QUALIFYING CONCRETE FOR A LOW-LEVEL WASTE REPOSITORY

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ABSTRACT

A waste repository for the belowground disposal of low-level radioactive waste, labelled IRUS (Intrusion Resistant Underground Structure), is planned at Chalk River Nuclear Laboratories. It relies greatly on the durability of concrete for a minimum of 500 years of service life. A research program based on laboratory testing to design a durable concrete and predict its useful engineered service life is in progress.

Durability of concrete depends on its resistance to deterioration from both internal and external causes. Since the rate of degradation depends to a major extent on the rate of ingress of aggressive ions into concrete, laboratory testing is in progress to establish the diffusion rates of ions, especially chlorides, sulphate and carbonate ions. A total of 1000 concrete specimens and 500 paste specimens are being exposed at 22° and 45°C to twenty-five different combinations of corrosive agents, including CO_2. Procedures to measure the ionic profile and to determine the factors controlling diffusion of ions in the various concretes have been developed. The paper presents the initial results from the research program and the longevity predictions to qualify concretes for the IRUS waste repository, based on twelve months of diffusion testing on laboratory specimens.

INTRODUCTION

Concrete durability depends on the quality of the concrete ingredients, their formulation, placement, and also on the service environment inside and outside of the repository, and hence is site dependent. AECL has identified various potential aggressive elements for the IRUS repository concrete at Chalk River Nuclear Laboratories [1]. Environmental factors considered included sulphate and chloride ion concentrations in ground water, carbonation effects, leaching of concrete, freeze-thaw deterioration, influence of microcracking due to design loads on the structure and alkali-aggregate reaction.

A cross-section of the IRUS disposal facility after closure is given in Fig. 1. The facility will be located in a free-draining sand dune, with the foundation of the repository placed one metre above the maximum recorded groundwater table. A plan of the facility during the operational phase is shown in Fig. 2. During the operational phase, the repository will be covered by a temporary weathershield building and served by a gantry crane. The bottom of the facility is designed to be permeable to allow any water that might enter the facility to drain freely rather than to remain in contact with the waste. Walls of the repository will be made of 0.6 metre-thick reinforced concrete, and the roof will have a minimum thickness of one metre. As an additional barrier to water infiltration, the reinforced concrete roof will be provided with a polyethylene sheet, 0.3 metre gravel layer, 1.1 metre of fine consolidated sand, and 0.3 metre of top soil. The additional cover materials will place the repository concrete below the frost level. Since the vault and roof are major components of the engineered barriers, the durability of concrete is an important aspect to the integrity of the disposal concept.

PROGRAM OBJECTIVES

The focus of the study is not on the development of a single highly durable concrete, but a methodology which would assess the durability aspects of a wide variety of concrete types and qualities subjected to different exposure conditions. This program has the following main objectives:

1. Design a concrete formulation for an engineered service life of 500 years or more based on the IRUS repository site environment. The intended duration of this research is 30 months.
2. Continue studies on a scientific basis on the durability aspects of concrete systems under different exposure conditions and create a data base. Predict the longevity of concrete types so as to design concrete formulations for selected repository environments. This study is of a generic nature and is expected to last 6 to 10 years.

PREDICTION METHODOLOGY

The prediction methodology basically involves establishing deterioration rate parameters of the concrete systems in a simulated repository environment. Deterioration due to physical agents such as freezing and thawing can be established for different qualities of concrete by standard laboratory tests. To simulate potential deterioration due to the chemical environment during the long service life, concrete specimens exposed to solutions containing selected

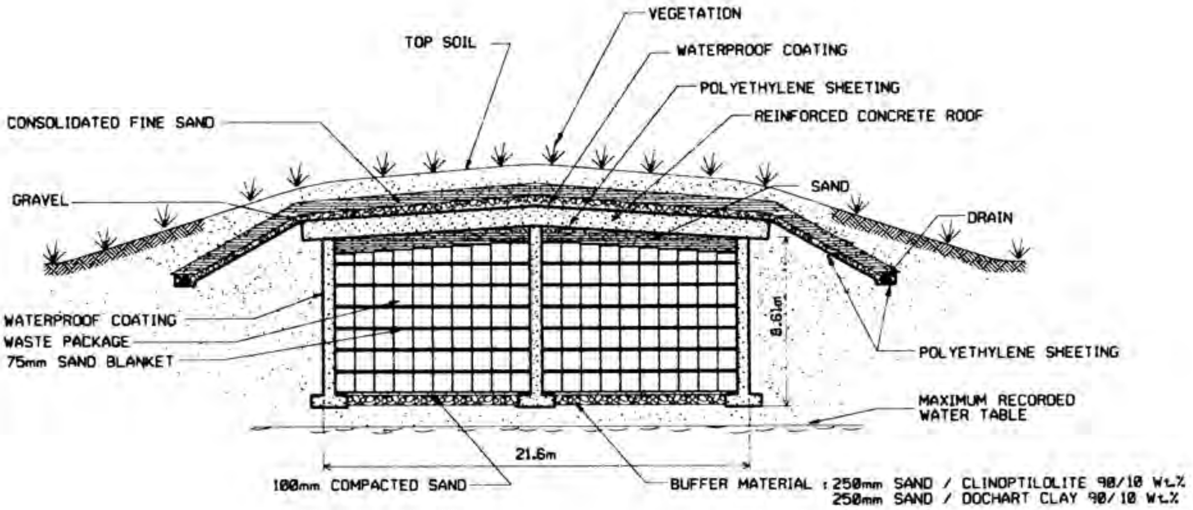


Fig. 1. IRUS Repository After Closure

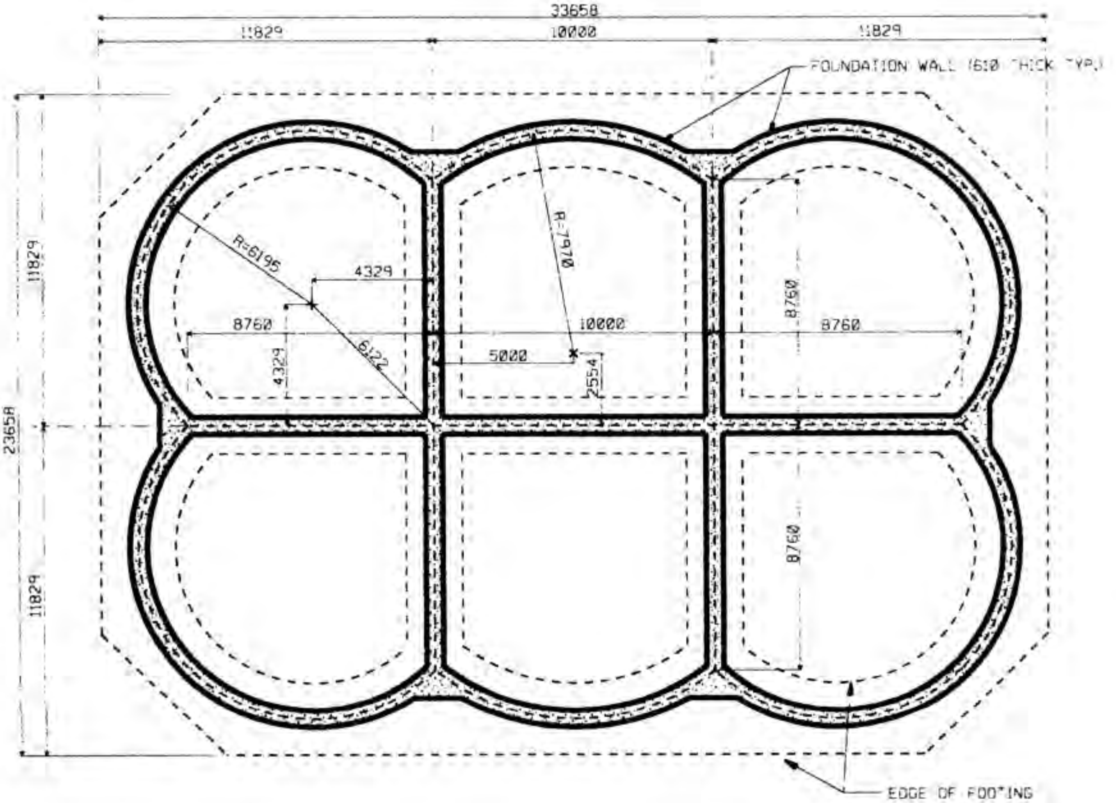


Fig. 2. Plan of IRUS Repository During Operation

ions and combinations of ions are to be studied. Since deterioration can be dependent on the depth of penetration of ions into the specimen, the rate of deterioration can be equated conservatively to the rate of penetration. If deterioration of a certain depth of concrete can be assumed as the failure of the repository structure, the time required for such an event or a longevity prediction can be made. The IRUS repository is being designed with a 75 mm concrete cover over the reinforcing bars. When the chloride ions diffuse through a distance of 75 mm, they will come into contact with the bars and will start the process of reinforcing steel corrosion. Once the steel corrosion has initiated, it is a matter of time before failure of the reinforced concrete vault occurs. For IRUS, a 75 mm depth of ionic ingress into concrete was selected as a conservative failure criteria for the preliminary longevity predictions.

Ionic ingress, in combination with other physical effects, can be considered for qualifying concrete for an engineered service life. The ionic ingress is a slow process and in the case of quality concrete systems is especially true. From the test results, it is clear that in a good quality concrete it will take many hundred years for the ions to reach the failure depth of 75 mm. Rate parameters are being established from short-term experimental data. Longevity of concrete systems can be predicted from equations developed either by statistical treatment of the data or by mathematical modelling. Confidence levels in these predictions will improve with the availability of longer-term data [1].

RESEARCH PROGRAM ON CONCRETE DURABILITY

Figure 3 shows the "Material Test Plan" for the study. The material testing is being done in 5 stages. In stage 1, concrete and paste systems with different parameters were manufactured to provide a total of 1000 concrete and 500 paste samples for durability testing. Mix designs for test samples incorporated Portland cement, sulphate resisting cement, fly ash, silica fume, slag and other additives. In stage 2, the specimens were subjected to material testing to determine freeze-thaw resistance, porosity, and compressive strength. In stage 3, the specimens were exposed to aggressive ions and ion combinations in 25 baths containing salt solutions of various concentrations. Specimens were taken out at various time periods of exposure and the reaction front was established using microprobe analysis techniques. A Cambridge Stereoscan S-250 Microscope was used for the examination, and a Tracor Northern TN 5500 Energy Dispersive X-Ray Analyser was used for the quantitative analysis. At each position along the profile, ion concentrations were expressed as mass percent of the paste. A value of 0.3% Cl and 3.0% of $SO_4^=$ was used to determine the position of the reaction front [2]. In stages 4 and 5, correlation of the data and longevity predictions are being at-

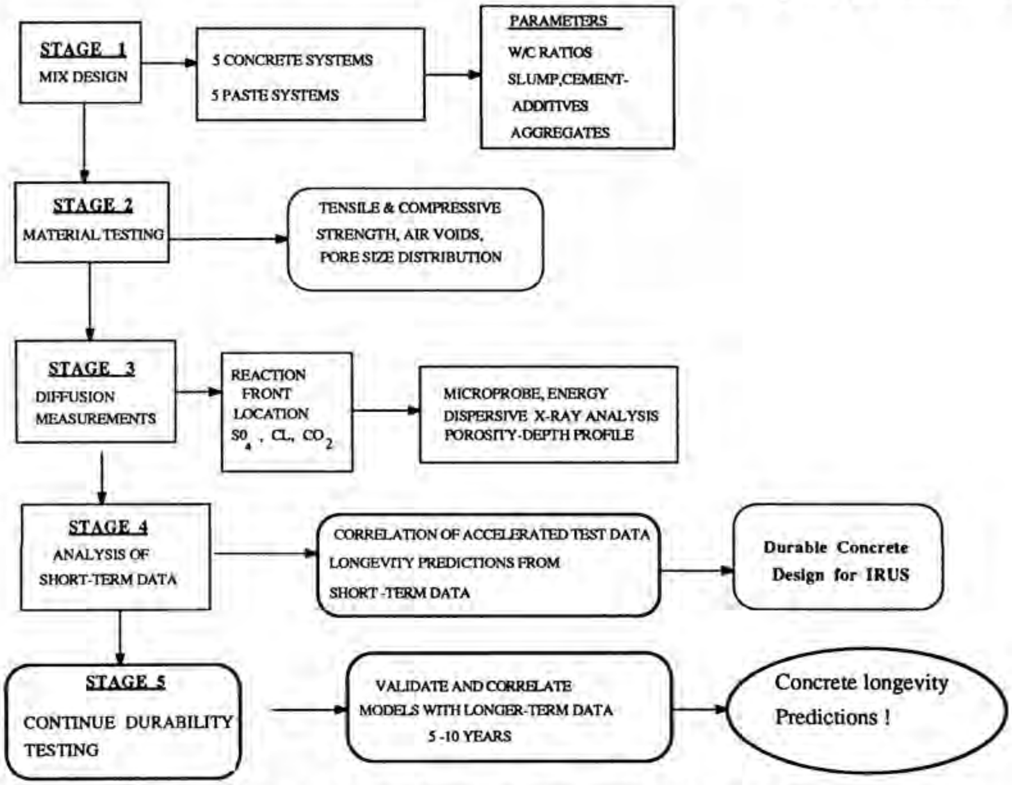


Fig. 3. Concrete Durability Research And Material Testing Plan

tempted. The research program is in stage 4 at present, and one-year diffusion test results are now available.

EXPERIMENTAL SET-UP

Binders

Portland cement (type 10), sulphate resisting cement (type 50), silica fume and blast furnace slag, and other additives were used for the study. The oxide analyses of the binders are given in Table I.

Aggregates

Unblended sand consisting mainly of quartz and feldspar was used. The potential alkali-reactivity of the sand was determined by accelerated mortar bar tests and monitoring length change during two weeks storage in 1 M NaOH solution at 80 degrees Celsius. The expansion of the specimens was well below the code-proposed limit of 0.1%, indicating innocuous aggregates [3].

Limestone coarse aggregate was also subjected to accelerated mortar bar testing and the standard concrete prism test. The expansion of concrete prisms after 8 weeks was just under 0.02%, compared to the code limit of 0.04%, and no further expansion was observed up to the present time (565 days) [4]. These results indicate that the aggregate is non expansive and that the selected limestone aggregate for IRUS from Arnprior, Ontario, would be unlikely to show expansion in concrete even in the long-service-life term. The specimens were also subjected to a magnesium sulphate soundness test, freeze-thaw cycling and petrographic evaluation, and the results were found to be satisfactory.

Concrete and Paste Systems

Table II shows the nomenclature and composition of the cement and paste systems. Two types of cement, the OPC (Type 10) and the sulphate resisting (Type 50), were used for the study. Systems 3 and 4 use slag replacing the cement content up to 75% by weight. The concrete and paste systems were designed with a water-to-cement ratio varying from 0.35 to 0.6 by weight.

TABLE I

Oxide Analysis Of Cements, Silica Fume And Slag

	Portland Cement Type		Slag	Silica Fume
	10	50		
SiO_2	19.43	20.71	35.30	95.17
Al_2O_3	4.18	3.77	10.62	0.21
Fe_2O_3	3.20	4.36	0.58	0.13
CaO	61.21	62.46	36.94	0.23
MgO	4.09	3.35	13.32	0.15
Na_2O	0.45	0.35	--	0.10
K_2O	0.89	0.87	--	0.27
C				1.56
L-O-I	1.53	0.88	1.16	2.30
SO_3	3.93	2.46	1.41	0.12
Free Lime	1.15	0.70	--	--

TABLE II

NOMENCLATURE AND COMPOSITION OF CEMENT SYSTEMS

		CEMENT		SLAG	FLY ASH	SILICA FUME	CURING TIME
S1	System 1	Type 10	100%	0	0	0	14 days
S2	System 2	Type 50	90%	0	0	10%	14 days
S3	System 3	Type 50	32%	65%	0	3%	28 days
S4	System 4	Type 50	65%	0	30%	5%	56 days
S5	System 5	Type 50	22%	75%	0	3%	28 days
			M1	Mix 1	0.35 w/c		
			M2	Mix 2	0.42 w/c		
			M3	Mix 3	0.50 w/c		
			M4	Mix 4	0.60 w/c		

Concrete Specimens

For each concrete mix, two concrete prisms 75x75x280 mm were cast for the study. All sides of the specimen were sealed with wax, leaving one long side (280 mm) unwaxed to allow unidirectional ingress of chloride and sulphate ions.

Compressive strengths of some of the test samples are shown in Table III. The compressive strengths were determined using the average value by loading 3 paste cubes to destruction. The results indicate the direct impact of water-to-cement ratio on the strength development. The strength increased in all test cases as the water-to-cement ratio decreased. The total porosity was evaluated using propan-2-ol as the displacement fluid, and microhardness measurements on paste systems 1 to 5. The porosity varies from 10% for system 5 to 50% for system 1, increasing with the water-to-cement ratio.

IONIC INGRESS

Depth of chloride ion penetration obtained from ionic profile data was plotted versus square root of time of exposure. This is conforming to the requirement for a diffusion based mechanistic model. The linear regression lines of results for exposure in Bath 15(49.49 g/L of NaCl at 22°C) are presented in Fig. 4 for S1(M(1-4)), Fig. 5 for S2(M(1-4)), and Fig. 6 for S5(M(1-4)). The data for the four experimental points up to 12 months are also presented in Table IV. It must be emphasized that the concentration of the chloride ion in the above Bath 15 is ten times the value identified as the worst case scenario for the IRUS repository. The linear regression analysis was carried out on each set of data using the starting time and zero ingress as the fifth point. Correlation coefficients, slopes, and intercepts of the regression line are included in Table IV. Values for ionic ingress after 500 years (77.46 months$^{1/2}$) exposure, are computed and also included in the table.

The correlation coefficients for S1(M(1-4)) are 1.00, 0.99, 0.99 and 0.97, respectively. Extrapolated values for ionic ingress after 500 years (77.46 months$^{1/2}$) exposure are 462, 697, 704 and 722 mm for S1M1, S1M2, S1M3 and S1M4, respectively. The correlation coefficients for S2 are all 0.90 or above, except M2, which is 0.84. The ingress values

TABLE III

Compressive Strength Of Paste And Concrete

Compressive strength of cement paste

Days	Mpa			
	S = System		M = Mix	
	S1M1	S1M2	S1M3	S1M4
1.0	37.7	25.5	17.1	11.5
3.0	43.4	31.5	23.4	16.8
7.0	49.0	34.9	26.7	19.0
	S2M1	S2M2	S2M3	S2M4
3.0	35.7	22.2	17.4	10.3
7.0	48.7	36.9	31.8	21.3
14.0	56.9	42.1	37.8	29.2
	S3M1	S3M2	S3M3	S3M4
3.0	13.4	8.7	5.3	3.2
14.0	48.2	35.7	25.2	20.2
28.0	58.0	45.9	32.5	26.0
	S4M1	S4M2	S4M3	S4M4
3.0	21.5	13.6	10.7	5.6
14.0	38.6	27.5	21.1	14.1
56.0	47.1	35.9	29.8	22.1
	S5M1	S5M2	S5M3	S5M4
3.0	11.4	7.7	5.2	2.5
14.0	46.1	34.6	25.3	18.6
28.0	54.6	42.2	32.6	26.3

Compressive strength at end of curing period

	paste	concrete
	Mpa	
S1M1	49.0	41.6
S1M2	34.9	31.1
S1M3	26.7	26.1
S1M4	19.0	22.6
S2M1	59.9	47.6
S2M2	42.1	42.1
S2M3	37.8	35.7
S2M4	29.2	31.3
S3M1	58.0	45.1
S3M2	45.9	41.0
S3M3	32.5	32.2
S3M4	26.0	26.8
S4M1	47.1	48.4
S4M2	35.9	39.0
S4M3	29.8	30.5
S4M4	22.1	31.3
S5M1	54.6	45.3
S5M2	42.2	36.1
S5M3	32.6	29.3
S5M4	26.3	20.0

extrapolated to 500 years are 255, 151, 355 and 382 mm for M(1-4), respectively, but M2 has some relatively low experimental points (Table IV) and the extrapolated values are probably too low. The ingress values for S2, however, are approximately half those of S1 for most mixes.

The correlation coefficients for S5 are all 0.92 or above except for M3. The extrapolated values of the ingress at 500 years exposure, however, are 132, 119, 107 and 171 mm for M(1-4), respectively, an average factor of about 5 times lower than S1.

Linear regression lines extending to 500 years are shown in Fig. 7 for S1, Fig. 8 for S2, and Fig. 9 for S3. Time required for the ionic ingress of 75 mm corresponding to the failure criteria is presented in Table V. From the table it can be observed that the rate of ingress of ions in S5M1 containing 75% blast furnace slag (replacing cement), is 14 times slower than that of S1M1, containing Portland cement with no supplementary materials added.

QUALIFYING CONCRETE FOR IRUS

To qualify a concrete for IRUS and establish its durabilty, deleterious effects of all relevant degradation mechanisms are to be considered in the evaluation. Microcracking due to internal strain from imposed loading on the structure is an important aspect to be considered. In a study jointly being conducted by AECL and the National Research Council Canada, and funded by EPRI, on "the effect of microcracking on reinforcement corrosion", it was found that microcracking due to design loads has a direct influence on the ionic ingress. The rate of ingress increased substantially with increased microcracking due to the higher stress levels in the specimen. Thus the longevity time prediction for any concrete under a given environment has to be adjusted to incorporate these additional effects. For IRUS, these effects are being investigated and will be incorporated in the final assessment. At present, the predictions are based entirely on the chemical attack due chloride ion diffusion into the concrete specimens.

Fig. 4 to 9: Penetration depths of chloride ions versus time$^{1/2}$ of immersion in Bath 15, regression lines.

The maximum chloride concentration expected during the service life of IRUS repository concrete was selected as 4.95 g/L of NaCl, and since this concentration is much lower than the ionic concentration in Bath 15, the rate of ionic ingress was much slower compared to the Bath 15 results.

TABLE IV

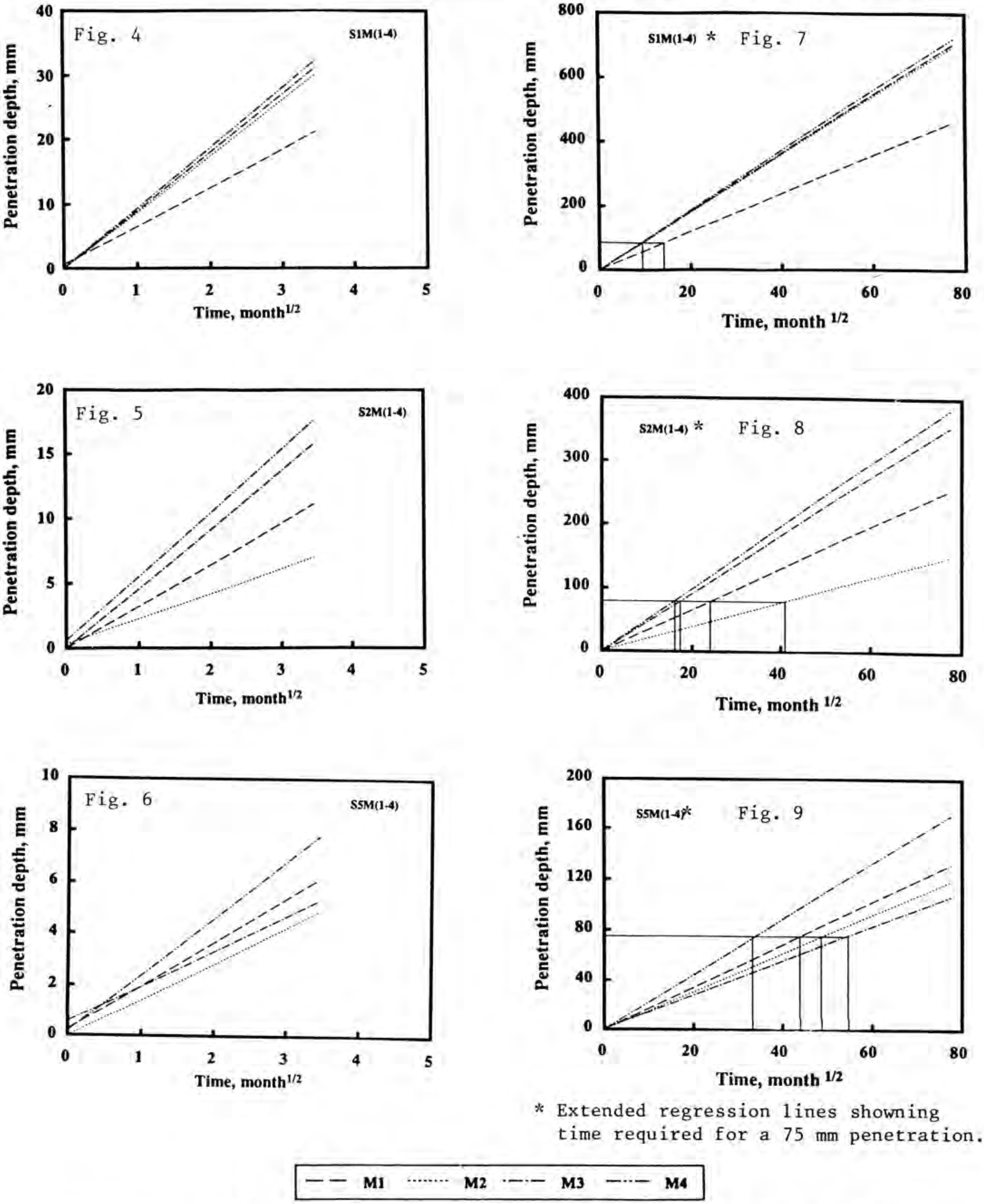
Chloride Ion Ingress (mm) In Concretes As a Function of Time of Immersion in Bath 15

Time$^{1/2}$ (months)$^{1/2}$	Mix 1 System			Mix 2 System			Mix 3 System			Mix 4 System		
	1	2	5	1	2	5	1	2	5	1	2	5
0.00	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
1.73	11.9	5.2	3.6	13.6	5.5	1.7	13.8	8.6	3.6	13.8	0.0	4.1
2.45	15.8	7.3	4.1	20.4	4.1	2.0	24.3	10.7	4.3	28.0	11.3	6.0
3.00	17.9	9.9	6.3	23.8	4.0	5.1	25.9	11.6	5.8	27.0	20.4	7.0
3.46	20.8	11.4	5.3	33.1	8.7	5.2	31.3	17.6	3.8	30.9	13.9	7.3
77.46	462	255	132	697	151	119	704	355	107	722	382	171
	Linear regression analysis											
Corr.Coeff.	1.00	1.00	0.96	0.99	0.84	0.92	0.99	0.97	0.87	0.97	0.90	0.99
Slope	5.96	3.29	1.70	9.01	1.94	1.55	9.09	4.58	1.37	9.31	4.92	2.20
Y-intercept	0.59	-0.24	0.25	-0.99	0.33	-0.49	-0.29	-0.06	0.57	0.12	0.61	0.19

TABLE V

BATH 15, Time Required For 75 mm Depth of Penetration

	MIX 1	MIX 2	MIX 3	MIX 4
	Time, Years			
SYSTEM 1	12	8	8	8
SYSTEM 2	44	124	22	19
SYSTEM 5	161	198	244	96



* Extended regression lines showning time required for a 75 mm penetration.

Fig. 4-9. Penetration Depths of Chloride ions Versus Time $^{1/2}$;of Immersion in Bath 15, Regression Lines.

The time required for 75 mm of depth of penetrations for the IRUS bath from the linear regression lines gives 1000 years for S5(M(1-2)), and for a 90% confidence limit of the data for a 75 mm depth of penetration provides 800 years, thus providing the indication that concrete can be qualified for an engineered service term of 500 years or more.

It is clear that having data for only one year decreases the level of confidence of the computations. Thus having only four experimental points increases the coefficient of variation and causes the prediction time for 75 mm penetration at 90% confidence to be low.

CONCLUSIONS

1. The rate of ingress of chloride ions into the three systems appears to be diffusion controlled.
2. The rates of ingress of chloride ions are decreased by the use of additions of silica fume and blast furnace slag to Type 1 cement.
3. The rate of ingress of chloride ions generally increases with water-to-cement ratio for the three systems.
4. Linear regression analysis of the limited data obtained by chloride exposure to 10 times worst case scenario indicates penetration of chloride ion as low as 107 mm in 500 years.
5. Linear regression analysis of the limited data obtained by exposure of specimens to IRUS bath indicates that time required for a 75 mm penetration can be over 1000 years.

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