Pergamon

CEMENT AND CONCRETE RESEARCH

Cement and Concrete Research 31 (2001) 1033–1041

Improving the properties of cement–fly ash grout using fiber and superplasticizer

Wei-Hsing Huang*

Department of Civil Engineering, National Central University, Jungli 32054, Taiwan, ROC

Received 14 September 2000; accepted 16 April 2001

Abstract

The physical properties of cement–fly ash grouts containing organic fiber and superplasticizer (SP) for isolation of low-level radioactive wastes are presented. In order to produce a workable, crack-resisting, impermeable, and durable grout, the grout mixes studied contain cement, fly ash, water, polypropylene (PP) fiber, and SP. Laboratory studies included viscosity, bleeding, setting time, compressive and flexural strengths, pore structure, water permeability, and durability of grout mixes. The findings indicated that grouts containing organic fiber were more crack-resisting and less vulnerable to environmental changes, but showed higher viscosity and permeability. With the incorporation of SP in grout mixes, the adverse effects introduced by organic fiber were corrected with additional improvements in viscosity, flexural strength, water permeability, and durability against wet–dry cycling and sulfate attack.

Keywords: Cement grout; Polypropylene fiber; Superplasticizer

1. Introduction

The current proposal for permanent disposal of the low-level radioactive wastes in Taiwan calls for stabilizing or encapsulating of these wastes in steel drums, which in turn are buried in shallow trenches. Some of the engineering barrier materials and isolation strategies are shown schematically in Fig. 1. In this disposal concept, grouts may be used as a solidification agent and as a barrier material.

Previous studies on grouts for containment of hazardous and radioactive wastes have postulated the use of cement–fly ash mixes in such applications [1]. However, cement grouts also have been criticized for being susceptible to cracking. At several disposal sites, cracks have formed in the soil cover or cap due to ground settlements and large quantities of water have entered through soil caps [2]. To mitigate this problem of cracking and deterioration, incorporation of fiber into cement grouts is considered as a potential solution. The addition of fiber to cement paste has been reported to improve the flexural and tensile strengths of cement paste and concrete, but there is limited information available on cement grouts. In this study, polypropylene (PP) fiber was selected because it is less susceptible to chemical attack and is expected to minimize brittleness of the grout barrier thus reducing the potential of cracking in the burial trench [3]. Also, laboratory experiences indicated that the use of fiber tends to reduce the flowability of grouts. Therefore, superplasticizer (SP) was adopted to improve the workability of grout mixes, especially those with low water/solid ratio.

In a waste disposal site, the primary mechanism for the likely introduction of hazardous and/or radioactive elements into the environment is through physical and chemical deterioration of the isolation barrier. The objective of this study was to determine the effect of the PP fiber and SP on the physical properties of cement–fly ash grouts, including viscosity, setting time, bleeding, compressive and flexural strength, water permeability, and pore size distribution. In addition, the durability against sulfate attack and wet–dry cycling of cement–fly ash grouts was evaluated in the laboratory.

* Tel.: +886-3-425-7062; fax: +886-3-425-2960.
E-mail address: t321655@ncu.edu.tw (W.-H. Huang).

0008-8846/01/$ – see front matter
PII: S0008-8846(01)00527-0

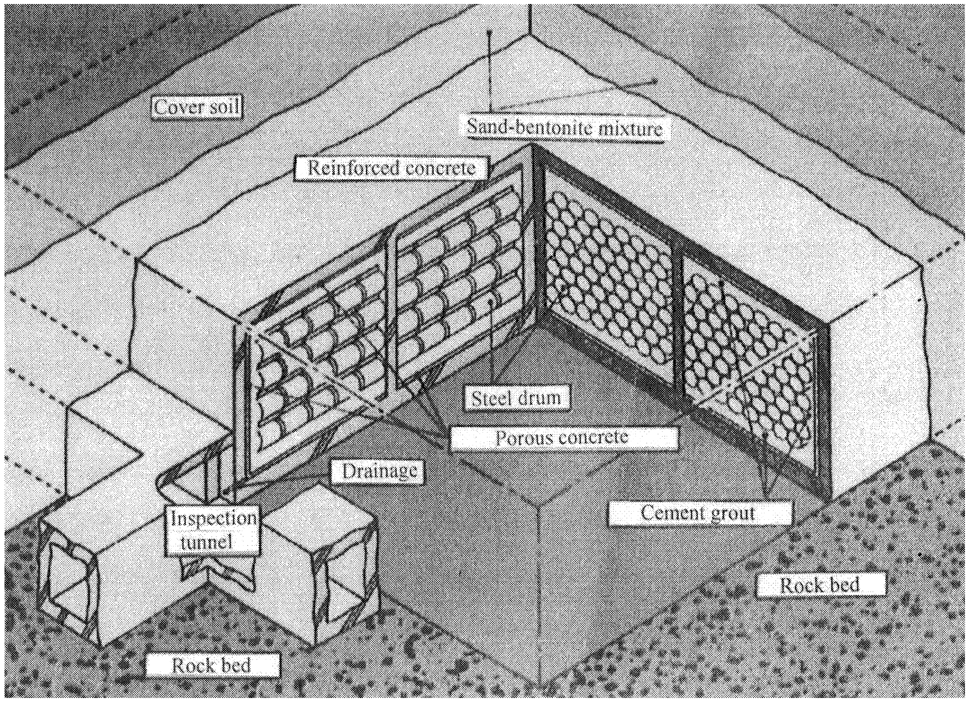

Fig. 1. Schematic diagram of the burial trench disposal concept.

2. Experimental

A standard grout mix prepared using 70% Type I Portland cement and 30% fly ash was developed as the control mix in the laboratory program [1]. The fly ash is a Class F fly ash obtained from Taichung Power Plant in central Taiwan. The PP fiber used is a 3-denier fibrillated fiber having a length of approximately 10 mm. One percent PP fiber (by weight) was added to the cement–fly ash mix. The liquid SP used contained 42% sulfonated naphthalene formaldehyde condensate conforming to ASTM C 494, Type F high-range water-reducing agent. Grout mixtures were prepared with water/solid (water-to-cement + fly ash) ratios ranging from 0.4 to 0.8. An SP dosage of 1% of the solids by mass was used for grout with its water/solid ratio up to 0.6. Table 1 summarizes the formulas used in the study with their notation used in this article.

Table 1
Grout formula and notation

Water/cement + fly ash	PP fiber (%)	SP (%)	Notation[a]
0.4	–	–	C-4
0.5	–	–	C-5
0.6	–	–	C-6
0.7	–	–	C-7
0.8	–	–	C-8
0.4	1	–	C-F4
0.5	1	–	C-F5
0.6	1	–	C-F6
0.7	1	–	C-F7
0.8	1	–	C-F9
0.4	–	1	SP-4
0.5	–	1	SP-5
0.6	–	1	SP-6
0.4	1	1	SP-F4
0.5	1	1	SP-F5
0.6	1	1	SP-F6

[a] C = control group; SP = superplasticizer group; F = fiber added.

Mixing of all grouts were accomplished using a three-blade paddle mixer as suggested in ASTM C 938. The exact amount of water was first poured into the mixer and the mixer started. Premixed cement–fly ash was added to the mixer in 2 min and kept mixing for 3 min. Delayed addition of SP is reported to decrease adsorption on C_3A after mixing, resulting in greater availability of SP to increase dispersion [4]. Therefore, SP was added to the mixes after addition of the cement, at about 4 min from the beginning of mixing. Finally, PP fiber was added at 1 min before the completion of mixing. The entire mixing procedure took about 5 1/2 min.

Hardened grout specimens prepared for strength and permeability tests were cured in a moisture room until the designated age for testing. Durability specimens were cured for 28 days and then subjected to either wet–dry cycling or submersion into magnesium sulfate solution. They were then tested for changes in water permeability and compressive and flexural strengths.

3. Results and discussion

3.1. Viscosity

The apparent viscosity of grout mixes was measured with a coaxial cylinder viscometer. Grouts with low

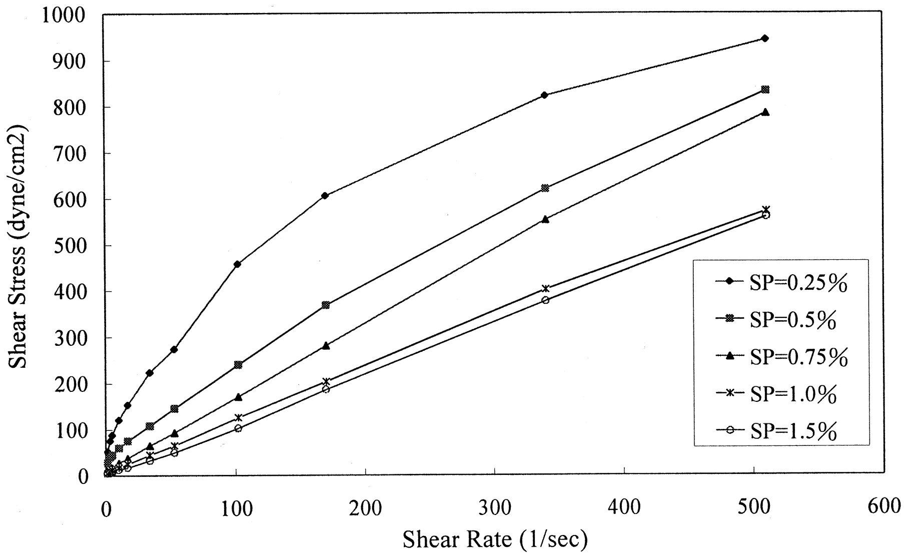

Fig. 2. Effect of SP content on the shear stress–shear rate relationship for cement–fly ash grout (water/solid = 0.4).

viscosity are preferred for penetrating fine fissures or long distances. High-viscosity grouts find applications in injection into wider fractures or limited depths. Fig. 2 shows the shear stress–shear rate relationship of the control grout mix (water/solid = 0.4) with a range of SP contents. The use of SP acts to impart a strong negative charge on cement particles, which considerably reduces the surface tension of the surrounding solution and hence significantly improves the viscosity of grouts. As expected, the apparent viscosity at any shear rate was decreased as the amount of SP increases. A greater effect is observed for SP contents of less than 1%. Also, as the SP content exceeded 0.75%, the yield stress decreased to almost zero and the shear stress–shear rate curves were virtually linear. This indicates that the rheological behavior of these grouts is approaching Newtonian flow over the time period of the measurement. As a result, the SP content was established at 1% throughout the study.

The flow property of grouts was determined using an ASTM C 939 flow cone. In this test, 1.725 l of grout flows from the discharge tube of the cone and the time of efflux is recorded. It should be noted that the ASTM flow cone has a different volume and geometry than a Marsh cone. The time of efflux for water through the ASTM cone is 8 s. Fig. 3 shows that the addition of PP fiber increases the efflux time, especially at high water/solid ratios. SP is found to be effective in improving the flow property of grouts, as manifested by the marked reduction in efflux time for grouts with and without fiber.

3.2. Time of set

The time required for grout to achieve the initial and final set is of great importance in the field. It is commonly recognized that the use of SP may cause significant retardation in setting times. In this study, the

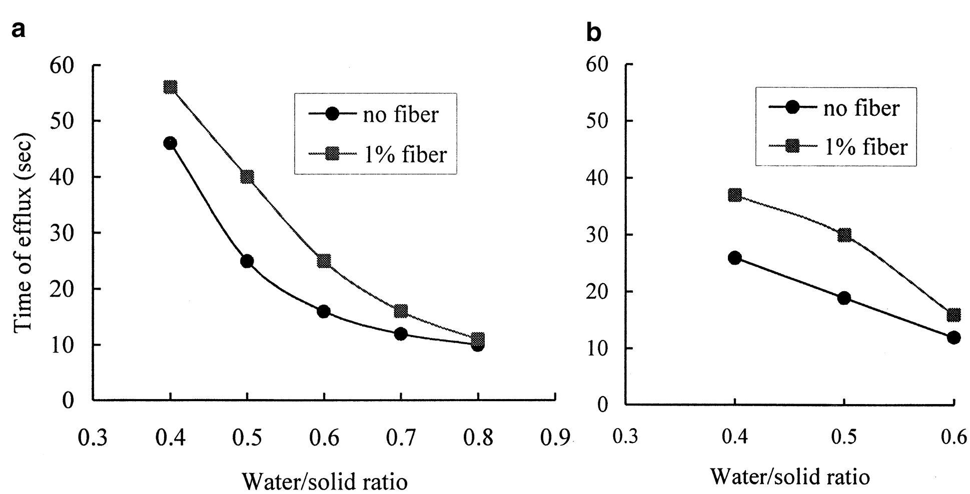

Fig. 3. Flow cone time of grout mixes: (a) no SP, (b) 1% SP.

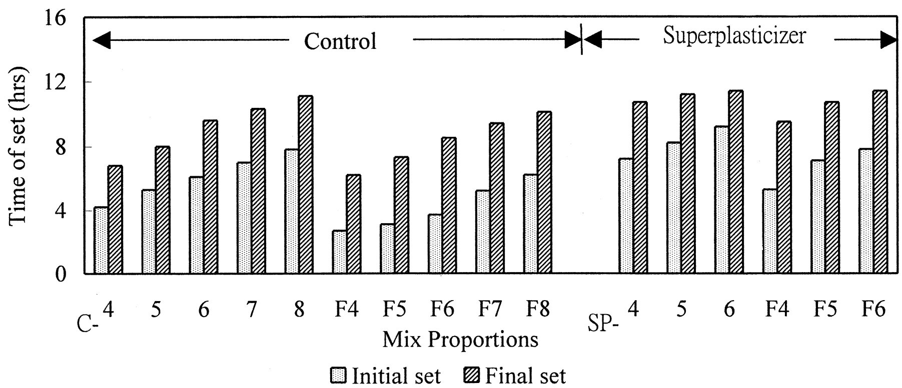

Fig. 4. Setting time of grout mixes.

initial and final setting times of grout mixes were measured by the Vicat apparatus (ASTM C 953) at 23°C. The results are summarized in Fig. 4. It was observed that 1% SP delays the initial and final setting times by 2–4 h, depending on the water/solid ratio. On the other hand, the addition of PP fiber shows an accelerating effect on the setting times of grout mixes, especially the initial set. In general, the initial set occurs at 4–8 h, and the final set is less than 12 h. For most field applications, the setting times for grout mixes studied are within acceptable range.

3.3. Bleed

Freshly mixed grout was evaluated for its final bleed in a glass graduated cylinder following the procedure described in ASTM C 940. Fig. 5 shows the final bleed as percentages of the initial volume of the grout. The water/solid ratio has a great effect on bleeding of the grout. The data indicate that, for water/solid ratio up to 0.5, the addition of PP fiber to the grout slightly increases the final bleed. However, this increasing effect becomes quite noticeable when the water/solid ratio is greater than 0.6. This can be explained by the easily accessible bleeding paths created by the slim fibers in the unset grout. At water/solid ratio higher than 0.6, the amount of water becomes so excessive that segregation of the light-weighted PP fiber from the solid can be observed. The use of SP is expected to increase the final bleed. This is due to the retarding effect of SP, which delays the cement reactions and allows time for increased bleeding.

Based on laboratory results on final bleed, it seems that the use of standard 70–30% cement–fly ash grout containing PP fiber should be limited to low water/solid ratios to insure effectiveness in the grouted fracture. If high water/solid ratios (> 0.6) are desired for specific applications, final bleed of the grout must be minimized by reducing the amount of fly ash and/or incorporating additives effective in eliminating bleed.

3.4. Compressive and flexural strengths

The compressive strength of the grout provides a good indicator of the quality of the hardened grout. The flexural strength is an important property because underground

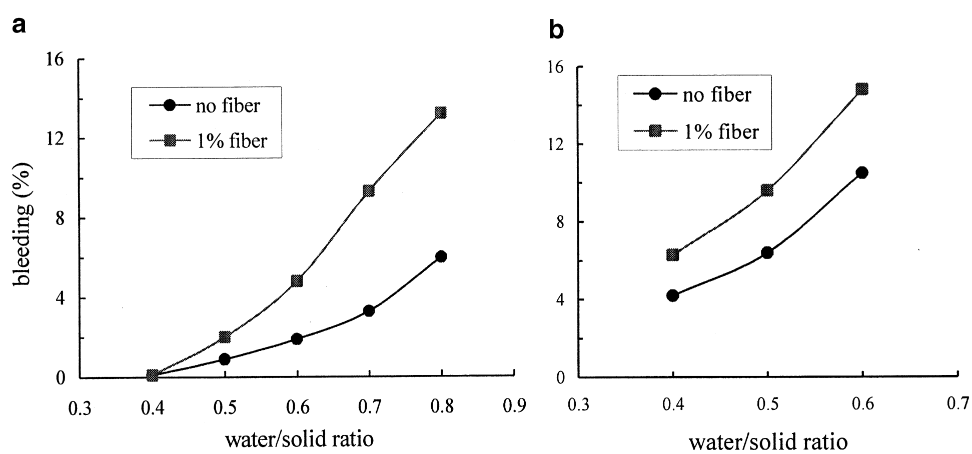

Fig. 5. Final bleed of grout mixes: (a) no SP, (b) 1% SP.

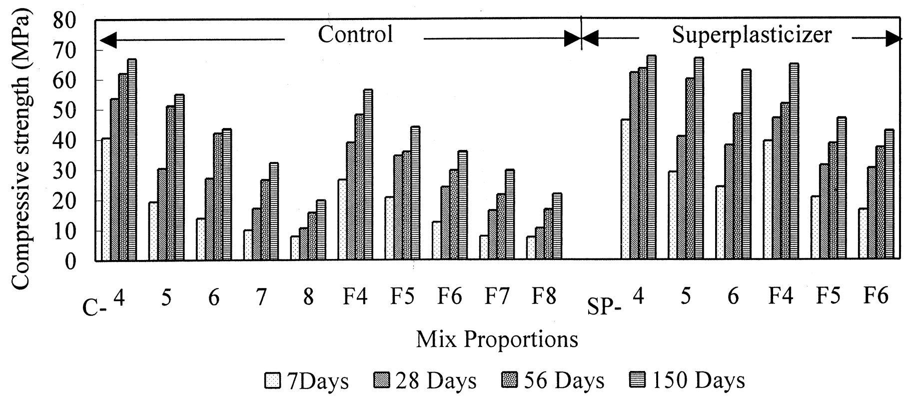

Fig. 6. Compressive strength development of grout mixes.

containment barriers are frequently subjected to high lateral earth pressure. In this study, the compressive and flexural strength tests were conducted on 50-mm cubes and $160 \times 50 \times 50$ mm prisms, respectively, at 7, 28, 56, and 150 days. Test results on compressive strength are shown in Fig. 6. In the control group, slight reductions in compressive strength can be observed with the use of PP fiber. This can be explained by the redistribution of void structure due to the inclusion of fiber, and the presence of weak interfacial bonds between the fiber and cement–fly ash grains [5]. On the other hand, the incorporation of SP increases the compressive strength of the grouts, especially for mixes with high water/solid ratio. This indicates that a better homogeneity of the cement grout structure is achieved in the well-dispersed system produced with the addition of SP.

Fig. 7 presents the flexural strength of grout mixes determined using a third-point loading method. The data show that the reduction in flexural strength resulting from fiber addition is similar to that in compressive strength. However, with the incorporation of SP, the fiber-laden mixes show improved flexural strength. And this effect is more pronounced for grout mixes with low water/solid ratio. This implies that the structural defects resulted from the addition of PP fiber to grout mixes can be modified by the use of SP, and the crack-resisting capability of PP fiber can thus be manifested.

3.5. Pore structure

Pore structure is a major component of the microstructure that affects water permeability and durability of cement grouts [6]. In this study, pore size distribution of the hardened grouts was determined using mercury intrusion porosimetry. The instrument has a pressure capacity of 420 MPa (60 ksi), and is capable of penetrating pores as small as 3 nm in diameter. In a cement paste, it is suggested that capillary pores larger than 10 nm influence mostly the strength and permeability; while gel pores smaller than 10 nm influence the drying shrinkage and creep [7]. The capillary pores can further be divided into large (>50 nm) and medium (50–10 nm) capillary pores. Based on this assumption, the pores of the grout were divided into capillary and gel pores. Fig. 8 shows the volume of pores in the three size categories for samples cured for 28 days.

In the control group, no significant difference in the total porosity exists between grouts containing PP fiber

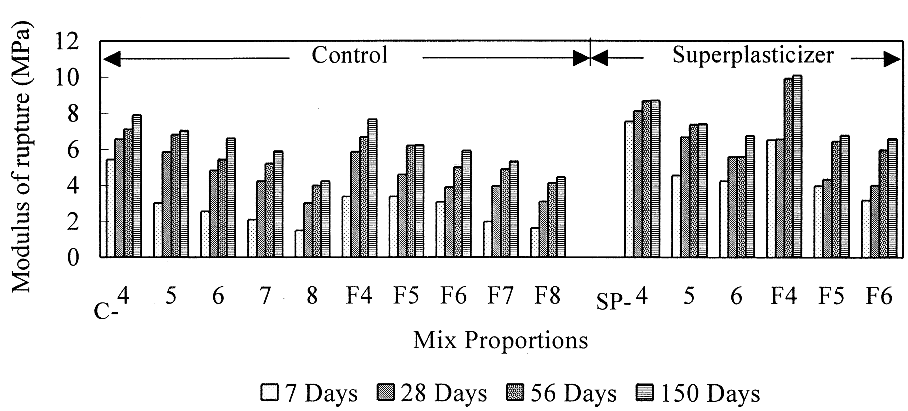

Fig. 7. Flexural strength development of grout mixes.

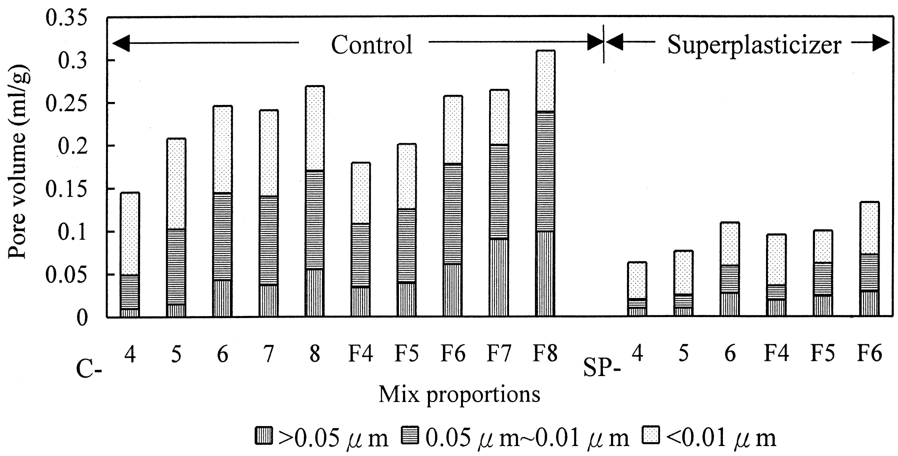

Fig. 8. Pore volume distribution of grouts at 28 days.

and those not. However, the amount of large pores increases with the addition of PP fiber. This is illustrated in Fig. 8 by comparing the proportions of pores in each size category. On the other hand, significant reductions in total porosity and pore size were observed in mixes containing SP. It was also found that the volume of large pores showed greater reduction with the use of SP. These findings are in agreement with other research on cement paste containing SP [8,9]. The decreases in the amount and size of pores result in a refined pore structure, which in turn is beneficial to the engineering and durability properties of the grout.

The improvements in the pore structure of grouts containing SP can further be manifested by microscopic examination on the hardened grout. Fig. 9 shows the photomicrographs taken from the hardened control and superplasticized grouts by a scanning electronic microscope. Pores are clearly illustrated at the interface of PP fiber and the cement–fly ash matrix. With the addition of SP, the pores around the periphery of the fibers are eliminated, hence the fiber–matrix bond is greatly improved.

3.6. Water permeability

The major function of subsurface grout barriers is to prohibit the migration of groundwater flow in or out of the waste. Most deleterious reactions, including sulfate attack, corrosion, alkali–aggregate reactions, and freezing and thawing, involve the ingress of water or aggressive solutions. Impermeable grouts not only provide hydraulic isolation but also enhance durability. Therefore, permeability is the most important property of watertight and durable grout barriers in aggressive underground environments.

Water permeability tests were performed on 28-day grout specimens using an apparatus similar to that adopted by Soongswang et al. [10]. The permeabilities of all grouts

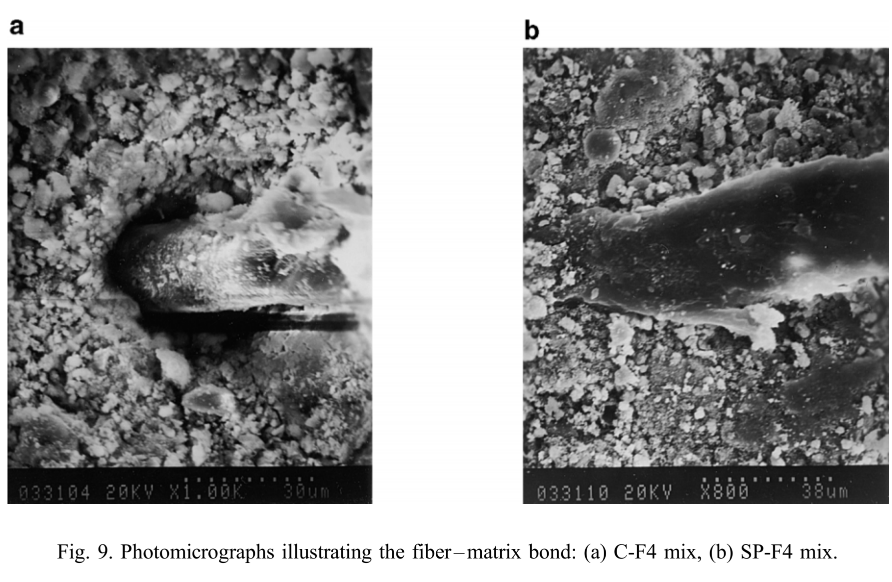

Fig. 9. Photomicrographs illustrating the fiber–matrix bond: (a) C-F4 mix, (b) SP-F4 mix.

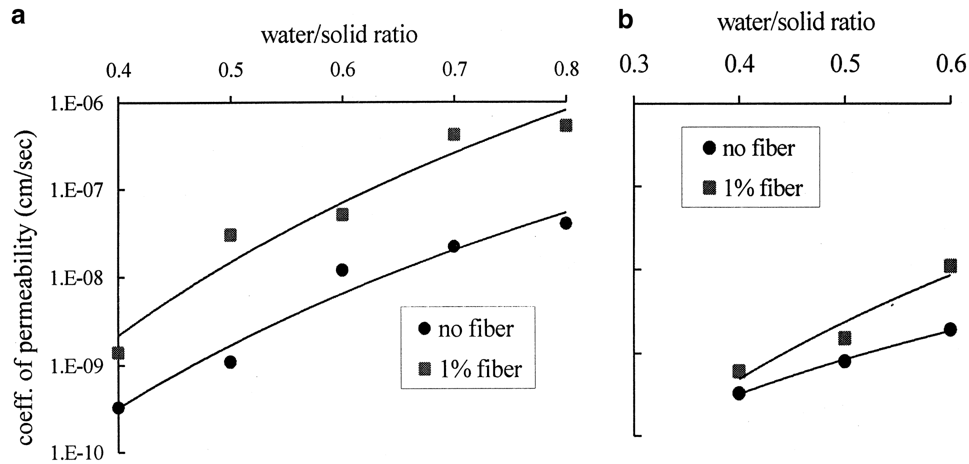

Fig. 10. Water permeability of grouts at 28 days: (a) no SP, (b) 1% SP.

measured on the 102-mm diameter specimens are presented in Fig. 10. The addition of PP fiber shows an increasing effect on the permeability of grouts, especially in the control group. This indicates that the openings at the fiber–matrix interface are providing convenient flow paths for water permeation. However, this can be offset by the addition of an SP. Due to the reduction in porosity and refinement of pores, the water permeabilities of grouts containing SP are approximately two orders of magnitude lower than those of control grouts. In general, grouts containing SP have coefficient of permeability lower than 10^{-8} cm/s.

In an attempt to predict the permeability of grout mixes, Fig. 11 was plotted to establish the relationship between the coefficient of permeability and the amount of capillary pores (>10 nm) in the grout. Fig. 11 shows that the permeability increases exponentially with the volume of large pores and the two parameters are well correlated. Since the measurement of the coefficient of permeability is rather complex and time-consuming, this correlation provides help in predicting the coefficient of permeability of grouts.

Based on the results derived from hardened grout, the addition of PP fiber alone seems to have adverse effects on compressive strength, pore structure, and water permeability of grout mixes. However, if SP was added at the same time, these adverse effects were corrected and the expected advantages of SP on fresh grout were still observed. This can be attributed to the dispersing effect imparted by SP, which provides improved homogeneity in the cement grout system. Therefore, in case PP fiber is introduced for a less brittle subsurface grout barrier, it is recommended that an appropriate amount of SP be incorporated in the grout formula.

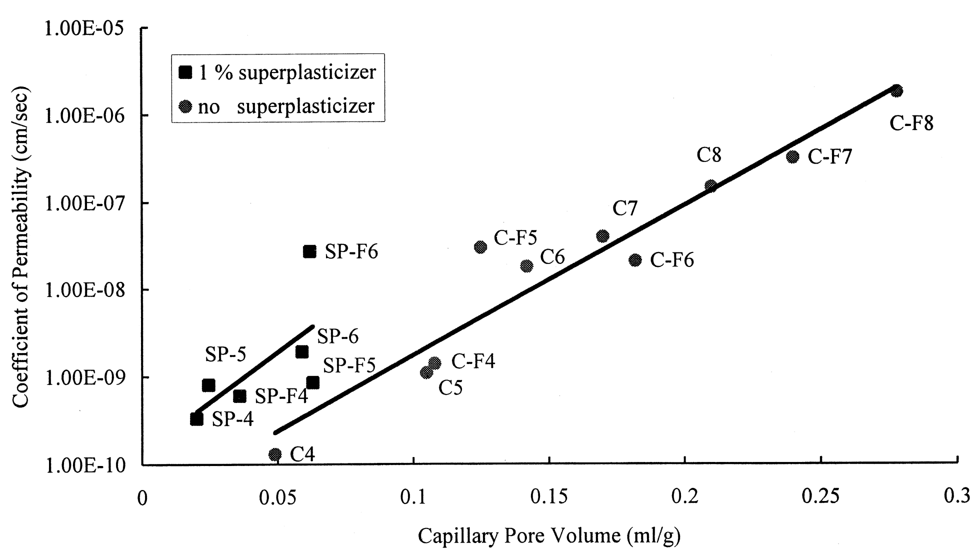

Fig. 11. Relationship between capillary pore volume and coefficient of permeability.

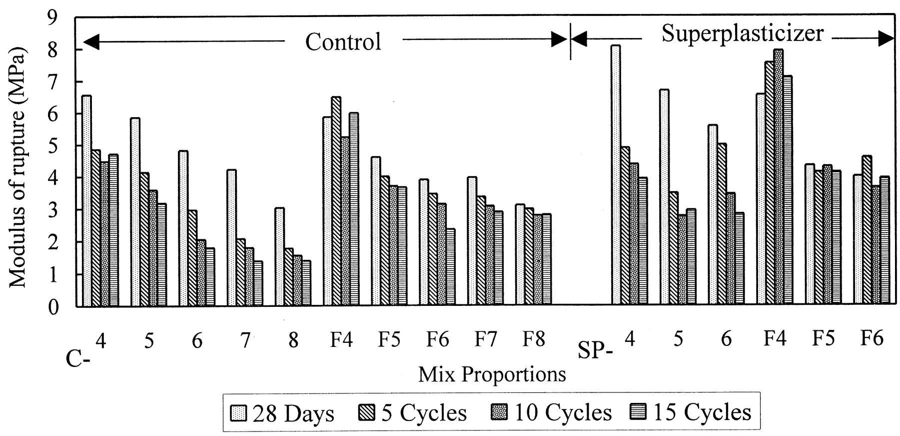

Fig. 12. Flexural strength of grouts experienced various cycles of wetting and drying.

3.7. Resistance to wetting–drying cycles

The proper formulation of grout mixes greatly depends on the expected performance over time in service. Changes in the properties of grout mixes in simulated in-service environment are of major concern. In this article, possible environmental changes that a subsurface burial trench might incur in Taiwan, including wet–dry cycling and sulfate attack, were evaluated.

Following moist curing for 28 days, grout specimens were exposed to wetting–drying cycles. This was accomplished by alternately submerging grout specimens in water for 24 h and drying in an oven maintained at 40°C for 24 h. Grouts were resaturated in water and tested for their water permeability and strength after 5, 10, and 15 cycles of wetting and drying.

The flexural strength of grout specimens obtained after various levels of wet–dry cycling are compared to the 28-day strength in Fig. 12. It is noted that grout mixes with no fiber experienced marked reduction in flexural strength after wet–dry cycling, while grouts containing PP fiber exhibited practically no reduction in flexural strength. This indicates that PP fiber helps in resisting the change in volume of the grout resulting from wet–dry process, because the fibrillated fiber will bridge the microcracks and inhibit their propagation.

Fig. 13 shows the change in coefficient of permeability (Δk) of grouts after exposure to 15 wet–dry cycles, expressed as a percentage of that of the moist-cured specimens. The equation used is Eq. (1)

$$\Delta k = -[(\log k_2 - \log k_1)/\log k_1] \times 100\% \quad (1)$$

where k_1 and k_2 are the coefficient of permeability of grouts before and after wet–dry cycles, respectively. Again, it can be seen that grouts containing PP fiber exhibit no increase in permeability, indicating that PP fiber is effective in resisting the development of cracks caused by drying shrinkage.

3.8. Resistance to sulfate attack

Grouts were submerged in 4.2% magnesium sulfate solution after 28-day moist curing and then tested for strength and water permeability. The measured strengths after 120-day submersion were compared to those obtained from moist cured grouts. In general, the changes in flexural strength after submersion in sulfate solution were found to

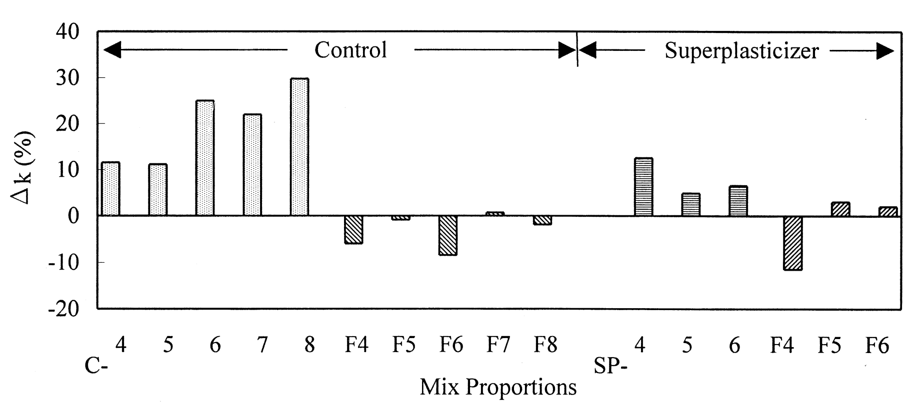

Fig. 13. Change in coefficient of permeability of grouts experienced 15 wet–dry cycles.

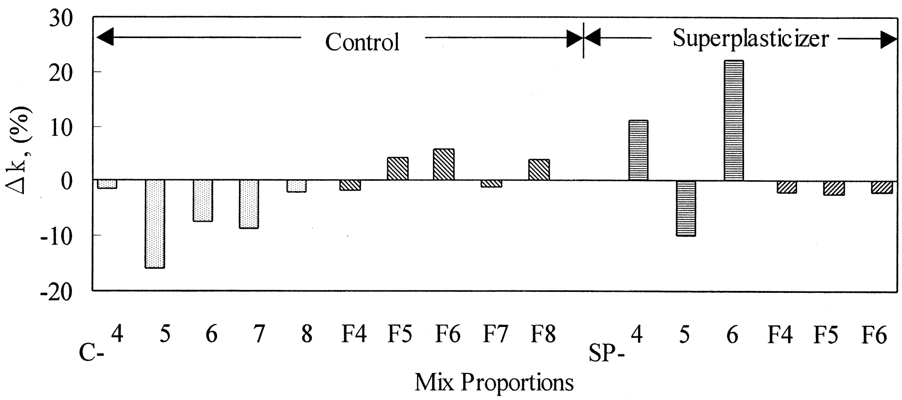

Fig. 14. Change in permeability after exposure to sulfate attack for 120 days.

be negligible. Also, no significant differences exist in the change of strength between the control and SP-containing grouts. Fig. 14 summarizes the change in water permeability of the grout mixes after exposure to sulfate solution. Grouts in the control group show insignificant change in water permeability. On the other hand, grouts containing SP alone show some increases, while the water permeability of mixes containing both PP fiber and SP remained unaffected.

4. Conclusions

The grouts tested in this study have applications in underground situations such as injection to subsurface voids and fracture zones, soil/rock–concrete interfaces, sealing containers, and intercontainer spaces of low-level radioactive wastes.

Substitution of 30% cement by Class F fly ash produces economical grouts with reasonable physical properties. Addition of PP fiber alone may have adverse effects on properties of fresh and hardened grout, but significantly improves the grout's resistance to cracking and changes in volume resulting from wet–dry cycles and sulfate attack. Grouts containing appropriate amount of SP show enhanced flowability and viscosity, higher strengths, and reduced permeability. Also, the adverse effects introduced by fiber addition are generally eliminated by the use of SP. To assure enhanced physical properties and durability of cement–fly ash grout, it is recommended that the use of PP fiber always be accompanied by the incorporation of SP.

Acknowledgments

This study was supported by the National Science Council under project number NSC 83-0410-E008-012 and NSC 84-2211-E-008-033.

References

[1] W.H. Huang, Properties of cement–fly ash grouts admixed with bentonite, silica fume, or organic fiber, Cem. Concr. Res. 27 (3) (1997) 395–406.
[2] E.D. Daniel, Shallow land burial of low level radioactive waste, J. Geotech. Eng. 109 (1) (1983) 40–55.
[3] I.O. Okonkwo, Improving the durability of radioactive waste grout barriers using polypropylene fibers, Proc. Waste Manage. 89 (1989) 715–722.
[4] M.L. Allan, L.E. Kukacka, Comparison between slag- and silica fume-modified grouts, ACI Mater J. 93 (6) (1996) 559–568.
[5] I.O. Okonkwo, Durability of particulate grouts: For isolation of low-level radioactive wastes, PhD dissertation, Purdue University, 1988.
[6] D. Perraton, P.C. Aitcin, D. Vezina, Permeabilities of silica fume concrete, D. Whiting, A. Walitt (Eds.), Permeability of Concrete, Am. Concr. Inst. SP-108, 1988, pp. 63–84.
[7] P.K. Mehta, Concrete: Structure, Properties, and Materials, Prentice-Hall, Englewood Cliffs, NJ, 1986.
[8] J.M. Khatib, P.S. Mangat, Influence of superplasticizer and curing on porosity and pore structure of cement paste, Cem. Concr. Compos. 21 (1999) 431–437.
[9] N.B. Singh, R. Sarvahi, Effect of superplasticizers on the hydration of cement, Cem. Concr. Res. 22 (2) (1992) 725–731.
[10] P. Soongswang, M. Tia, C. Bloomquist, L.M. Sessions, An efficient test set-up for determining the water-permeability of concrete, Transp. Res. Rec. 1204 (1988) 77–82.