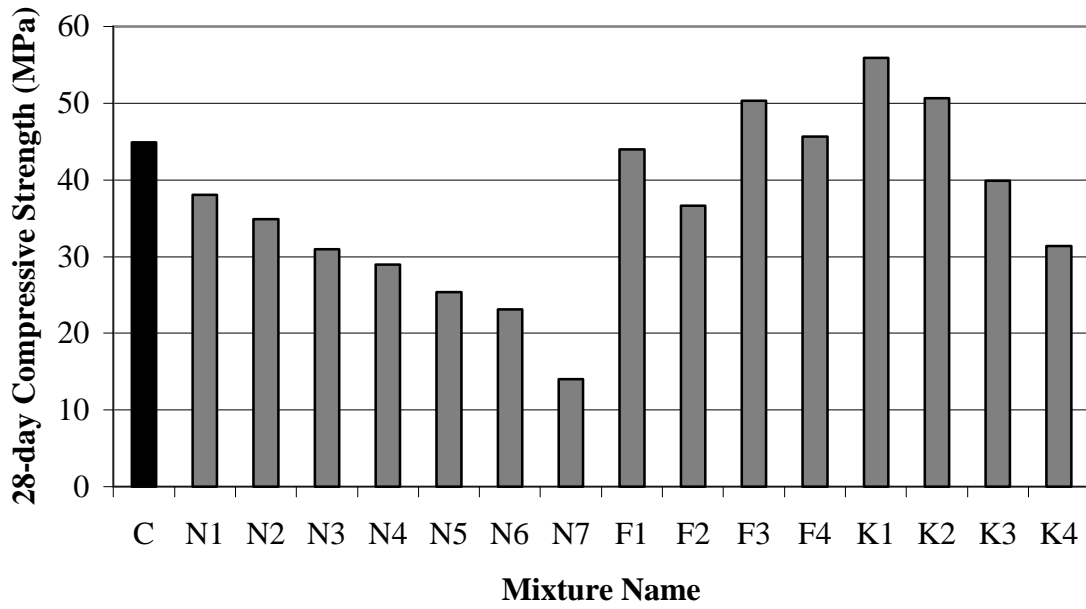


Fig. 1. Compressive strength of concrete at 28 days

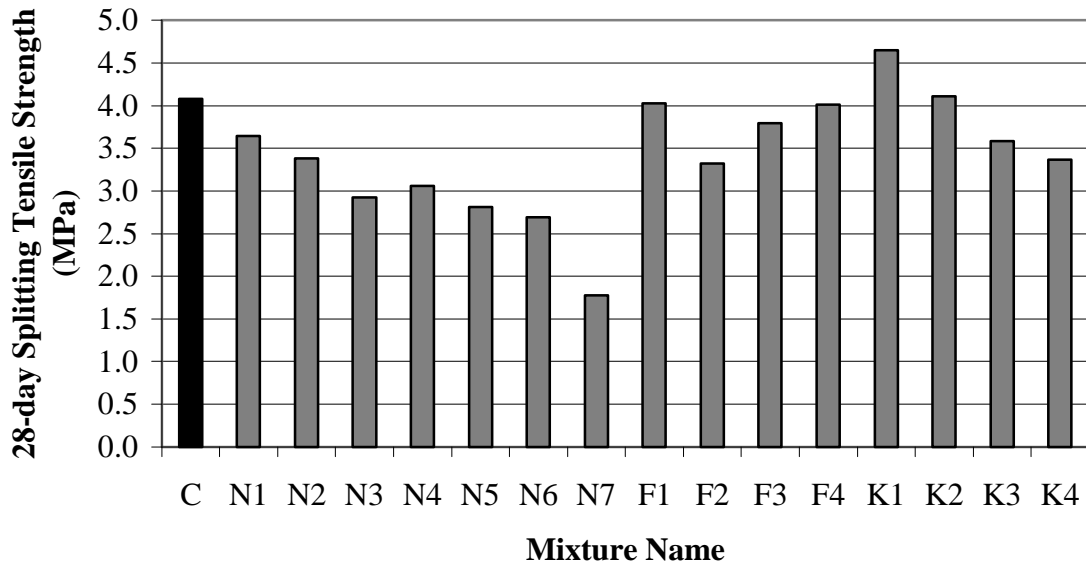


Fig. 2. Splitting tensile strength of concrete at 28 days

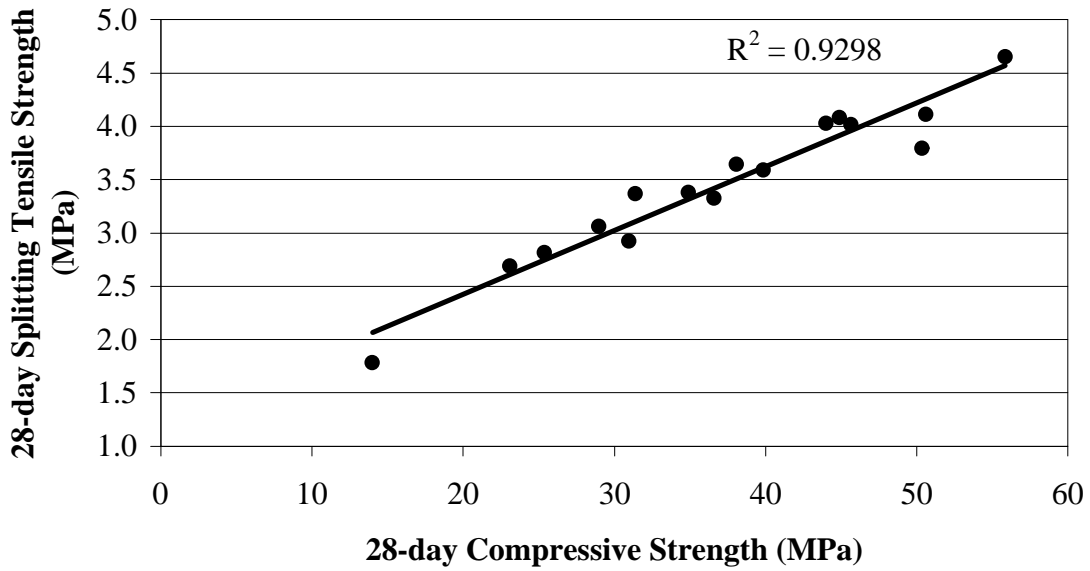


Fig. 3. Relation between 28-day compressive and splitting tensile strengths of concrete

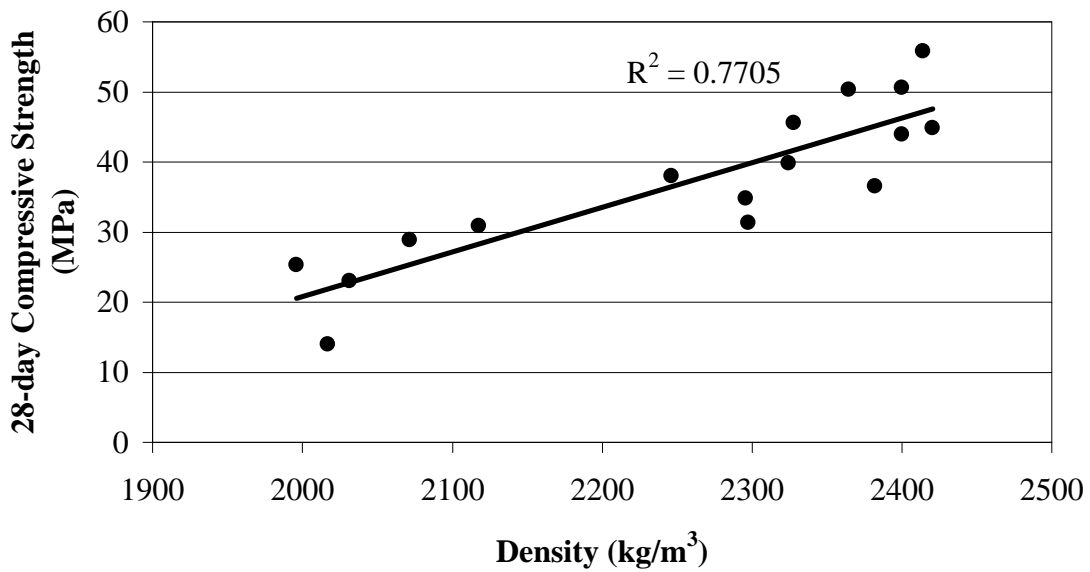


Fig. 4. Relation between density and 28-day compressive strength of concrete

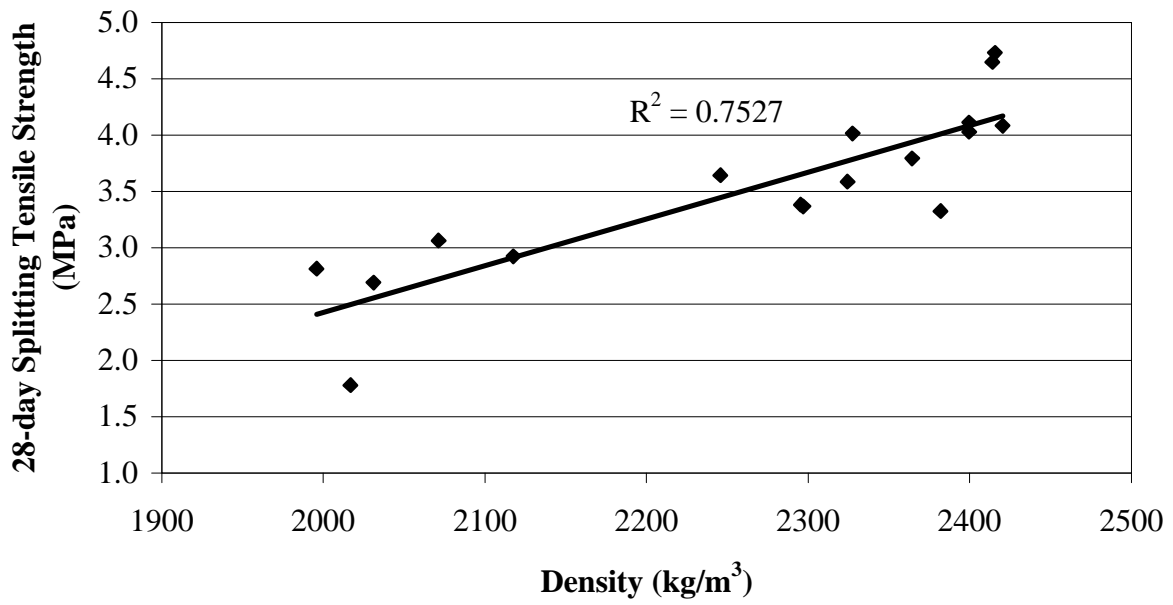


Fig. 5. Relation between density and 28-day splitting tensile strength of concrete