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Study on the durability of the low diffusion layer for sub-surface radioactive waste disposal facilities in JAPAN

K. NIWASE [1*], S. KUDO [1], Y. EDAMATSU [2], N. SUGIHASHI [3] and H. YANOKURA [3]

[1] *Japan Nuclear Fuel Limited, Japan,* [2] *Newjec Inc, Japan,* [3] *Shimizu Corporation, Japan*
[*] *504-22, Aza Nozuki, Oaza Obuchi, Rokkasyo-mura, Kamikita-gun, Aomori-ken , 039-3212, Japan*
kazuhito.niwase@jnfl.co.jp, sunao.kudou@jnfl.co.jp, edamatsuys@newjec.co.jp, n-sugihashi@shimz.co.jp, h-yanokura@shimz.co.jp

ABSTRACT

Among low-level radioactive waste (LLRW) disposal facilities, an engineered barrier system for a sub-surface disposal facility is being planned in Japan. This facility will be required the assessment of multi engineered barrier in several tens of thousands years. Multi engineered barrier materials are also used with the two kinds of cementitious materials and a bentonite. The low diffusion layer made of cementitious materials of them, shall delay the migration due to the diffusion of a radionuclide. Therefore the durability and low diffusion, cracks control are the important matters in the low diffusion layer. This paper describes the characteristics of the low diffusion layer for the sub-surface disposal facility in laboratory comparative testing and the field demonstration tests from the view points of the durability. As a result, it was demonstrated that the low diffusion layer with selected materials and mix proportion could exhibit the required initial performance and the durability.

Keywords. diffusion coefficient, crack, diameter distribution of fine pores, sub-surface disposal facility, radioactive waste disposal facilities

INTORODUCTION

In Japan we are planning to construct sub-surface disposal facilities for relatively high low-level radioactive waste. To ensure the safety of the facilities, we should not bequeath a huge burden to future generations, thus the facilities must be maintenance-free. The safety assessment is related to the great variety of natural phenomenon or artificial events, etc. from the physical, chemical and geochemical aspects over a long period of time. Therefore, it is complicated to set the conditions for the assessment and the uncertainties derived from the long period are unavoidable. It is important to design the barriers that avoid a serious functional loss by utilizing the individual advantage of multiple barriers (roughly classified into the engineered barrier and the natural barrier which is surrounding bedrock) and compensating their weakness. It is also important to indicate a margin of safety ratio for an entire system in order to evaluate the barriers. It is necessary to verify the performance of the engineered and natural barriers for durability of ultra-long-term which is over tens of thousands of years, as radioactive waste for the burial disposal contains radionuclides (or radioactive atoms) that require over a long period to be confined and controlled to migrate (or to migrate from one medium to another). The engineered barrier mainly consists of a low water permeability layer and a low diffusion layer. The former is made up of bentonite material for controlling the migration of radionuclide based on advection, and the latter made up of cementitious material for controlling the migration based on diffusion. This design concept of the planned sub-surface disposal facility (Kyoya 2005) is shown in Figure 1. This paper describes the characteristics of the low diffusion layer for the sub-surface disposal facility from the view points of the durability.

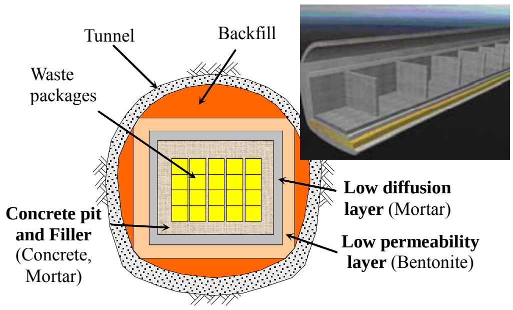


Figure 1. Sub-surface disposal facility

DESIGN PERFORMANCE OF THE LOW DIFFUSION LAYER

The selection of material and mix proportion design is carried out for the purpose of advancing the performance requirements as follows. The performance requirement for the strength is the increase of compressive strength. It for the physical and chemical is the improvement of low permeability, suitability of setting time, no alkali aggregate reaction, and decrease of big voids and making densification. It for the crack control is the minimization of heat of hydration and drying shrinkage, autogenous shrinkage. It for the construction performance is high flowability and segregation resistance. An indicator of the diffusion coefficient is used to quantitatively evaluate the design requirement of "reduced leakage speed by controlling the diffusion of radio nuclides" for a low diffusion layer. The diffusion coefficient could be evaluated by classifying the original zone and leaching zone, crack zone as shown in Figure.2. In this requirement, the diffusion coefficient of tritium in the original zone is less than 1E-12 m^2/s. And that the crack area is less than 0.05% of surface area in a member of the low diffusion layer.

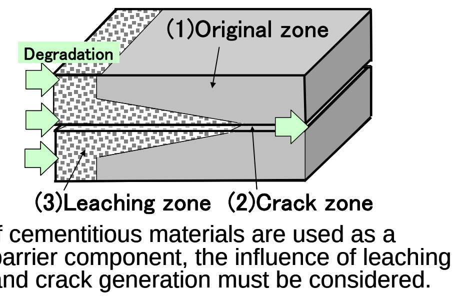


Figure 2. Concept of diffusion coefficient

LABORATORY TEST

Comparative Tests of Materials and Mix Proportions in the Laboratory. For an efficient comparative study on the characteristics of materials determined upon searching related literature, combinations of performance items were organized to be mainly spotlighted regarding the materials and mix proportions. The performance items were compared and considered for a total of 33 as shown in the table 1.

Table 1. The mix proportions

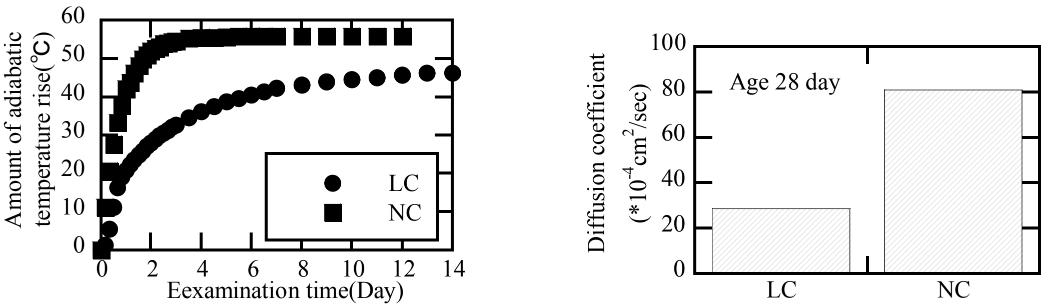
Parameter	Mix proportion №		W/C	s/a	Unit quantity(kg/m^3)													
					W	C	F	K	LP	E	S1	S2	S1'	G1	G2	Ad1	Ad2	AE
Cement	1	LC	45.0%	45.0%	150	333	–	–	–	–	338	534	–	1042	–	1.70 P×0.5%	–	0.009
		NC	45.0%	45.0%	150	333	–	–	–	–	337	533	–	1040	–	2.70 P×0.8%	–	0.012
Aggregate	2	S1+G1	45.0%	45.0%	150	333	–	–	96	–	759	–	–	1040	–	2.00 P×0.6%	–	0.010
		S1+G2	45.0%	45.0%	150	333	–	–	96	–	753	–	–	–	1101	2.70 P×0.8%	–	0.015
		S2+G1	45.0%	45.0%	150	333	–	–	–	–	–	890	–	1042	–	6.70 P×2.0%	–	0.007
		S2+G2	45.0%	45.0%	150	333	–	–	–	–	–	890	–	–	1104	6.70 P×2.0%	–	0.013
		S1,2+G2	45.0%	45.0%	150	333	–	–	–	–	338	534	–	–	1104	2.30 P×0.7%	–	0.009
		S1'+G1	45.0%	45.0%	150	333	–	–	–	–	–	–	840	1042	–	2.30 P×0.7%	–	0.012
Admixture mineral	3	A20	45.0%	44.4%	150	266	67	–	–	–	331	522	–	1042	–	1.70 P×0.5%	–	0.017
		A30	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	1.70 P×0.5%	–	0.023
		B20	45.0%	44.5%	150	266	67	–	–	–	332	524	–	1042	–	1.70 P×0.5%	–	0.017
		B30	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	1.70 P×0.5%	–	0.018
		K50	45.0%	44.5%	150	166	–	167	–	–	332	524	–	1042	–	1.20 P×0.35%	–	0.008
		K70	45.0%	44.4%	150	100	–	233	–	–	330	521	–	1042	–	1.20 P×0.35%	–	0.008
Expansive admixture	4	E20	45.0%	44.2%	150	213	100	–	–	20	328	518	–	1042	–	1.70 P×0.5%	–	0.015
		E30	45.0%	44.2%	150	203	100	–	–	30	328	518	–	1042	–	1.70 P×0.5%	–	0.015
Limestone fine powder	5	LP0	45.0%	42.7%	160	249	107	–	–	–	308	487	–	1042	–	1.80 P×0.5%	–	0.028
		LP20	45.0%	54.0%	160	249	107	–	71	–	362	571	–	837	–	3.60 P×1.0%	–	0.025
		LP40	45.0%	54.0%	160	249	107	–	142	–	334	527	–	837	–	3.90 P×1.1%	–	0.025
Chemical admixture	6	Ad2	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	–	3.30 P×1.0%	0.167
Water-cement ratio	7	WC40	40.0%	43.0%	150	262	113	–	–	–	312	492	–	1042	–	1.30 P×0.35%	–	0.023
		WC55	55.0%	45.8%	150	191	82	–	–	–	349	551	–	1042	–	1.60 P×0.6%	–	0.020
Air content	8	A2.5	45.0%	45.8%	150	233	100	–	–	–	349	551	–	1042	–	1.70 P×0.5%	–	0.007
		A6.5	45.0%	42.5%	150	233	100	–	–	–	306	483	–	1042	–	1.30 P×0.4%	–	0.032
Mixmum size of coares aggregate	9	Gmax13	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	2.30 P×0.7%	–	0.030
		Gmax05	45.0%	100.0%	244	379	163	–	–	–	564	890	–	–	–	0.50 P×0.1%	–	0.043
		LP40Gmax05	45.0%	100.0%	230	358	153	–	204	–	510	805	–	–	–	3.80 P×0.75%	–	0.015
Amount of coares aggregate	10	Gvol800	45.0%	54.9%	150	233	100	–	–	–	407	643	–	842	–	3.30 P×1.0%	–	0.030
		Gvol600	45.0%	65.6%	150	233	100	–	–	–	486	768	–	642	–	6.70 P×2.0%	–	0.027
Flowability	11	SL12	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	0.70 P×2.0%	–	0.020
		SL21	45.0%	44.2%	150	233	100	–	–	–	328	517	–	1042	–	2.00 P×0.6%	–	0.023
		SF50	45.0%	54.0%	160	249	107	–	142	–	334	527	–	837	–	3.20 P×0.9%	–	0.030
		SF70	45.0%	54.0%	160	249	107	–	142	–	334	527	–	837	–	4.50 P×1.25%	–	0.350

C: Cement, F: Fly-ash, K: Blast-furnace slag, LP: Limestone fine powder, E: Expansive admixture, S1: Lime crushed sand(MATSUDATE), S2: Sand(NIWAKAMAI), S1': Lime crushed sand(RISHIRI), G1: Lime crushed stone(MATSUDATE), G2: Crushed stone(KOMAMINE), Ad1: High-range water reducing and air-entraining agent(Polycarboxylate), Ad2: High-range water reducing and air-entraining agent(Naphthalene series), AE: Air-entraining agent

The test method used for comparison and consideration was a representative one for evaluating the performance requirements in accordance with JIS(Japanese Industrial Standard) and JCI(Japan Concrete Insititute), JSCE(Japan Society of Civil Engineers) ,so on.

The followings present part of the test results that became important factors in selecting a material and mix proportion.

Cement Types. As shown in Figures 3, the priority ratings of the LC mix (using in the low-heat portland cement) are high for adiabatic temperature rise, and diffusion coefficient.

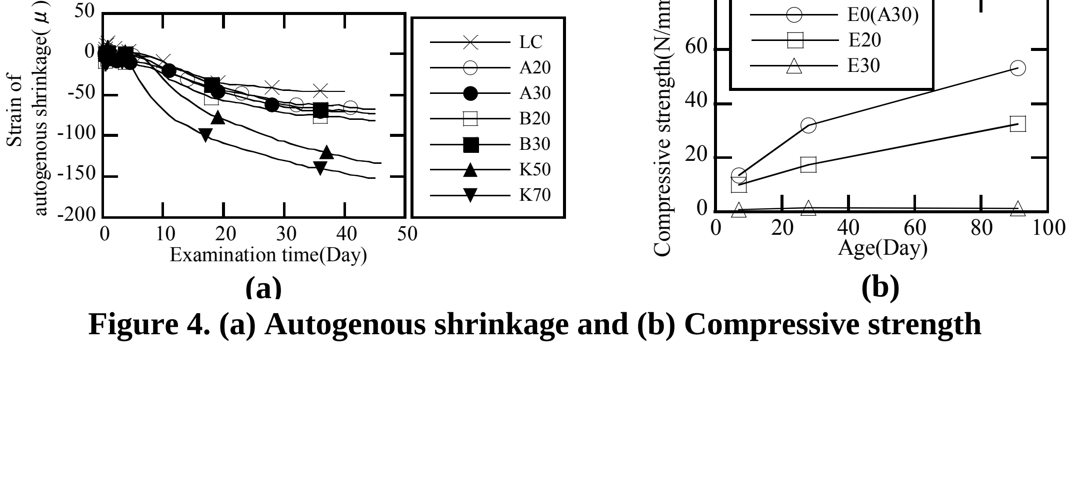


(a) **(b)**

Figure 3. (a)Adiabatic temperature rise and (b)Diffusion coefficient.

Aggregate Types. The aggregate type of S1' + G1 are highly effective in terms of drying shrinkage.

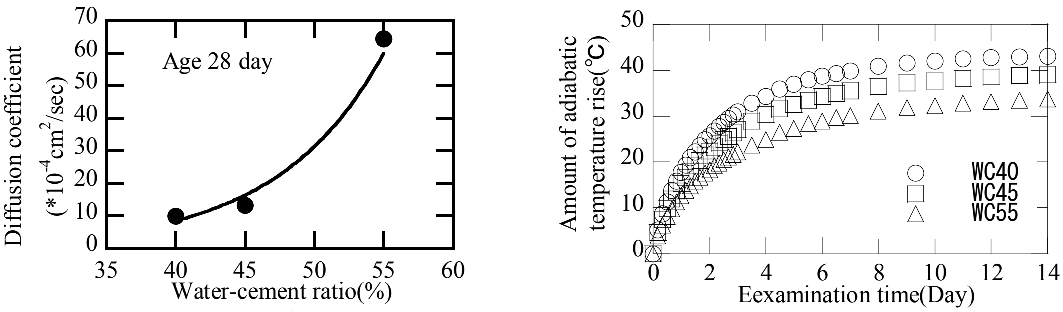
Addition Types and Quantities. The use of fly-ash and blast-furnace slag can reduce the adiabatic temperature rise and the diffusion coefficient of water. The use of blast-furnace slag as shown in Figures 4(a) tends to increase autogenous shrinkage. Hence, the use of fly-ash (A30) as an addition is therefore effective. The use of limestone fine powder also increases segregation resistance and gives high-fluidity concrete high filling performance. Adding an expansive admixture, on the other hand, considerably reduces compressive strength as shown in Figure 4(b). Particularly notable was when the dose was 30 kg/m^3. At that level of dose, there were cases where the model cracked and expanded, resulting in destruction. It was therefore necessary to consider appropriate doses and types, among other considerations, for expansive admixtures.



(a) **(b)**

Figure 4. (a) Autogenous shrinkage and (b) Compressive strength

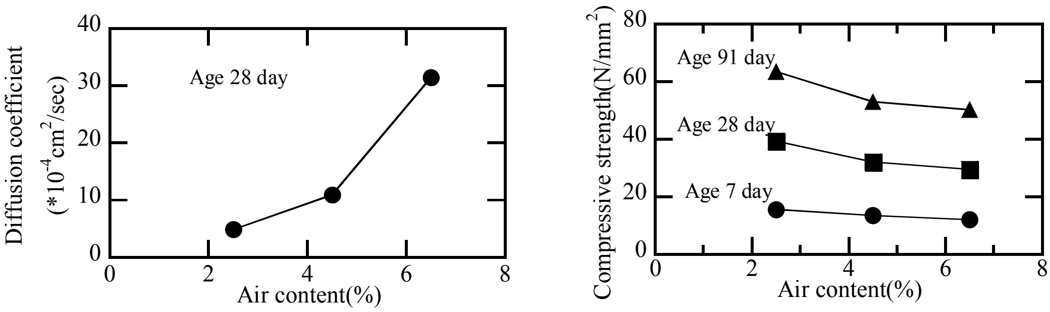
Water-Cement Ratio. As shown in Figure 5(a), the smaller is the W/C, the smaller is the diffusion coefficient. However, such a big difference does not occur when the W/C is less than 45%. Conversely, the adiabatic temperature rise increases as the W/C declines as indicated in Figure 5(b). Based on these findings, the most appropriate W/C is to be at about 45%.



(a) (b)

Figure 5. (a) Diffusion coefficient and (b) Adiabatic temperature rise

Air Content. As shown in Figures 6, as the air content declines, the diffusion coefficient goes down and compressive strength increases. At this facility where freezing and thawing action does not constitute an issue, the air content is better when as low as possible.



(a) (b)

Figure 6. (a) Diffusion coefficient and (b) Compressive strength

Summary of Laboratory Comparative Testing Results. For selecting mix proportions satisfying the materials and mix proportion parameters selected in laboratory comparative testing, a mortar is selected for the low diffusion layer, so that the mortar eliminates the transition zone that is said to form around the coarse aggregate and turn it into capable of reducing the amount of void 100 nm or more in pore diameter (Niwase 2010) to improve the low permeability. Moreover, since the homogeneity, filling performance and other characteristics of the slump-type mix proportion vary with construction workmanship, it was also decided to target a slump flow-type mix proportion that can minimize the effects of construction quality. Of all the mix proportions used in laboratory comparative testing, the mix proportion No.LP40Gmax05 shown in the table 1 most satisfies these parameters, along with the material and mix proportion parameters selected. Based on the findings, the

LP40Gmax05 mix proportion was provisionally set to the mix proportion by: (1) changing from blending sand made by lime crushed sand and land sand (S1 + S2) to sand made by lime crushed sand (S1'), and (2) changing the air content from 4.5% to 2.5%.

The laboratory comparative testing revealed that, the larger the amount of limestone fine powder, the more effective it is in enhancing fresh mortar performance. However, no testing has been conducted at a dose of more than 40%. Regarding the dose of limestone fine powder, breeding, breeding under pressure and efflux time through funnels were considered. The breeding and breeding under pressure were considered indicators of segregation resistance, while the efflux time through funnels was considered an indicator of fluidity. As these test results, breeding is constant at a limestone fine powder amount of 60% or more, but the efflux time through funnels is considered to grow at a dose of 80%, along with increased viscosity. The dose of limestone fine powder was therefore considered appropriate at about 60%.

It was also necessary to further consider expansive admixture and limestone fine powder, which had been insufficiently considered in comparative testing, and then make a final decision on the mix proportion. In laboratory comparative tests, as using the expansive admixture replacing cement with it’s 20 or 30 kg/m^3, this resulted in an excessive rise in expansion amount and a decline in compressive strength. It was then failed to obtain the crack control effect of a typical expansive admixture. The use of an expansive admixture can be expected to improve the effect on the crack control. For that reason, the expansive admixture was enhanced to ensure that: (1) the length change rate in a restrained expansion test (JIS A 6202) would be in the range of 0.015 to 0.025(%), and (2) the compressive strength would be comparable to that achieved when not adding the material aged for seven days. Figure 7(a) shows the test results of the length change rate achieved when changing the dose to 0, 15 and 20 kg/m^3 replacing cement with in the provisional mix proportion. This revealed that the rate of 20 kg/m^3 satisfied the target length change rate (0.015 to 0.025(%)) at an age of seven days. Figure 7(b) shows the results of compressive strength test at seven days. The compressive strength was comparable despite changes in the amount of expansive admixture. These findings were considered appropriate to replace 20 kg/m^3 of expansive admixture with cement.

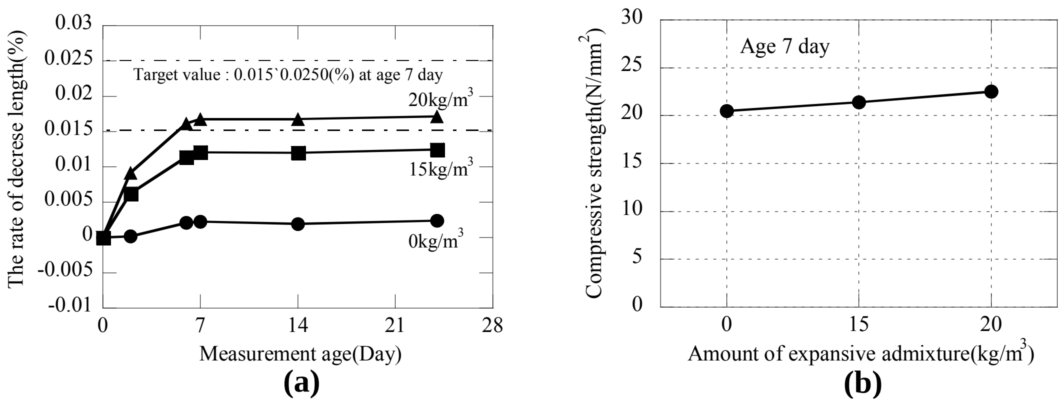


Figure 7. (a) Length rate of change and (b) Compressive strength

Based on the findings above, the material and mix proportion for the low diffusion layer selected in laboratory tests were as listed in Table 2 and Table 3.

Table 2. Selected materials

Material	Sign	Specification
Cement	LPC	Low-heat Portland cement Density=3.22 g/cm^3, Specific surface=3,470cm^2/g
Fine aggregate	S1	Lime crushed sand : Density=2.68g/cm3, FM=2.68
Limestone fine powder	LS	Limestone fine powder Density=2.71 g/cm^3, Specific surface=4,970cm^2/g
Fly-ash	FA	Density=2.25 g/cm^3, Specific surface=3,730cm^2/g
Expansive admixture	LEX	Improved expansive admixture ：Density=3.15g/cm^3
Chemical admixture	SP	Superplasticizer: Polycarboxylate type
	AS	Air-entraining agent: Polyalkylenglycol derivertives

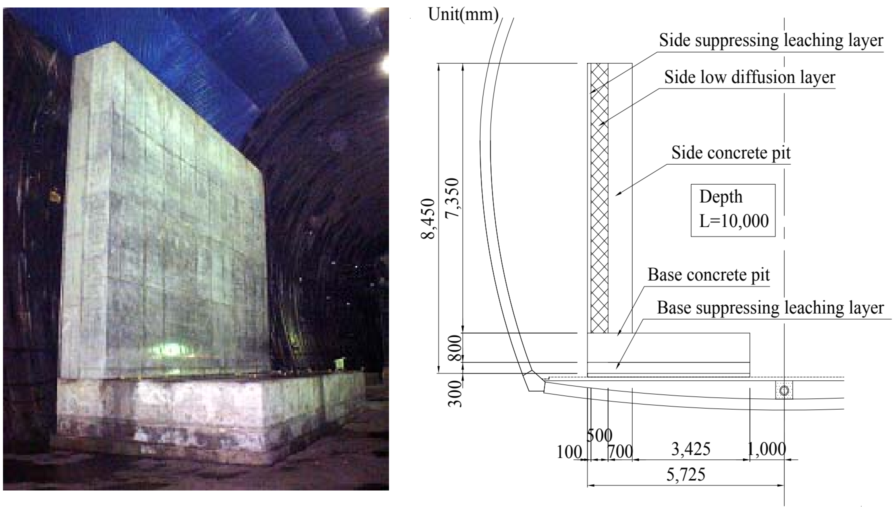
Table 3. Selected mix proportion

Mix proportion No.	W/B (%)	W/P (%)	Slump flow (cm)	Air (%)	Unit quantity (kg/m^3)						
					W	LPC	FA	LEX	LS	S1	SP (P×%)
DLP60	45.0	28.1	65±5	2.5	230	338	153	20	307	1223	0.69

Where, B=LPC+FA+LEX、P=LPC+FA++LEX+LS

FIELD DEMONSTRATION TESTING

Field demonstration testing was conducted by using full-size models as shown in the figure 8, in order to confirm the actual construction performance on site and the crack control effect of the low diffusion layer mix proportion selected in the laboratory testing. This field demonstration testing was conducted inside a test cave with about 18-m wide and 16-m high excavated at about 100-m under the ground surface of a candidate site for constructing the facility in order to simulate the construction environment as well as possible.



(a) **(b)**

Figure 8 Full-size model (a) Overview, (b) Cross section

To evaluate the effects of construction procedure, a fresh mortal performance test was conducted at the locations of mixing plant, unloading a track agitator and placing when the full-size model was built, and a study was conducted on the effects of construction method on the fresh characteristics of the low diffusion layer. Moreover, after hardening, core samples were made from such different locations as the higher and lower parts of the hardened that model to evaluate whether constructing a homogeneous building frame would be possible. It is possible to evaluate the selected material and mix proportion of the low diffusion layer considering the other factors comprehensively. Core samples were also used to confirm and evaluate changes in the diffusion coefficient with aging of the material. The crack control effects were also evaluated by observing the state of crack occurrence, thereby considering these effects on the diffusion coefficient. Core samples were also used to confirm and evaluate changes in the diffusion coefficient with aging of the material. It is possible to evaluate the selected material and mix proportion of the low diffusion layer considering the other factors comprehensively.

Evaluation of the Effects on Construction Method. Quality control testing was conducted at three locations such as at mixing, unloading and after pumping. The quality control test items were mortar temperature, slump flow, efflux time through V-funnel for mortar and compressive strength. These test results shows that the low diffusion layer made in this demonstration test had the required fresh performance and other required the qualities. The hardened characteristics were tested up to the age of one year. A comparative study was made of: (1) samples made at construction and cured in water in a test cave, (2) samples prepared by drilled cores out of the higher and lower parts of the full-size model as shown in the Figure 9, and (3) data about the laboratory test results reported (Niwase 2010).

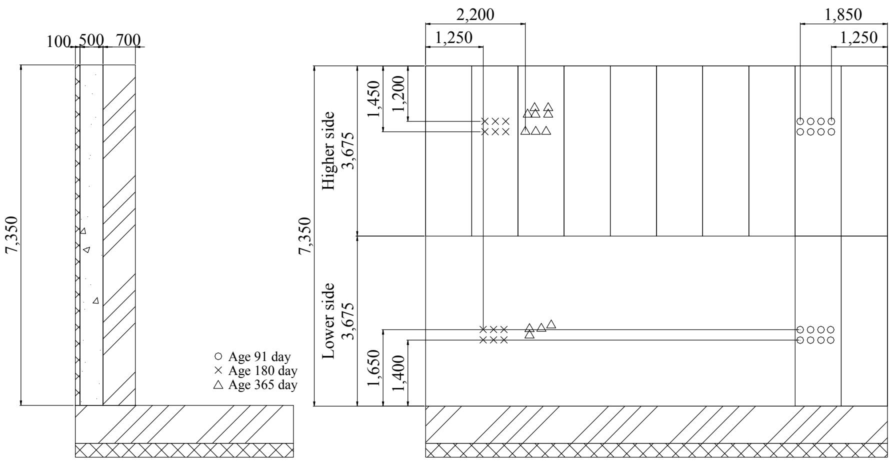
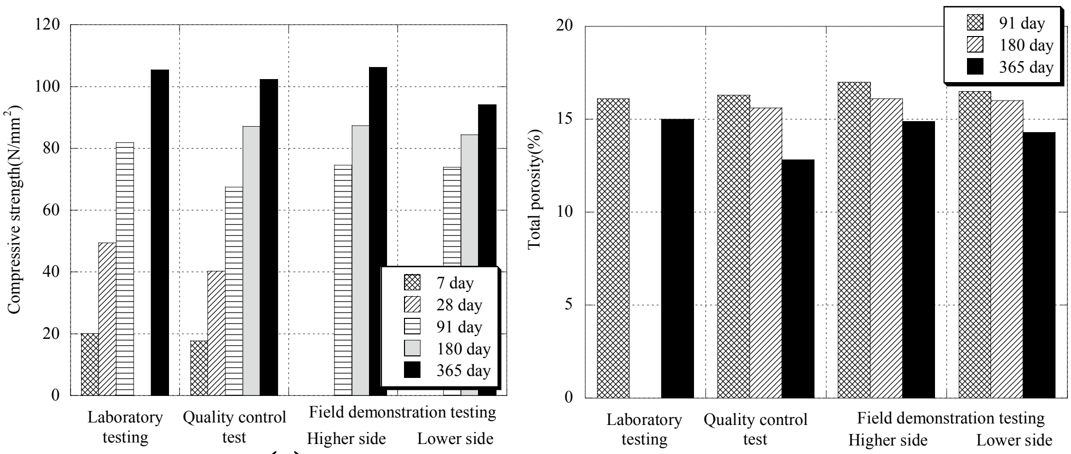


Figure 9 Core sampling positions

Figure 10(a) shows the compressive strength in laboratory testing is higher than in other test phases for two identically aged. However, compressive strength can be well approximated using a logarithmic function of the integrated temperature, so that differences in compressive strength depending on the test phase are due to differences in integrated temperature in other word curing temperature. Figure 10(b) similarly shows the total porosity. The downward trend in total porosity according to increase the age is observed in any of test phases. The results above allowed us to confirm the occurrence of compressive strength and the densification due to the progress of hydration and pozzolanic reaction with aging of the material. It was observed that the progress in such strength and densification can be evaluated not by the location in the member or where the specimen was collected, but mainly by using the integrated temperature. Prior literature clearly indicates the correlation between compressive strength and the porosity with regard to low diffusion and many other properties. The findings of the present experiments are allowed to demonstrate that a full-size model of a homogeneous low diffusion layer could be built equally on the member by using selected materials and mix proportions, methods of construction in the low diffusion layer.



(a) **(b)**

Figure 10 (a)Compressive strength and (b)total porosity

Evaluation of the Diffusion Coefficient of the Original Zone. All test results in the effective diffusion coefficient of tritium were the order of 1E-13 m^2/s. Figure 11 shows the relation between the porosity and effective diffusion coefficient for the low diffusion layer. Comparing concrete of a general mix proportion with the same porosity also shows a very small diffusion coefficient for the low diffusion layer. These findings supported that the direction of mix proportion design was correct. The findings above demonstrated that until the equivalent age about one year, the diffusion coefficient of the original zone achieves a sufficiently small required value.

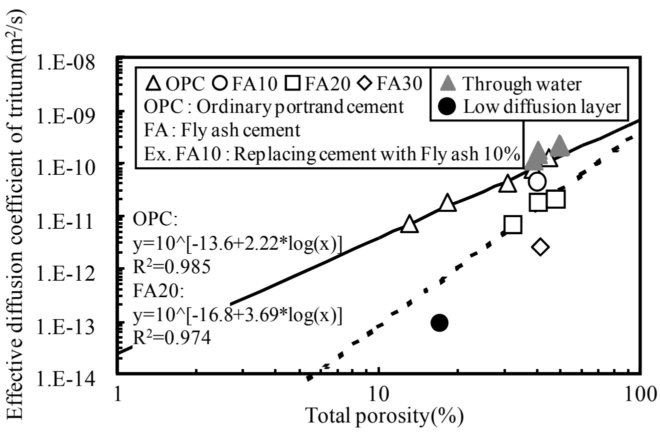


Figure 11 Total porosity and effective diffusion coefficient of tritium

Evaluation of the Crack Control Effect. Figure 12 shows the crack generating situation. The outermost surface generated three cracks with a maximum crack width of 0.06 mm. At the material aging of 42 days, the first crack generated in the middle in the lengthwise direction. Then at material aging of 59 days, the second and third cracks formed in the span location at half the lengthwise half span divided by the first crack. No other cracks subsequently generated up to material aging of 700 days. The evaluation of these actually generated cracks will certainly reduce diffusion performance. However, the diffusion coefficient of the original zone can be considered very small enough to satisfy the requirement in the low diffusion layer, even though the cracks are accounted about 0.05% of the area in the surface of the low diffusion layer, assumed to be passing-through cracks. The results of crack observation by using a bore hole camera revealed an estimation of cracks not passing-through the member. Consequently, an evaluation that specifies the cracks as passing-through cracks is sufficiently conservative. The materials and mix proportion of the low diffusion layer selected based on the considerations above was evaluated and proved to exhibit the required crack control effect.

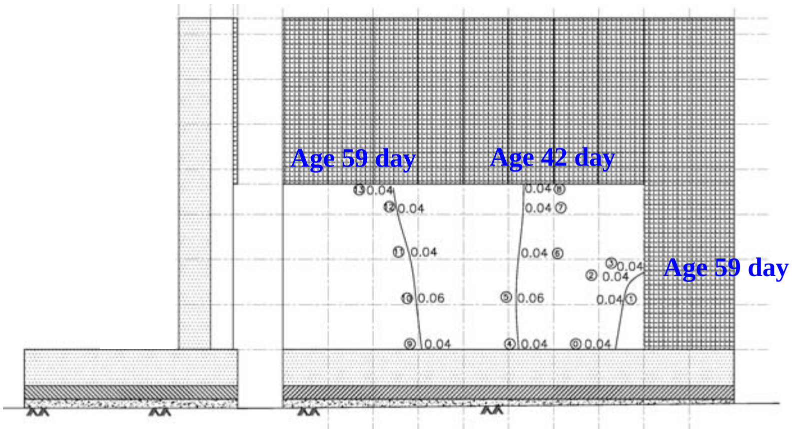


Figure 12 Crack generating situation

SUMMARY AND FUTURE CHALLENGES

It was demonstrated that the materials and mix proportion was evaluated as being appropriate and the low diffusion layer built by using a general method of construction could exhibit the required initial performance. In the future, evaluation studies will continue for the diffusion coefficient of the sound region and the effects of cracks and other defects that may occur due to the leaching and the corrosion of steel, which are deterioration phenomena.

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