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SEAWATER CORROSION TESTS
FOR
LOW-LEVEL RADIOACTIVE WASTE DRUM CONTAINERS

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日本原子力研究所
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SEAWATER CORROSION TESTS
FOR
LOW-LEVEL RADIOACTIVE WASTE DRUM CONTAINERS

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(Received October 15, 1985)

This report is a part of corrosion tests of drums under various environmental conditions (seawater, river water, coastal sand, inland soil and indoor and outdoor atmosphere) done at Japan Atomic Energy Research Institute sponsored by the Science and Technology Agency. The corrosion tests were started in November, 1977 and completed at March, 1984.

This report describes the results of the seawater corrosion tests which are part of the final report, "Corrosion Safety Demonstration Test"(Japanese), and it is expected to contribute the safety assessment of sea dumping of low-level radioactive waste packages.

Keywords: Corrosion Test, Radioactive Waste, Safety Evaluation,
Waste Disposal, Waste Storage, Demonstration Test,

放射性廃棄物ドラム缶容器の海水中腐食試験

日本原子力研究所東海研究所環境安全研究部

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(1985年10月15日受理)

昭和52年度から昭和58年度まで、科学技術庁の委託により、種々の環境条件下（海水、陸水、海岸砂、土、室内及び野外大気）における低レベル放射性廃棄物ドラム缶容器の耐食性実証試験を行った。本報告はこの内の海洋処分に対応する部分に関するものである。約5年間の海水中腐食試験結果をもとに、長期にわたる海水中におけるドラム缶の腐食進行状態の推定を行った。

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1. Preface

The amount of low-level radioactive waste has increased year after year with the increasing use of nuclear power generation, and the measures for their disposal have become an important problem in the country's effort to promote nuclear power generation.

Regarding the disposal of low-level radioactive waste, the Japan Atomic Energy Commission, in October 1976, gave a principle for the disposal of this waste on land and at sea. As for disposal at sea, the criteria for it are given in Ordinance No. 56 of the Prime Minister's Office "Rules for Waste outside Nuclear-fuel-material-handling Factories or Works" (December, 1978) and in the Notification No. 9 of the Science and Technology Agency, Prime Minister's Office "Determination of Detailed Waste Treatment and Disposal Techniques outside Nuclear-fuel-material-handling Factories or Works" (December, 1978). The Radioactive Waste Disposal Ad Hoc Committee, Atomic Energy Commission (in June, 1982) discussed land disposal by utilizing out-of-site storages, trenches, pits, structures or underground caves.

The corrosion resistance of low-level radioactive waste drum containers under a disposal environment is an important item for evaluating the safety of this type of disposal system. Although several test examples on the corrosion resistances of low-level radioactive waste drum containers are available, systematic, quantitative data on this subject do not exist. With regard to radioactive waste disposal, such natural conditions as geology, topography and climate, and the siting and social conditions in Japan are more severe than in the United States and almost all European countries, necessitating safety evaluation with

a much higher accuracy.

These "Corrosion Safety Demonstration Tests" were carried out to demonstrate the corrosion resistances, under various environmental conditions (seawater, river water, coastal sand, inland soil, and indoor and outdoor atmosphere), of drum containers in which low-level radioactive waste are enclosed, verifying thereby their corrosion integrity under various environmental conditions.

In the fiscal 1977 special account budget for promoting electric power development, the trust contract for the radioactive waste safety demonstration tests was allocated, followed by the conclusion of the fiscal 1977 trust contract between the Science and Technology Agency and the Japan Atomic Energy Research Institute on the safety demonstration tests for low-level radioactive-waste-enclosing containers "Corrosion Safety Demonstration Tests" in November, 1977. Since then, a survey on the tests,¹⁾⁻⁶⁾ the construction of laboratory and installation facilities for the testing equipment have been carried out. A series of demonstration tests were started in November, 1978 and completed at the end of fiscal 1983. This paper describes the results of the seawater corrosion tests which are part of the final report", "Corrosion Safety Demonstration Tests".

2. Corrosion Safety Demonstration Test Committee

After completion of the trust contract for the "Corrosion safety demonstration tests", the "Corrosion Safety Demonstration Test Committee" consisting of many competentmen was set up at our institute in May, 1978. Since then, seventeen committee meetings have been held up to the end of the tests in March, 1984 to discuss the testing program and the results of tests.

The contents of the report were approved by the committee after very detailed discussions by the committee members and the researchers responsible for the tests.

3. Test Bodies

3.1 Drum Containers

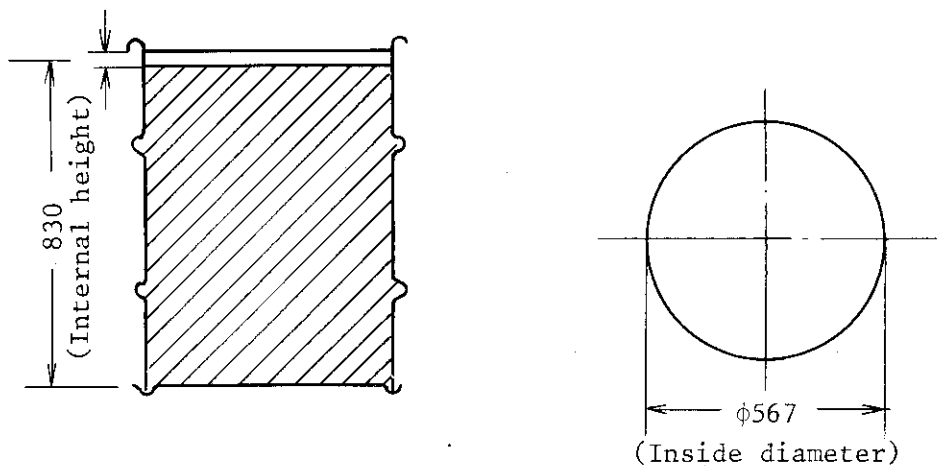
The drums used for the low-level radioactive waste container were employed with simulated cemented waste enclosed (Fig. 3-1). The drums were 200-l, open-type ones mainly of the H-grade type (1.6-mm thick steel plate) with one kind being of the M-grade type (1.2-mm thick steel plate) for comparison purposes. The material of the drums (including the bands) consisted hot-rolled mild steel and first-class band steel (SPHC) specified in JIS G3131-1977. In addition, the stainless steel drums used partly were made of cold-rolled stainless steel plates specified in JIS G4305-1977.

Although coating for the drum containers is not specified in JIS Z1600-1977, several anticorrosion-treated or coated test bodies were tested in addition to the ones coated with melamine coatings used widely in nuclear power plants. If possible, the anticorrosive drum containers should be put into practical use without any remarkable alterations in the present mass production modes and costs; in other words, with only minor modifications to the conventional drum containers. For this purpose, five common anticorrosion methods were chosen and applied to the drum containers: they involve painting, electric anticorrosion, galvanization, rust-free metals, etc. Non-Coated drum containers (M-and H-grade) were tested for comparison purposes, too. The specifications of the drum containers used for tests are shown in Table 3-1.

3.2 Test Pieces

In the corrosion tests for the drum containers enclosed with

simulated cemented waste, it was difficult to perform a variety of tests on them because of the limitations in the testing periods and the number of test bodies, and to measure directly the weight changes and minute surface changes. For the purpose of supplementing the drum container tests, test pieces were prepared and tested. As shown in Table 3-2, the test pieces used were made of the same materials as those of the test drum containers. The plates were 150-mm in length, 75-mm in width and 1.6-mm in thickness to which various equivalent surface treatments and coatings were applied. In addition, stainless steel was also used for comparison purposes. The protective coatings of drum containers are often damaged during handling before the containers are disposed of. To examine the effects of the damaged coatings before disposal concerning corrosion, artificially damaged test pieces, as shown in Fig. 3-2, were used in comparison with those that were undamaged.



Item		Specifications	Remarks
Kind of drum		H class (200 ℓ)	In accordance with JIS Z 1600-1979
Cement-solidified simulated wastes	Normal portland cement	28.45	
	River sand (max. particle size, 5 mm)	59.28	
	Water	11.86	
	Sodium sulfate (anhydrous)	0.35	
	Ethyl alcohol	0.06	
	Total	100.00	
	Uniaxial compressive strength	150	28-day curing

Fig. 3-1 Specifications of simulated low-level radioactive waste enclosed in a drum.

Table 3-1 Kinds and specifications of test drums

Kind	Test drum No.	Classification of anticorrosion treatment	Drum grade	Steel plate thickness (mm)	Steel plate treatment	External-surface coating			Internal coating			Number of tested drums								
						Undercoating	Surface coating	Total film thickness (Average) (u)	Undercoating	Surface coating	Total film thickness (Average) (u)	Sea water tank	Beach tank	Soil tank	Indoor (I)	Indoor (II)	Outdoor air	Total		
Drum for comparison	1	Non-coated	M	1.2	Derusting	—	—	—	—	—	—	1	1	1	1	0	0	5		
	2	"	H	1.6	"	—	—	—	—	—	—	1	1	1	1	0	2	7		
Present drum	3	Coated	M	1.2	Zinc phosphate film treatment	—	Single coating with baked-on melamine	32	21	27	—	Double coating with baked-on epoxy	28	2	2	2	1	1	12	
	4	"	H	1.6	"	—	"	36	29	24	—	"	25	2	2	2	2	1	12	
Anticorrosion drum	5	"	H	1.6	Sand blast	Zinc-bearing anticorrosion primer	Double coating with baked-on epoxy	218	215	195	Zinc-bearing anticorrosion primer	"	156	2	2	2	2	1	1	12
	6	Electric anticorrosion treatment	H	1.6	Zinc phosphate film treatment	—	Single coating with baked-on melamine	22	20	21	—	"	50	2	2	0	2	0	0	6
Anticorrosion drum	7	Zinc metallic	H	1.6	Sand blast	Zinc metallic anticorrosion primer	Single coating with baked-on epoxy	180	207	170	Zinc-bearing anticorrosion primer	Single coating with baked-on epoxy	66	0	0	2	0	1	1	5
	8	Galvanization	H	1.6	Derusting	Galvanizing	Single coating with baked-on epoxy	60	50	52	Galvanizing	Single coating with baked-on epoxy	45	2	2	2	2	1	1	11
	9	Stainless steel (SV304)	H	1.5	"	—	—	—	—	—	—	—	—	2	2	2	2	1	1	11

Table 3-2 Specification of test piece coating

Test piece No.	Classification of anticorrosion treatment	Steel plate thickness (mm)	Steel plate treatment	Coating		Total film thickness (μ)
				Undercoating	Surfacecoating	
2	without coating	1.6	Derusting	—	—	—
3, 48	Coating	1.2	Zinc phosphate treatment	—	Single coating with baked-on melamine	20 ~ 30
4, 5, 47	Coating	1.6	Zinc phosphate treatment	—	Single coating with baked-on melamine	20 ~ 30
54	Coating	1.6	Sand blast	Zinc-bearing anticorrosion primer	Single coating with epoxy	120 ~ 180
55	Coating	1.6	Sand blast	Zinc-bearing anticorrosion	Single coating with tar-epoxy	200 ~ 280
56	Coating Electric anticorrosion treatment	1.6	Zinc phosphate treatment	—	Single coating with baked-on melamine	20 ~ 30
57	Coating Zinc metallic	1.6	Sand blast	Zinc metallic	Single coating with epoxy	100 ~ 150
43	Coating Galvanization	1.6	—	—	Single coating with baked-on melamine	20 ~ 30
41, 62	Stainless steel	1.5	—	—	—	—

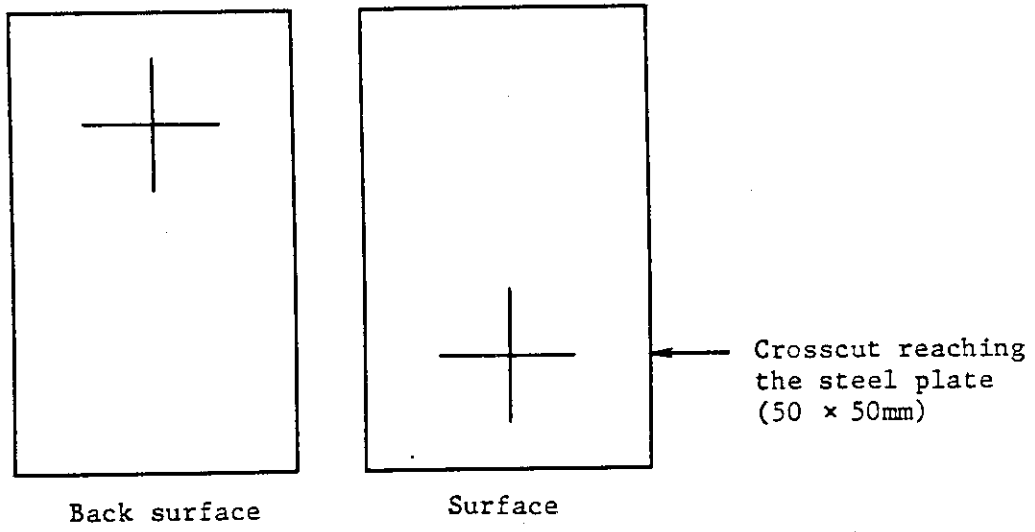


Fig. 3-2 Artificially damaged test pieces.

4. Testing Methods and Results

4.1 Drum Containers

4.1.1 Testing Methods

The simulated-contaminated-waste-enclosed drum container shown in Table 3-1 was immersed in a 10-compartment seawater testing tank shown in Fig. 4-1. The tank was further divided into two 5-compartment through which seawater was allowed to flow at a rate of 1 to 3 m/sec. The temperature of the seawater in the tank was controlled at 25 ± 5 °C by a temperature controller, and the environmental conditions such as temperature, pH, dissolved oxygen and electric conductivities were periodically measured.

4.1.2 Environmental Conditions

The drum containers were immersed in seawater under the environmental conditions shown in Table 4-1. The environmental conditions during the seawater immersion tests are given in Figs 4-2 to 5 and Table 4-2.

(1) Temperature

The temperatures of the seawater in the tanks were measured at the inlets and outlets of the No. 1 and No. 2 tanks. The temperature change during the test period is shown in Fig. 4-2, indicating that the average seawater temperature throughout the testing period was about 2.3°C.

(2) Dissolved Oxygen

The seawater was automatically sampled at the inlet and outlet of the tank to automatically measure the dissolved oxygen by using a dissolved oxygen meter. The concentration of dissolved oxygen was 1.0 to 2.0 ppm lower at the outlet than at the inlet up to 600 days after the

start of the test, with the difference becoming gradually smaller. The average amount of dissolved oxygen through the test period was about 8 ppm.

(3) Acidities

Like in the amount of the dissolved oxygen, seawater samples for pH measurement were taken automatically at the inlet and outlet of the tank, and the pH values were automatically measured by a pH meter. As shown in Fig. 4-4, the pH values during the test period were fairly stable, ranging from 7.4 to 8.1.

(4) Electric Conductivities

Seawater samples were automatically taken at the inlet and outlet of the tank, and measured automatically by an electric conductivity meter. The results obtained during the test period are shown in Fig. 4-5, which indicates electric conductivities of 35 to 43 m Ω /cm with no great variation.

(5) Chemical Composition of the Seawater

The sea water to be tested was taken from the waste cooling water of JPDR (Japan Power Demonstration Reactor) and differs a little from ordinary seawater; namely, the seawater for testing included sodium hypochlorite added intentionally to prevent marine life from breeding, and a minor amount of iron and other fine metallic particles because the seawater had been used for cooling. To examine these problems and perform tests under the same conditions as those of ordinary seawater, the followings were checked in advance:

- i) Sodium hypochlorite added to the seawater for testing is considered to be almost decomposed when it is used for testing. If sodium

hypochlorite remains, it has a bad effect concerning corrosion, so that the seawater was used after confirming the presence of sodium hypochlorite by measurement when the tank became full.

ii) A filter was provided at the inlet of the tank to filter iron and other metallic particles.

iii) Seawater was periodically analyzed. Table 4-2 shows the analytical results of the seawater for testing. No special component was observed when compared with ordinary seawater.

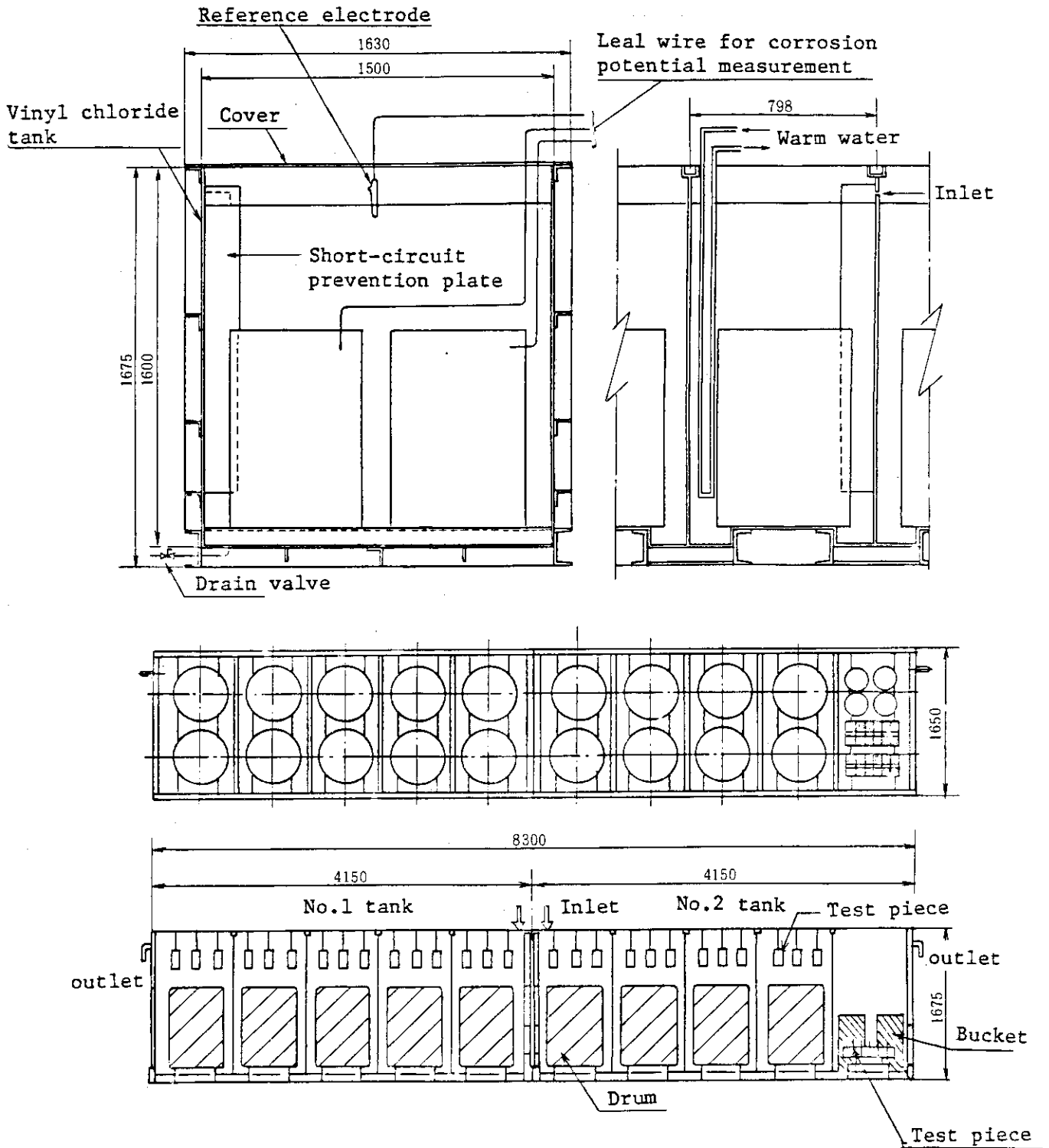


Fig. 4-1 Seawater tank for testing the drums.

Table 4-1 Comparison using environmental conditions between ocean disposal and drum tests.

Euvironment	Disposal environment	Testing environment	
		Target	Measure value*
Corrosion factor	Deep sea: 4,000 m or over		
Pressure	> 400 kg/cm ²	< 1 kg/cm ²	
Temperature	2 ~ 5 °C	25 ± 5 °C	23 °C
Dissolved oxygen	7 ~ 8 ppm	5 ~ 10 ppm	8 ppm
Flow rate	About 2 cm/sec	2 cm/sec	1 ~ 3 cm/sec
Acidity	pH 7 ~ 8	pH 7 ~ 8	pH 7.8
Electric conductivity	—		40 m /cm
Water quality	Deep sea condition		
Sediment	Deep sea condition		
Sea bottom soil	Existence of soft mud		

* Average measured value for 4 years and 7 months

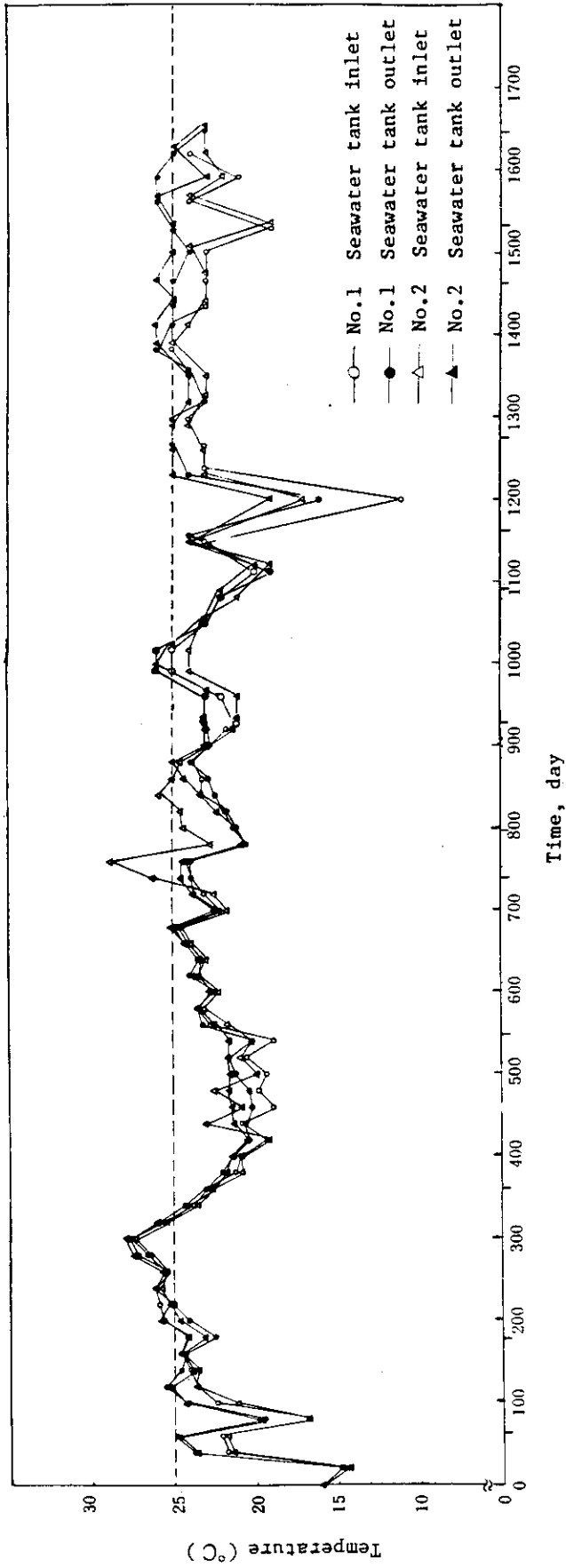


Fig. 4-2 Seawater temperature during the seawater corrosion tests.

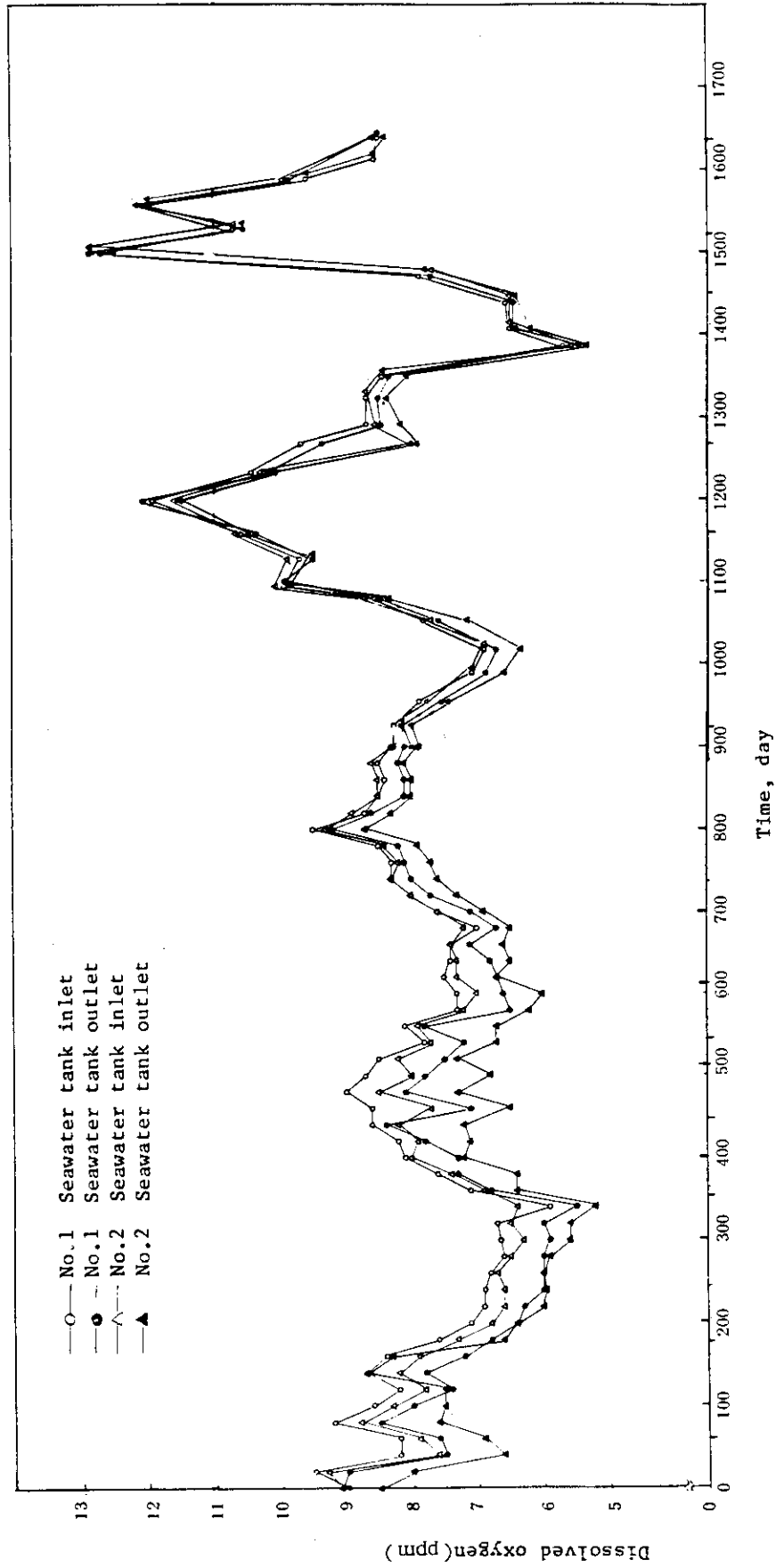


Fig. 4-3 Dissolved oxygen in seawater in the seawater corrosion tests.

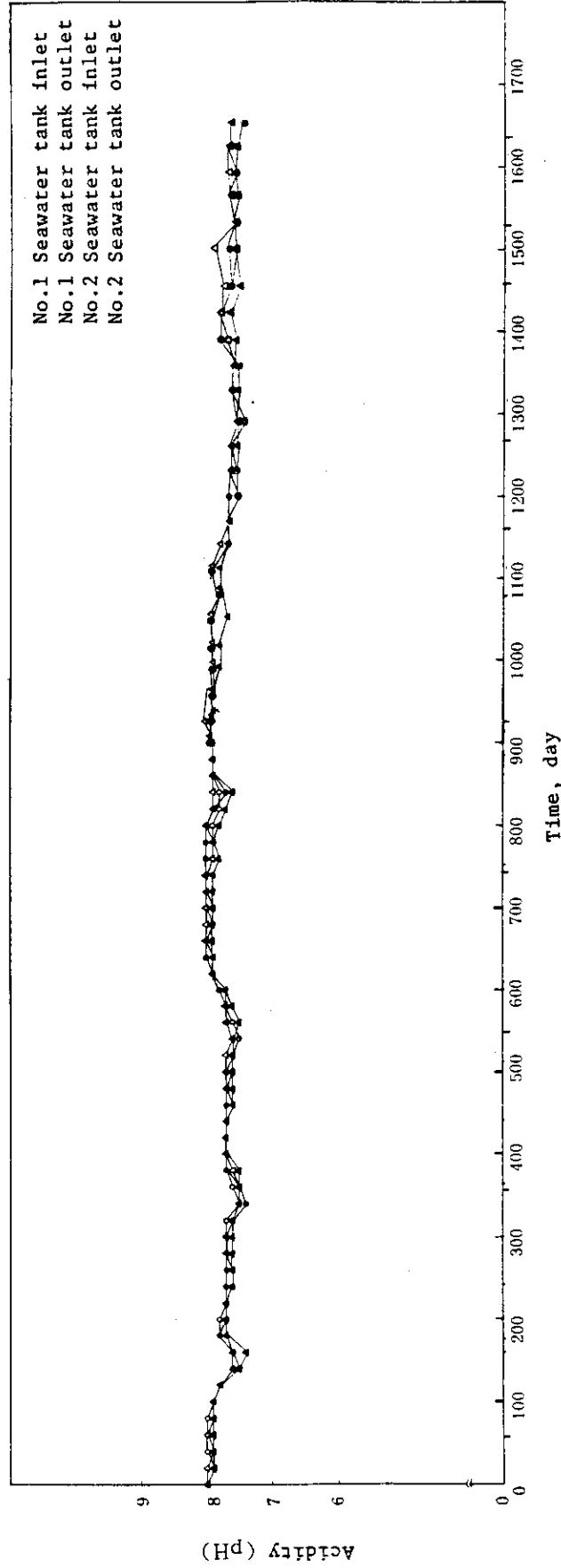


Fig. 4-4 Seawater acidity in the seawater corrosion tests.

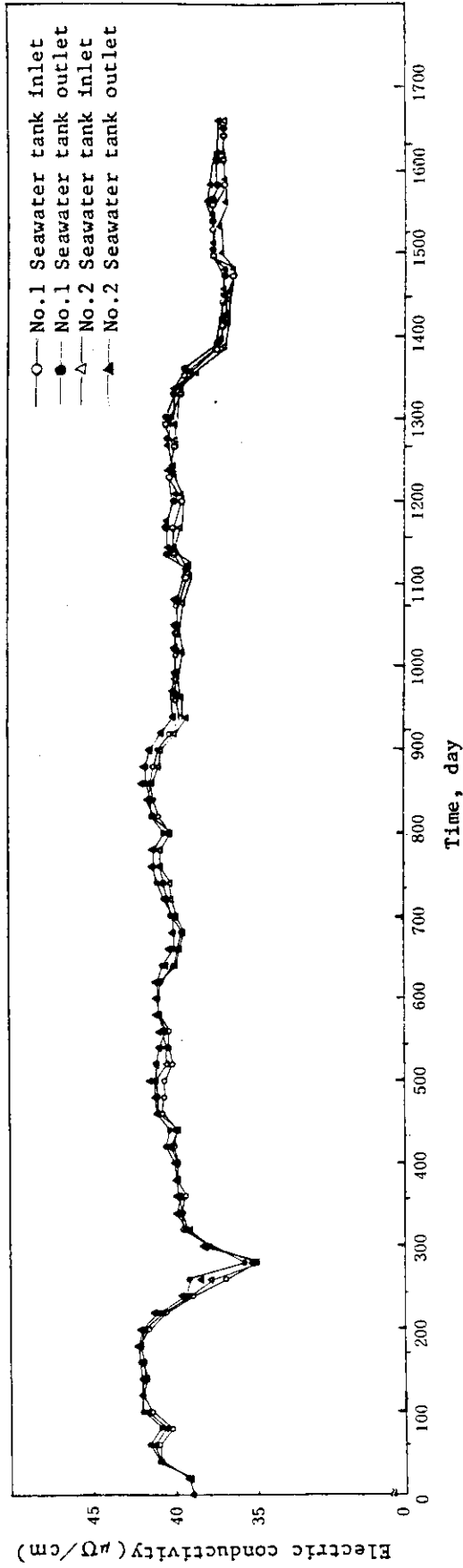


Fig. 4-5 Electric conductivities of seawater in the corrosion tests.

Table 4-2 Analyses of seawater for testing.

Component	Analyses (mg / l)							Analytical method
	Sampled on Jan. 8, '79; analyzed on Jan. 30, '79	Sampled on Dec. 6, '79; analyzed on Dec. 19, '79	Sampled on Feb. 14, '80; analyzed on Feb. 21, '80	Sampled on Jan. 30, '80; analyzed on Aug. 20, '80	Sampled on Nov. 4, '80; analyzed on Nov. 24, '80	Sampled on Dec. 11, '81; analyzed on Dec. 26, '81	Sampled on Dec. 10, '82; analyzed on Jan. 31, '83	
Sodium ^(Na)	10,000	—	—	9,600	10,500	9,500	11,000	JIS-K0101,36
Potassium ^(K)	750	—	—	450	420	390	390	JIS-K0101,37
Total nitrogen ^(T-N)	0.10	0.17	0.25	0.21	0.11	N.D	0.11	JIS-K0101,37 Total organic nitrogen
Nitrogen as ammonia ^(NH₄-N)	N.D	N.D	N.D	N.D	N.D	N.D	N.D	JIS-K0102, 17-1-2-13
Nitric acid ^(NO₃)	0.40	—	—	—	—	0.09	0.18	JIS-K0102, 17-3-1
Chlorine ^(Cl)	19,227	—	—	18,300	19,100	17,900	18,600	JIS-K0101,25-2
Residual chlorine ^(Cl₂)	< 0.1	N.D	N.D	N.D	N.D	N.D	N.D	JIS-K0101,22-1
COD	1.8	0.7	0.7	1.2	0.5	1.6	1.1	JIS-K0101,16: KMnO ₄ consumption at 1,100 °C
Residue on evaporation	38,685	—	42,991	41,040	48,900	36,800	38,050	JIS-K0101,15-1
Suspended substance ^(SS)	4.0	—	1.0	< 1	2.0	2.0	1.0	JIS-K0101, 5-5-2
Dissolved oxygen ^(O₂)	—	—	5.1	5.4	5.7	5.4	6.7	JIS-K0102,24-3
BOD	—	—	< 1	< 1	< 1	< 1	< 1	JIS-K0102,16
Sulfuric acid ^(SO₄)	2,520	2,440	2,650	2,570	2,640	2,510	2,640	JIS-K0101, 31-1-2
Phosphoric acid ^(PO₄)	H.D	—	H.D	H.D	H.D	H.D	H.D	JIS-K0101,32-1
Iron ^(Fe)	0.12	0.24	H.D	0.15	H.D	H.D	0.50	JIS-K0102,47-2
Silicic acid ^(SiO₂)	< 2	H.D	H.D	0.63	1.3	0.2	0.8	JIS-K0101, 33-1.2
Calcium ^(Ca)	630	—	377	364	376	364	380	JIS-K0101,38
Magnesium ^(Mg)	1,600	—	1,324	1,250	1,260	1,280	1,240	JIS-K0101,39
Copper ^(Cu)	N.D	N.D	N.D	N.D	N.D	N.D	N.D	JIS-K0102,37-2
Zinc ^(Zn)	0.12	0.05	0.04	0.68	0.03	0.63	0.03	JIS-K0102,38-2
Nitrogen as nitric acid ^(NO₃-N)	—	0.17	0.09	0.05	0.01	0.02	0.04	Method specified in 8.11 of the marine observation guideline

4.1.3 Corrosion Potentials

The corrosion potentials of drum containers were measured to estimate the corrosion tendency of the drum containers by the seawater immersion tests.

4.1.3.1 Measuring Methods

After connecting lead wire to the upper part of the external drum surface, the corrosion potential of the drum container immersed in the seawater was measured once a day automatically by computer control with a silver-chloride electrode being a reference. For the measurement of potentials the electrometer HE-102 fabricated by Hokuto Denko Co., Ltd. was used.

4.1.3.2 Results

The results of the measurement are shown in Fig. 4-6, in which the average value for two same-specification drum containers is given.

Though a highly durable silver chloride electrode was used as a reference for the measurement, the results thus obtained were converted to the values on the calomel electrode basis ⁷⁾ and shown in Fig. 4-6. The trends in corrosion of various test bodies estimated from the measured corrosion potentials are shown below.

(1) Non-coated Drum Containers

The No. 1 (without coating, $t \approx 1.2$ mm) and No. 2 test bodies (without coating, $t \approx 1.6$ mm) showed corrosion potentials of 500 to 600 mV immediately after the start of the tests through their ends. These values corresponded to the corrosion potentials of the stainless steel, of which the drum containers are made, in the seawater tank.

(2) Ordinary Drum Containers

The No. 3 (melamine coating, $t \approx 1.2$ mm) and No. 4 (melamine

coating, t ... 1.6 mm) test bodies show a potential around 650 mV 100 days after the start of tests, followed by 580-to 600-mV potentials, which are nearly the same as those in the No. 1 test body, to the ends of the tests. A series of these facts suggests that the corrosion of the stainless steel plate itself proceeded to some degree. In addition, the stable potential of this test body may indicate nearly uniform corrosion. In addition, a potential of about 900 mV immediately after the tests may probably be that of zinc galvanized over the bands, and decreases to a constant value as the zinc dissolves.

(3) Anticorrosive Drum Containers

The corrosion potential of the No. 5 test body is around 650 mV 100 days after the start of the test, indicating the progress in corrosion of the steel plate is like the ordinary melamine-coated drum container. The results of external appearance examination (See 6.1.4) suggest that since little deterioration is observed in the coating itself over the test body instead of the generation of iron rust on the band, the potential is responsible for the iron band after the dissolution of the galvanized zinc.

The No. 6 test body (electrical anticorrosion treatment & melamine coating) shows a stable potential of about 900 mV through the test period. The potential clearly corresponds to that of zinc, showing the continuation of the sacrificial anode reaction. This estimation may be endorsed by the fact that no rust generation was observed inspite of nearly complete spallation or relief of the melamine coating.

The No. 8 test body (galvanization & melamine coating) showed a potential of around 900 mV up to 400 days after the start of the test, but changed rapidly to around 600 mV, which is nearly equal to that of an ordinary melamine-coated drum container, after about 580 days. This fact suggests that the steel-plate-protecting function of the galvanized

zinc layer begins to deteriorate after 400 days and is completely disappears after 580 days, agreeing with the results of the visual observation.

The No. 9 test body (stainless steel) showed a constant potential of about 300 mV throughout the test period. The bent in the upper part of the test drum is provided with mild-steel core wire (see Photo. 4-5), and is attacked severly, including the nearby stainless steel. A potential of 300 mV may indicatc severe corrosion.

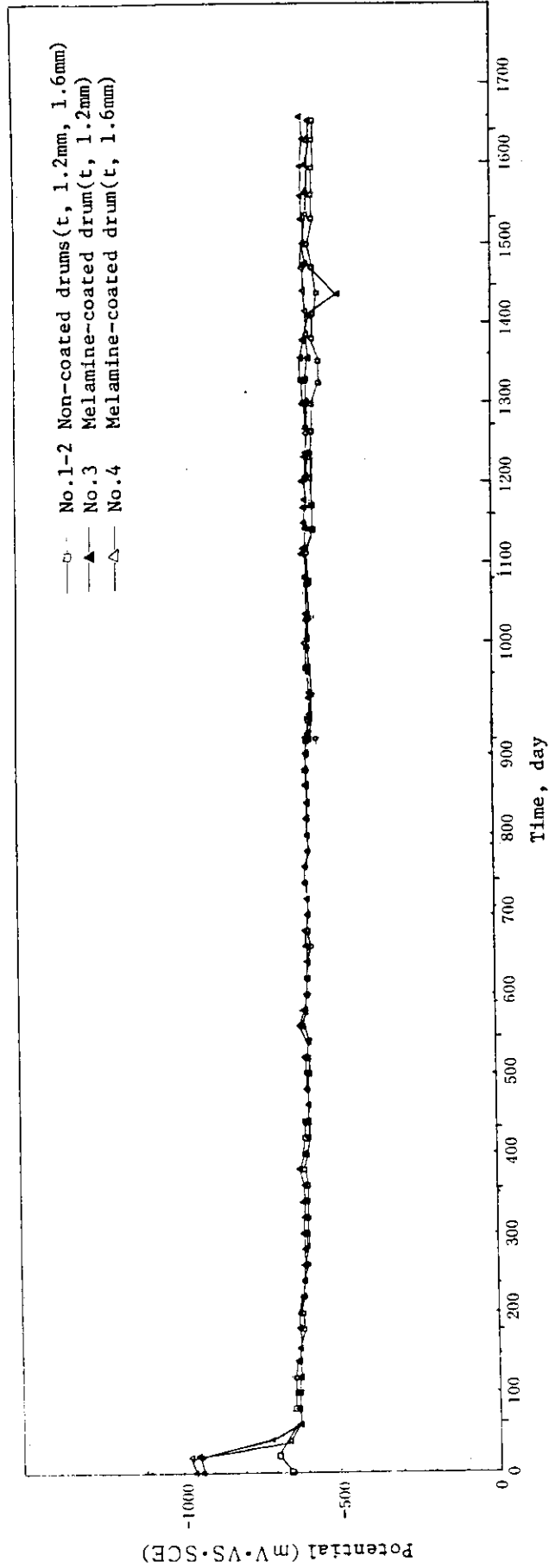


Fig. 4-6-(1) Corrosion potential of the drums in the seawater corrosion tests.

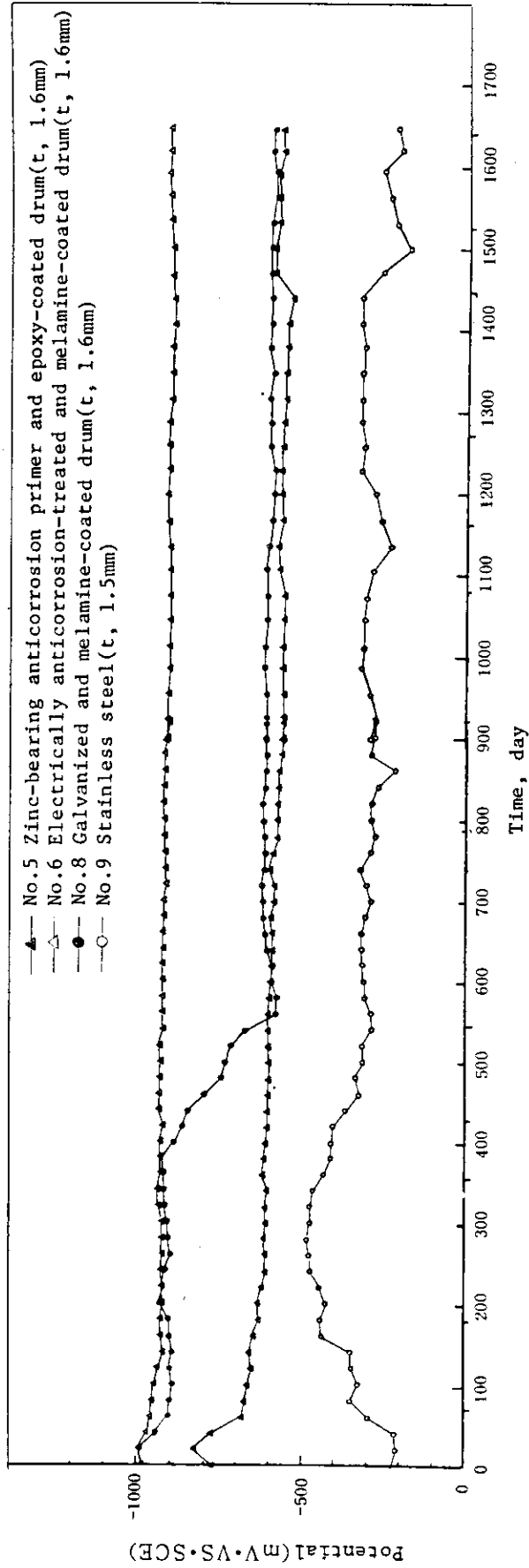


Fig. 4-6-(2) Corrosion potential of the drums in the seawater corrosion tests.

4.1.4 External Appearance Examination

4.1.4.1 Examination Methods

With regard to the drum containers used in the seawater immersion tests, after the seawater in the testing tank was completely drained, the top plates and drum shells were visually observed and they were photographed.

As to the external appearance examination after the seawater immersion tests, the external surfaces of the top and bottom plates as well as the shells were visually observed and photographed, the shells were longitudinally cut, the simulated cemented waste was removed from the drum containers, and the internal surfaces of the bottom plates and shells were visually observed and photographed.

4.1.4.2 Results

The results of visual observation (Photos 4-1 and 4-2) for the drum containers after the seawater immersion tests is summarized as follows:

(1) External Surfaces

The surfaces of the No. 1 (without coating, $t \approx 1.2$ mm) and No. 2 test bodies (without coating, $t \approx 1.6$ mm) were covered with thick rust, suggesting severe corrosion.

Regarding the No. 3 (melamine coating, $t \approx 1.2$ mm) and No. 4 test bodies (melamine coating, $t \approx 1.6$ mm), rust usually generated around the upper bents, welds and spliced parts to which stress was concentrated. Considerably large amounts of rust generated on the top plate, because the coating film was damaged when impurities and corrosion products settled over the top plate were removed at the time of the visual observation and thereby deterioration of the film proceeded. Compared with these parts, rust generation in the shell was small (see (Photo. 4-1)).

In the No. 5 test body (epoxy-resin coating), small blisters occurred around the peripheral part of the top plate and spliced parts, but rust rarely occurred and, accordingly, the coating film itself was at any rate sound (Photo. 4-2).

As for the No. 6 test body (electrical anticorrosion treatment & melamine coating), blisters and spallings were generated over the whole coating film but no rust, and consequently, the steel plate itself was sound. In general, the electric anticorrosion treatment may sometimes make the surface of the steel plate alkaline, leading to the generation of blisters and spallings because of poor alkali resistance of the melamine coating film. This may be why a great deal of spallings occurred on the coating film over the No. 6 test body. Since the steel plate was protected from rust generation by the sacrificial anodic reaction of zinc, no rust generated on the steel plate in spite of the blisters and spallings on many parts of the coating film.

Many blisters as well as rust were found over the whole surface of the No. 8 test body (galvanization & melamine coating). Like in the No. 3 and No. 4 melamine-coated drum containers, rust tended to generate on the top plate rather than on the shell, because damage was caused when impurities and corrosion products settled over the top plate were removed.

The inside of the upper bent of the No. 9 test body (stainless steel) was provided with mild-steel core wire (See Photo. 4-4), and rust by corrosion was observed. Over the surface of the test body a thin layer of adhered substances consisting mainly of calcium carbonate was formed, with the substance mixed with the rust. Therefore, although rust was seemingly generated over the test-body surface, it can be readily removed, and the surface thereby reveals itself to be sound and still keeps the luster of stainless steel.

(2) Internal Surfaces

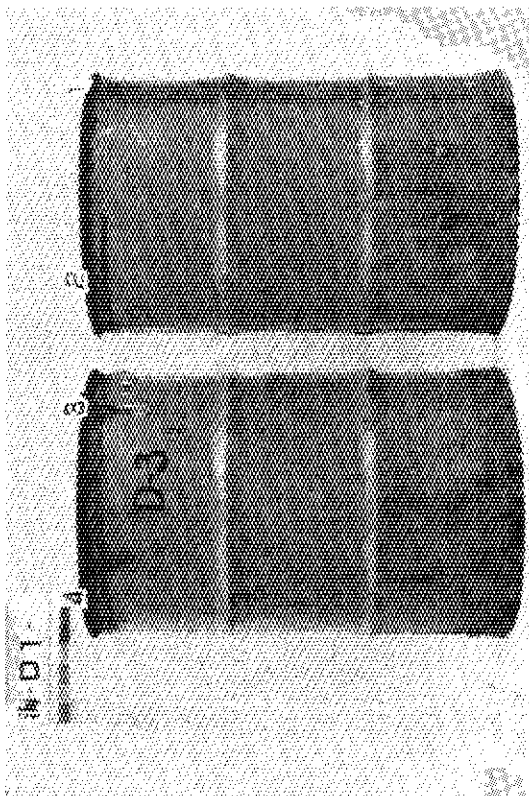
Although seawater seeped into the No. 1 test body (without coating, $t \dots 1.2$ mm), no rust was found on the shell and bottom plate which were in contact with the simulated cemented waste, and accordingly sound. A small amount of rust was observed on the upper part of the shell and top plate both of which were not in contact with the cemented waste. With regard to the seawater that had seeped into the drum container, the amount of dissolved oxygen decreased, and its pH value became higher because the alkaline components leached out of the cemented waste, resulting in a decrease⁸⁾ in corrosivity of the steel plate. No seepage of seawater was observed for the No. 2 test body (without coating, $t \dots 1.6$ mm). Although thin rust layers were observed on the top plate and part of the shell which were not in contact with the simulated cement waste, the bottom plate and part of the shell which were in contact with it were sound.

No seawater seepage was found for the No. 3, 4, 5 and 6 test bodies (internal surface \dots epoxy-resin coating). Although a small amount of blisters and rust were observed on the top plate and uppermost part of the shell which were not in contact with the cemented waste, the shell and bottom plate which were in contact with it were sound.

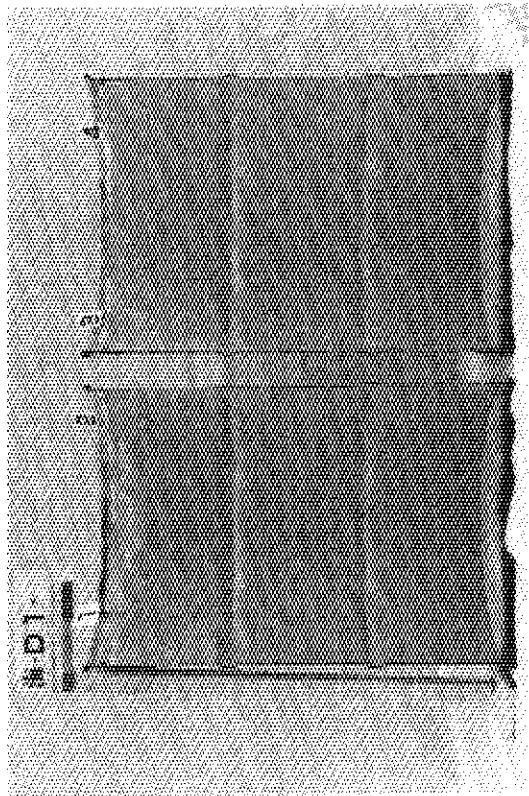
Regarding the No. 8 test body (internal surface \dots galvanization & melamine coating), seawater did not seep into the drum container, but the generation of blisters and rust on the top plate and the uppermost part of the shell were greater than that in the other-type of drum containers.

In two No. 9 test bodies (stainless steel), remarkable gap corrosion⁹⁾ was observed in the top plates and the uppermost part of the shells both of which were in contact with rubber packing, and formed through

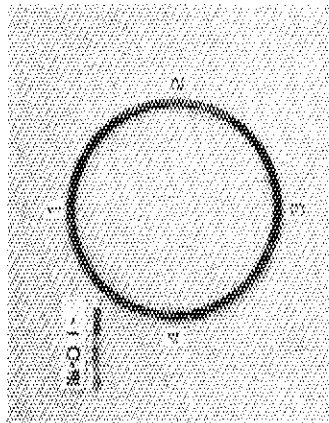
the holes. Although seawater flowed into the drum container through the holes, the internal surface of the test body was not effect except for the above-mentioned gap corrosion, and was considered to be sound.



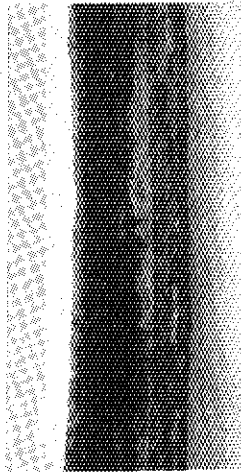
External surface of shell



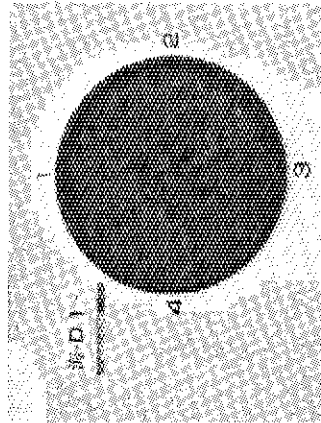
Internal surface of shell



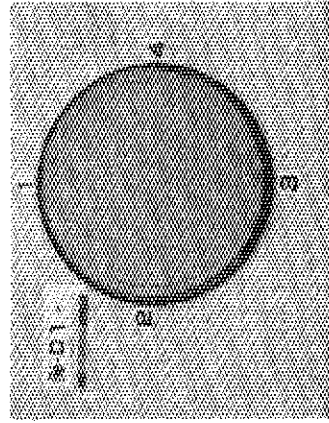
External surface of band



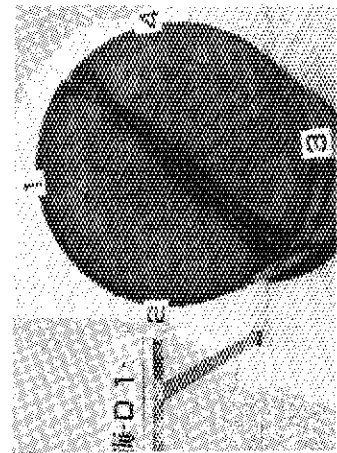
Internal surface of band



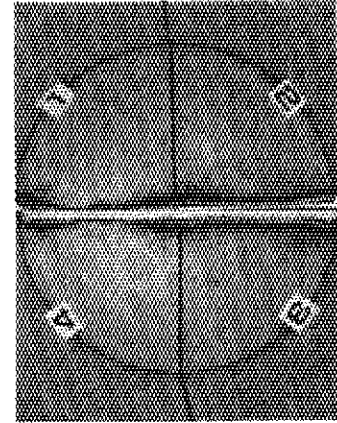
External surface of top plate



Internal surface of top plate



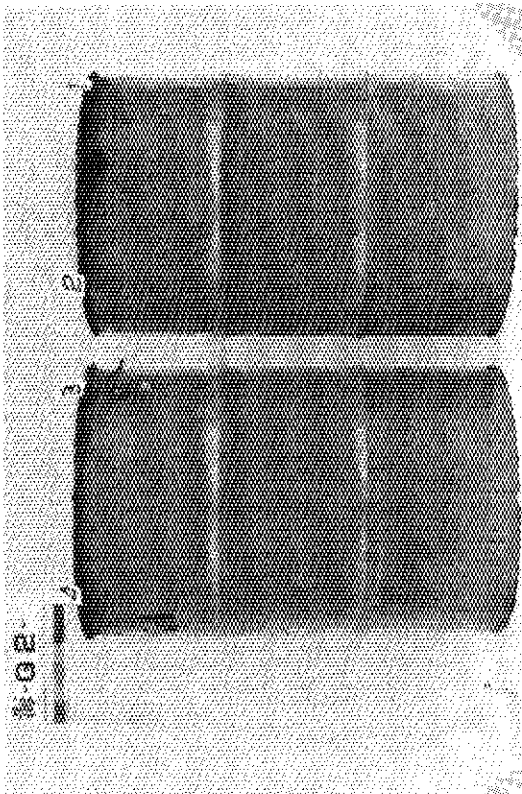
External surface of bottom plate



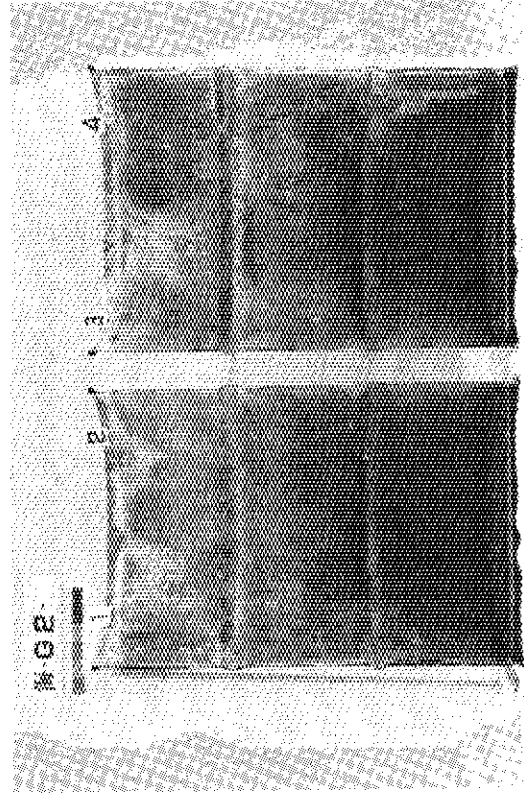
Internal surface of bottom plate

No. 4 drum; Melamine-coating (1.6 mm.)

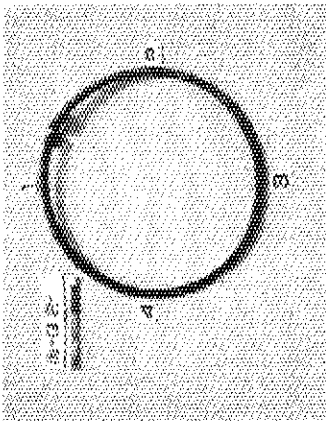
Photograph 4.1 Drum after the seawater corrosion tests (55 months).



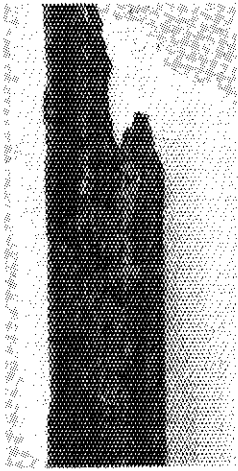
External surface of shell



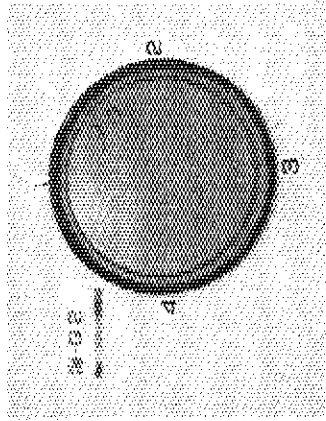
Internal surface of shell



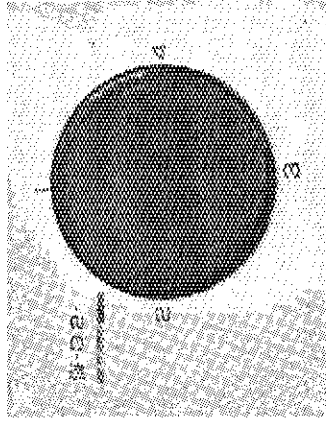
External surface of band



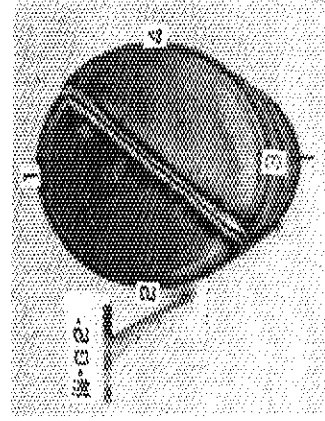
Internal surface of band



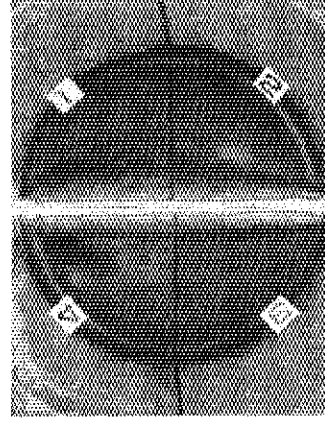
External surface of top plate



Internal surface of top plate



External surface of bottom plate



Internal surface of top plate

No.5 drum; Zinc-bearing primer and epoxy-coating (1.6 mm.)

Photograph 4.2 Drum after the seawater corrosion tests (55 months).

4.1.5 Coating Deterioration Tests

4.1.5.1 Testing Methods

(1) External Surfaces

Various parts of the coated container were tested once a year to once in 6 months during an immersion test and after the immersion test; the parts tested were the external surfaces of the top plate, shell and bottom plate (excluding the bottom plate for the tests during immersion), with the shell divided further into the upper, middle and lower parts (see Fig. 4-7). As for these parts, the areas of the coating in which blisters and / or rust generate were measured in terms of percentage to evaluate the change in the coating. With regard to No. 6 test body (electrical anticorrosion treatment & melamine coating), the sacrificial anodic reaction due to anticorrosive zinc as well as melamine coatings protected the stainless steel itself, and zinc consumption was obtained by a weight measurement at the end of the test. For the No. 8 test body (galvanization & melamine coating), the consumption of the galvanized layer beneath the melamine coating was obtained by measuring the thickness of the layer.

(2) Internal Surfaces

The deterioration of the internal coating of the drum container depends on contact with cemented waste as well as the seawater seepage. To evaluate the deterioration of the coatings by calculating the percentage of the coated area in which blisters and / or rust generated, part of the drum container which was not in contact with the cemented waste was divided into the top plate and uppermost zone of the shell, whereas part of the drum container which was in contact with the cemented waste in the bottom plate and the shell except for its uppermost zone. In the case of the No. 8 test body (galvanization & melamine coating),

the consumption of zinc was calculated by measuring the thickness of the galvanized layer beneath the coating.

4.1.5.2 Results

Tables 4-3 and 4-4 give the results of the coating deterioration tests at the end of the immersion tests, while Fig. 4-8 shows the secular change in the deterioration of the coatings.

The consumption of anticorrosive zinc of the No. 6 test body (electrical anticorrosion treatment & melamine coating) was 1.005 g (initial amount before test ... 6.300 g) after a 55-month test. As for the galvanized layer (25 μ before test) of the No. 8 test body (galvanization & melamine coating) no change in the amount was found in the internal surface between before and after testing, indicating no zinc consumption. Regarding the external surface, the galvanized layer was almost lost, indicating zinc exhaustion in the anearly stage.

(Uppermost part of shell) part which was
not in contact with cemented waste

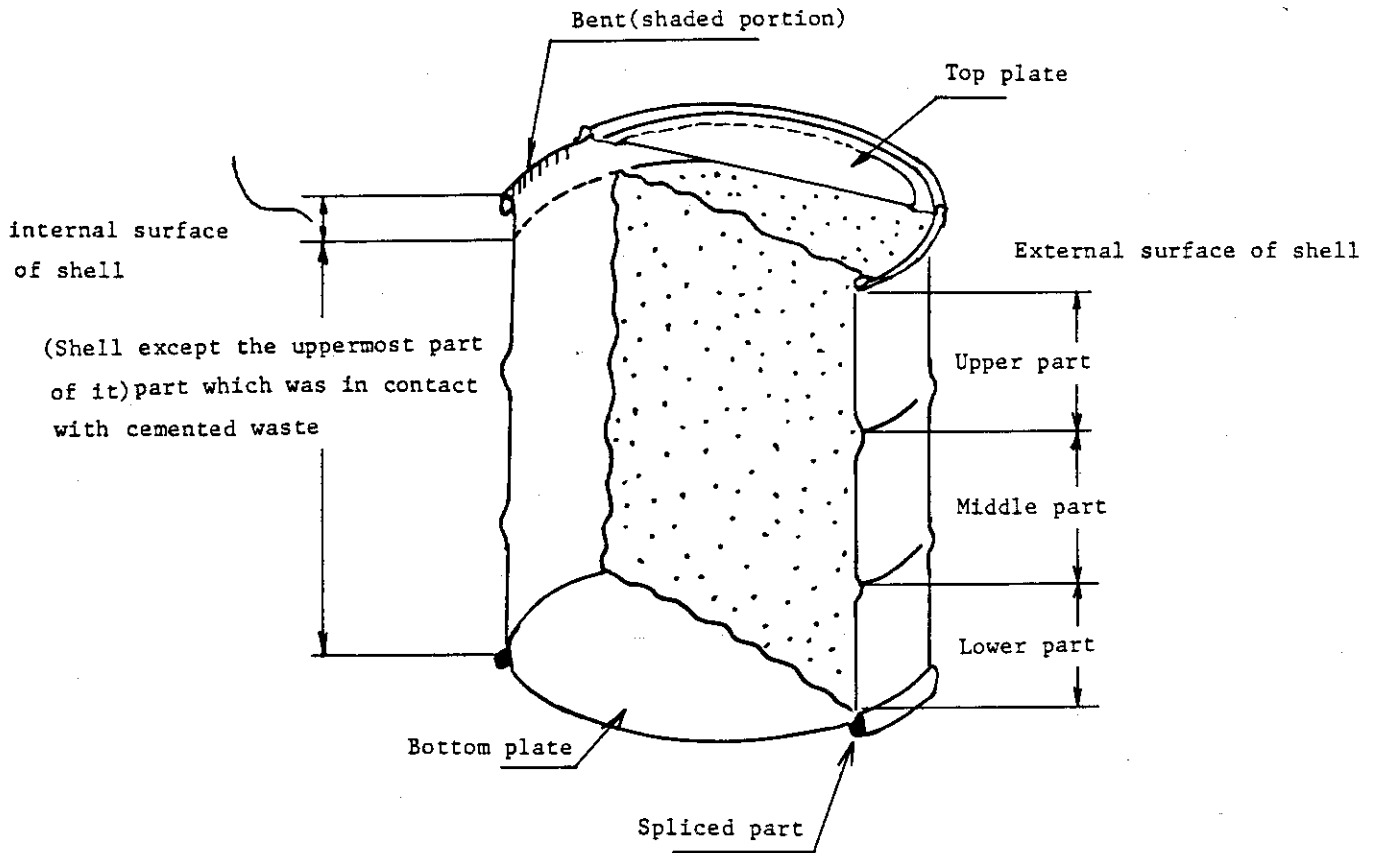


Fig. 4-7 Classification of external and internal surfaces of a drum shell in the coating deterioration tests.

Table 4-3-(1) Deterioration of external-surface coatings of drums after the seawater corrosion tests.

Test drum No.	Classification of anticorrosion treatment	Steel plate thickness (mm)	Testing period	Test part	Blister ¹⁾ generated area	Rust ²⁾ generated area	Remarks
3	Melamine coating	1.2	3 years	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	10 3 8 8 15	80 5 7 25 15	
3	Melamine coating	1.2	55 months	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	25 10 35 35 10	70 10 10 25 30	Cracks in the Shell
4	Melamine coating	1.6	3 years	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	10 2 5 10 10	50 5 5 12 10	
4	Melamine coating	1.6	55 months	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	30 10 40 60 20	70 10 10 35 30	
5	Zinc-bearing anticorrosion primer and epoxy coating	1.6	4 years	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	1 3 3 3 1	1 2 0.5 1 1	
5	Zinc-bearing anticorrosion primer and epoxy coating	1.6	55 months	Top plate Upper part of shell Middle part of shell Lower part of shell Bottom plate	5 10 10 10 2	2 5 0 1 0	

- Notes: 1) The "blister" mentioned here means one beneath which no rust is generated and still having a steel-plate-protecting function.
2) The aprt beneath coated blisters in which rust is generated over the steel plate is included in the rust-generated zone.

Table 4-3-(2) Detevioration of external-surface coatings of drums after the seawater corrosion tests.

Test drum No.	Classification of anticorrosion treatment	Steel plate thickness (mm)	Testing period	Test part	Blister ¹⁾ generated area	Rust ²⁾ generated area	Remarks
6	Electvical anticorrosion treatwent and melamine coating	1.6	55 months	Top plate	90	0	This "blister" includes spallings.
				Upper part of shell	50	0	
				Middle part of shell	45	0	
				Lower part of shell	95	0	
				Bottom plate	50	0	
6	Electvical anticorrosion treatment and melamine coating	1.6	55 months	Top plate	95	0	This "blister" includes sapplings.
				Upper part of shell	95	0	
				Middle part of shell	90	0	
				Lower part of shell	95	0	
				Bottom plate	70	0	
8	Galvanization and melamine coating	1.6	55 months	Top plate	5	95	This "blister" includes spallings.
				Upper part of shell	40	10	
				Middle part of shell	40	2	
				Lower part of shell	55	4	
				Bottom plate	20	1	
8	Galvanization and melamine coating	1.6	55 months	Top plate	40	60	This "blister" includes spallings.
				Upper part of shell	55	5	
				Middle part of shell	63	1	
				Lower part of shell	60	1	
				Bottom plate	90	0	

- Notes: 1) The "blister" mentioned here means one beneath which no rust is generated and still having a steel-plate-protecting function.
 2) The part beneath coated blisters in which rust is generated over the steep plate is included in the rust-generated zone.

Table 4-4-(1) Deterioration of internal-surface coatings of drums after the seawater corrosion tests.

Test drum No.	Classification of anticorrosion treatment	Steel plate thickness (mm)	Testing period	Seawater seepage	Test part	Blister- ¹⁾ generated area	Rust- ²⁾ generated area	Remarks
3	Epoxy coating	1.2	3 years	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	0 0 0 0	
3	Epoxy coating	1.2	55 months	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	5 2 0 0	Cracks in the shell
4	Epoxy coating	1.6	3 years	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	0 0 0 0	
4	Epoxy coating	1.6	55 months	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	5 0 0 0	
5	Zinc-bearing anticorrosion primer and epoxy coating	1.6	4 years	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	0 5 0 0	
5	Zinc-bearing anticorrosion primer and epoxy coating	1.6	55 months	Without	Top plate Upper part of shell Shell Bottom plate	10 0 0 0	1 1 0 0	
6	Zinc-bearing anticorrosion primer and epoxy coating	1.6	52 months	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	0 2 0 0	Electrically anticorrosion-treated external surface

- Notes: 1) The "blister" mentioned here means one beneath which no rust is generated and still having a steel-plate protecting function.
2) The part beneath coated blisters in which rust is generated over the steel plate is included in the rust-generated zone.

Table 4-4-(2) Deterioration of internal-surface coatings of drums after the seawater corrosion tests.

Test drum No.	Classification of anticorrosion treatment	Steel plate thickness (mm)	Testing period	Seawater seepage	Test part	Blister-generated area	Rust-generated area	Remarks
6	Epoxy coating	1.6	55 months	Without	Top plate Upper part of shell Shell Bottom plate	0 0 0 0	0 0 0 0	Electrically anticorrosion-treated external surface
8	Galvanization and welawive coating	1.6	52 months	Without	Top plate Upper part of shell Shell Bottom plate	2 0 (40)* (40)*	5 5 0 0	Coated spalling due to adhesion to the cemenled body
8	Galvanization and welawive coating	1.6	55 months	Without	Top plate Upper part of shell Shell Bottom plate	2 10 (40)* (40)*	2 5 0 0	Coated spalling due to adhesion to the cemenled body

- Notes: 1) The "blister" mentioned here means one beneath which no rust is generated and still having a steel-plate protecting function.
- 2) The part beneath coated blisters in which rust is generated over the steel plate is included in the rust-generated zone.

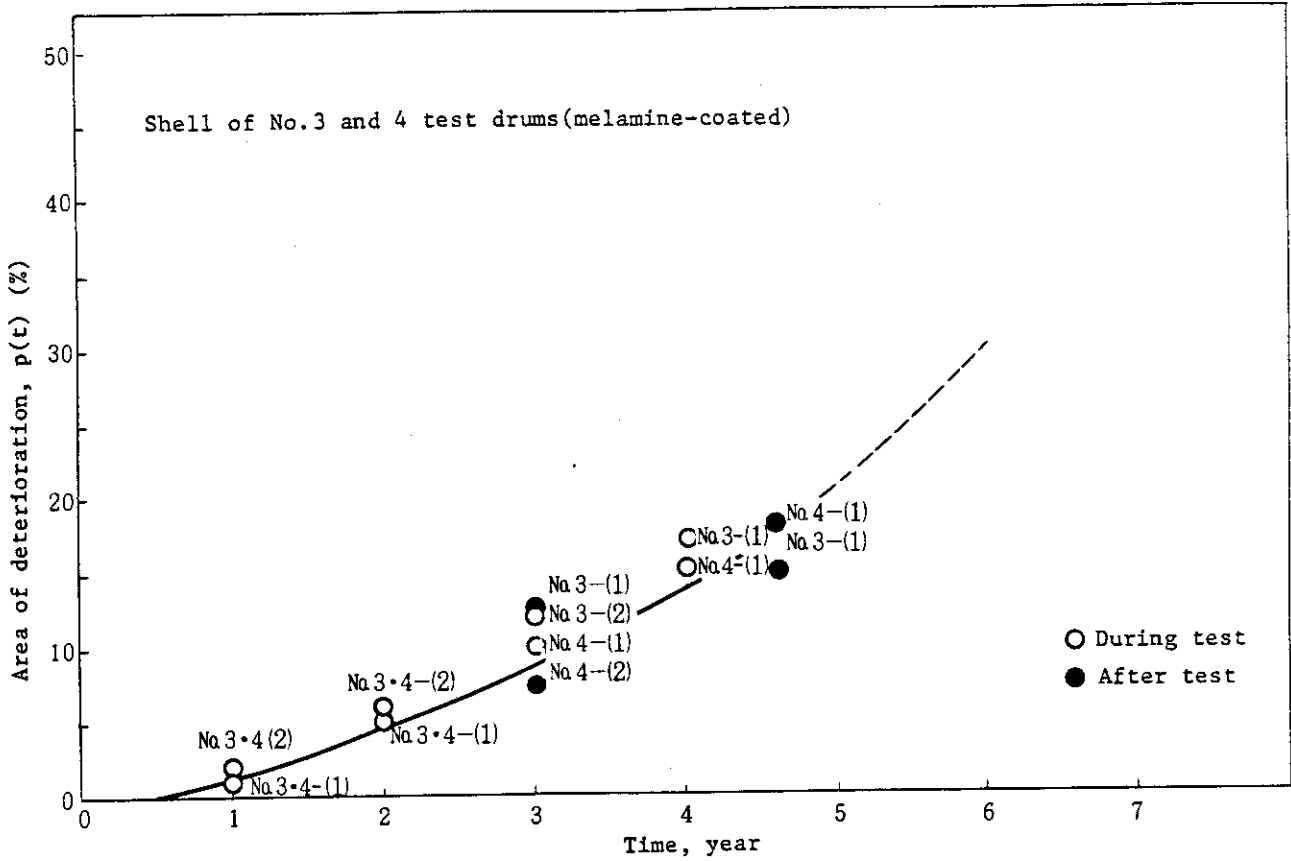


Fig. 4-8-(1) Deteriorated area of external surface coating of drums in seawater.

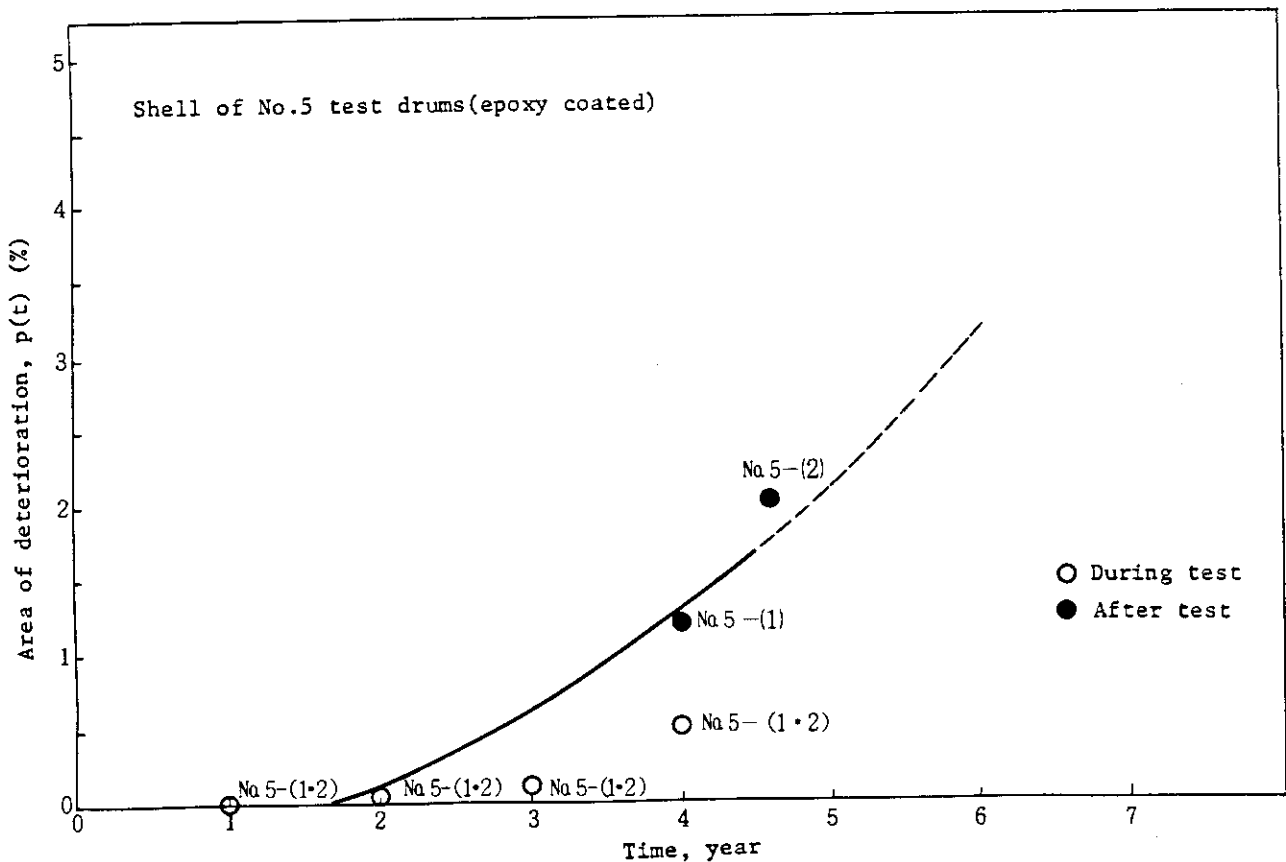


Fig. 4-8-(2) Deteriorated area of external surface coating of drums in seawater.

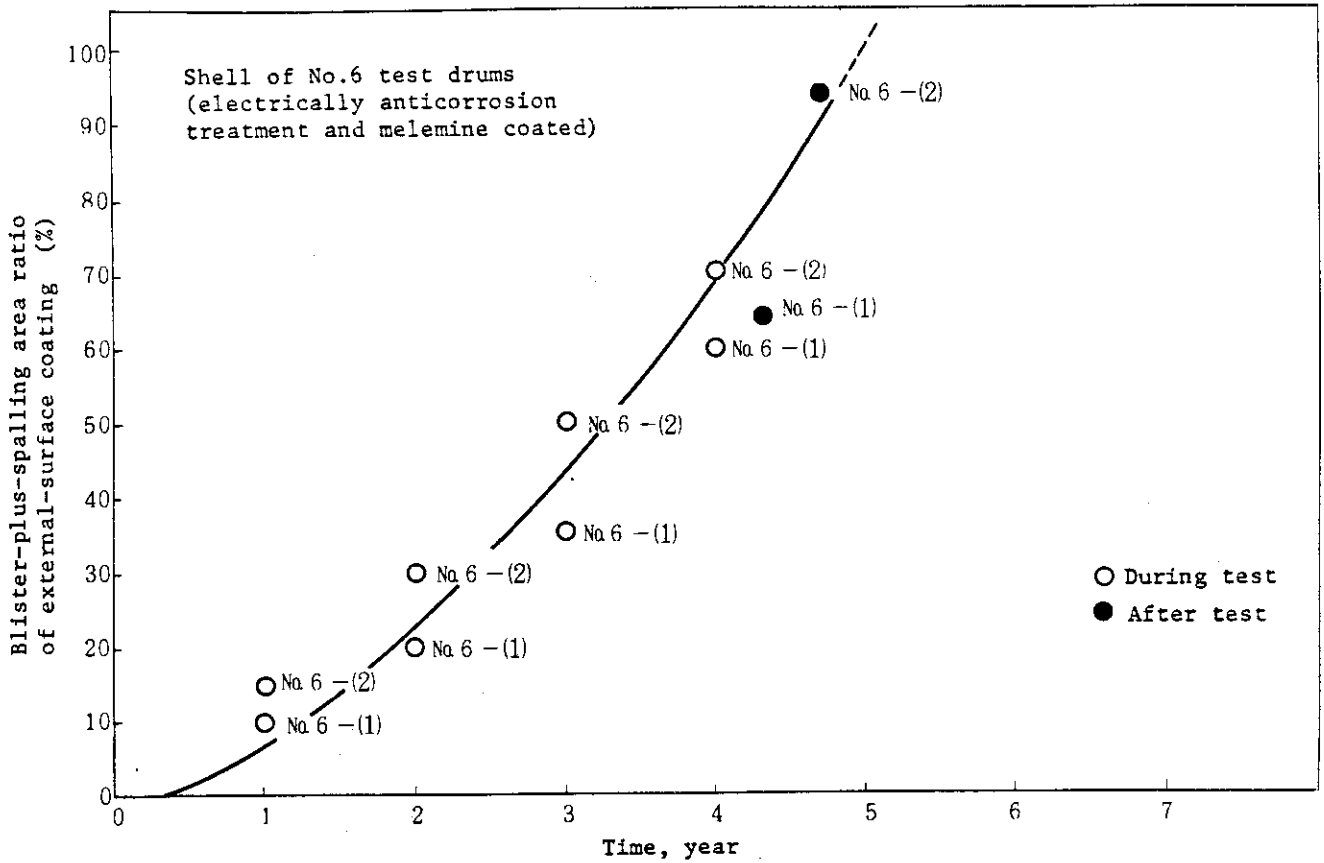


Fig. 4-8-(3) Deteriorated area of external surface coating of drums in seawater.

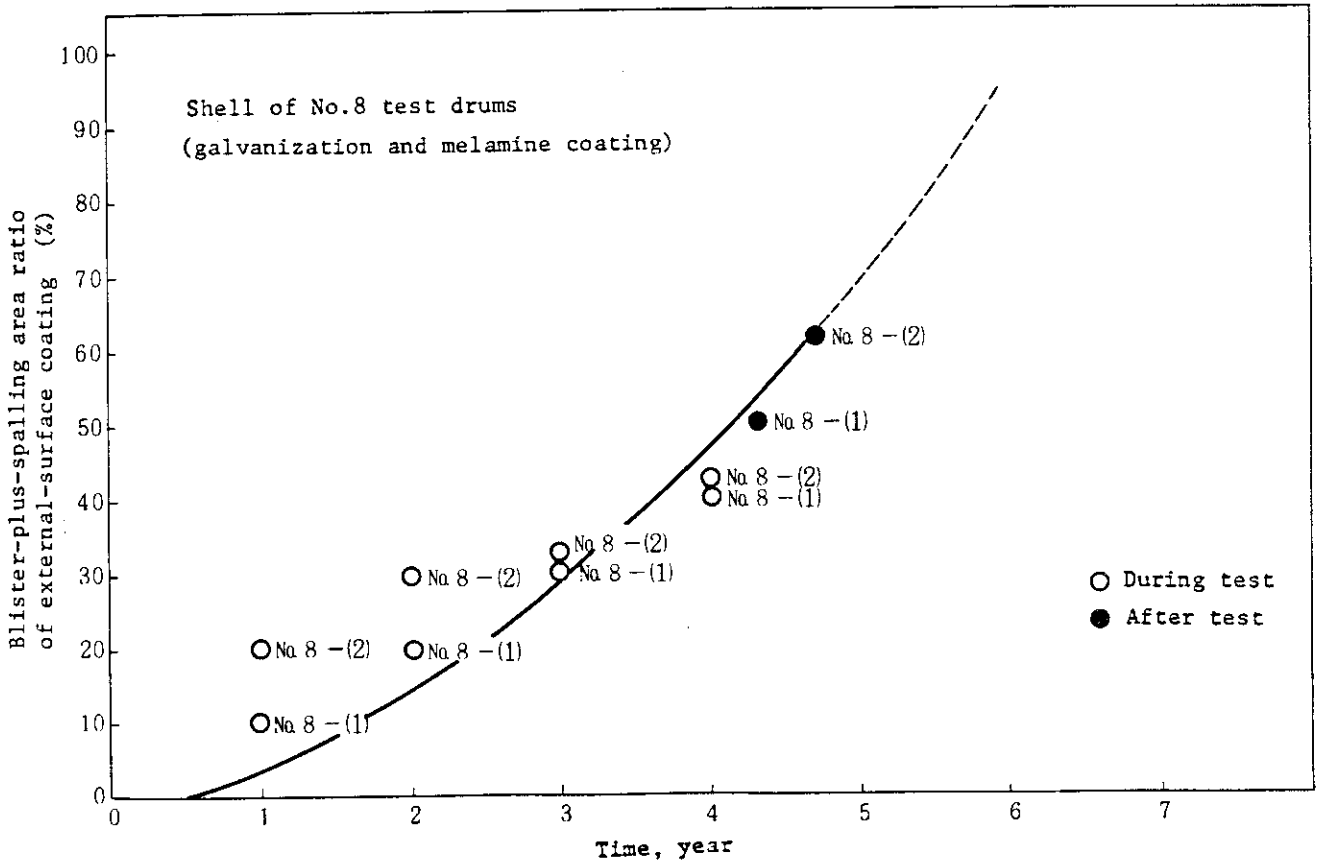


Fig. 4-8-(4) Deteriorated area of external surface coating of drums in seawater.

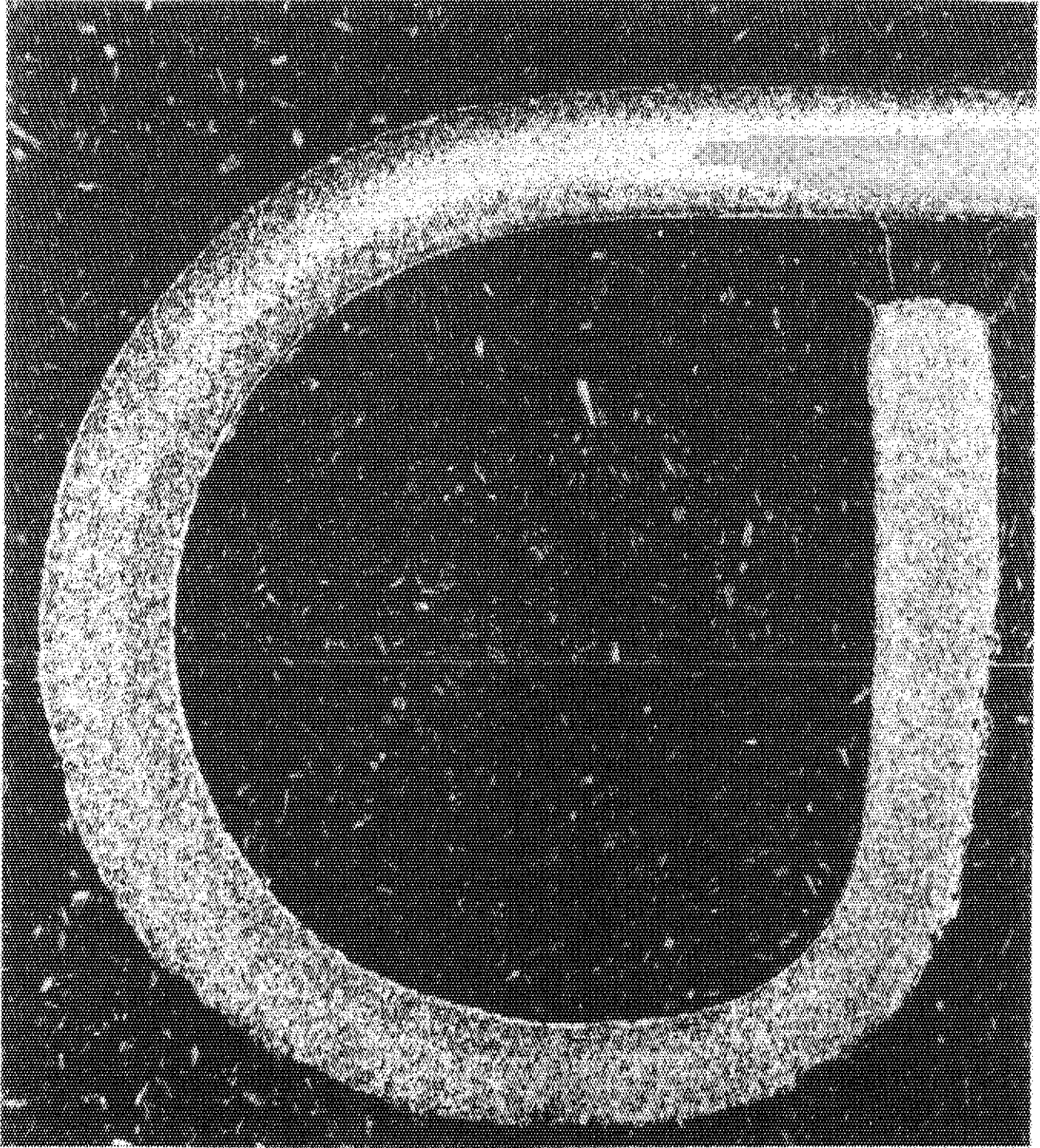
4.1.6 Microscopic Examination

4.1.6.1 Examination Methods

After finishing external appearance examinations and coating deterioration tests, the highly worked parts of the drum containers such as their bents, welds, ring-band parts, spliced parts, peripheral parts of the top plates and bands were sampled, followed by microphotographs of their cross sections to observe the state of corrosion.

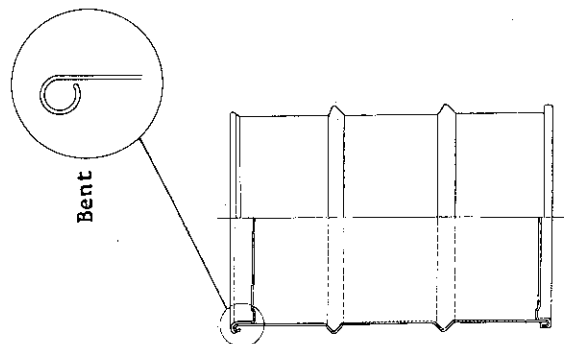
4.1.6.2 Results

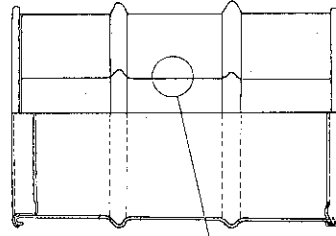
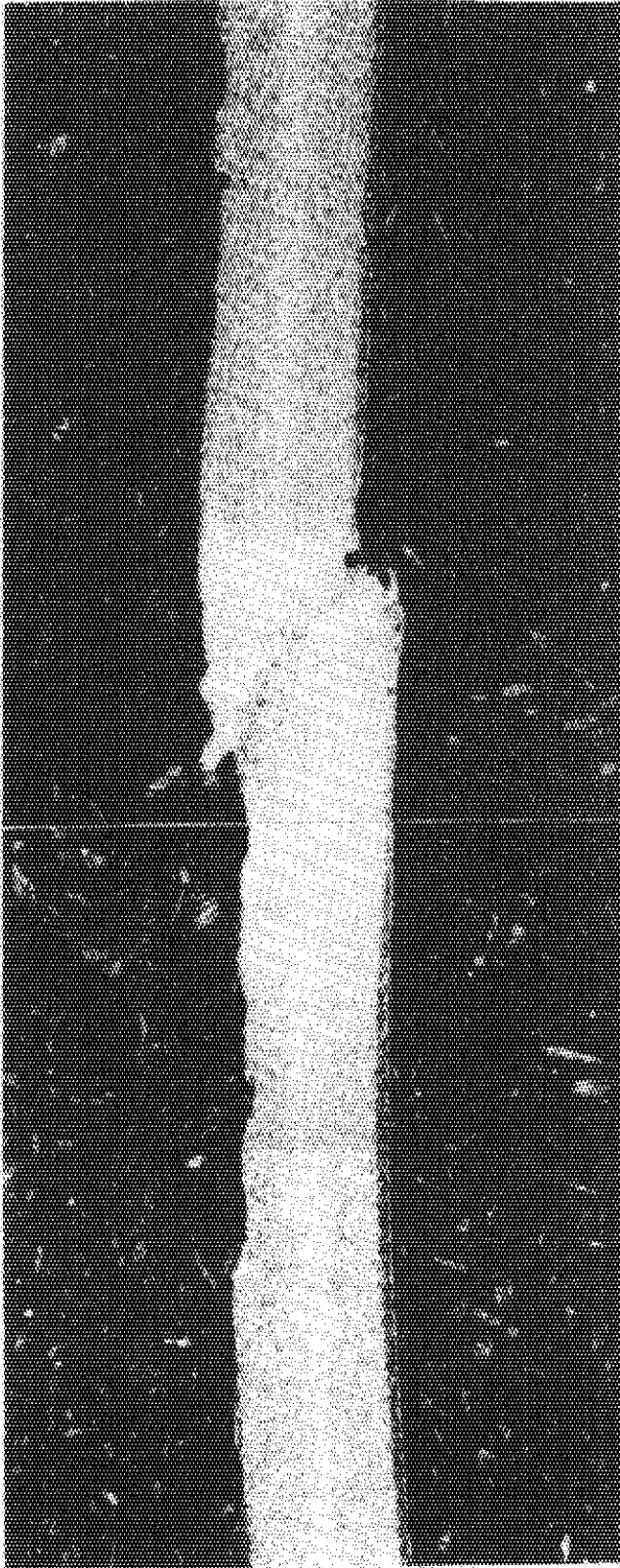
The results on the No. 2 (without coating, $t \approx 1.6$ mm) test bodies are shown in Photo. 4-3 and no significant differences were observed in the state of corrosion in the highly worked zones of the steel plates. The results on the bent of the No. 9 test body (stainless steel) are given in Photo. 4-4, showing that about 6.5-mm-dia. iron core wire was completely dissolved and corrosion took place in the vicinity. In addition remarkable gap corrosion was found along the contacting part between the top plate and the shell.



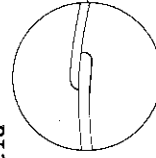
Bent of No.2 test drum(without coating, t-----1.6mm)

Photograph 4-3-(1) Enlarged cross-sectional photograph of each part of a drum after a 55 month seawater corrosion test.



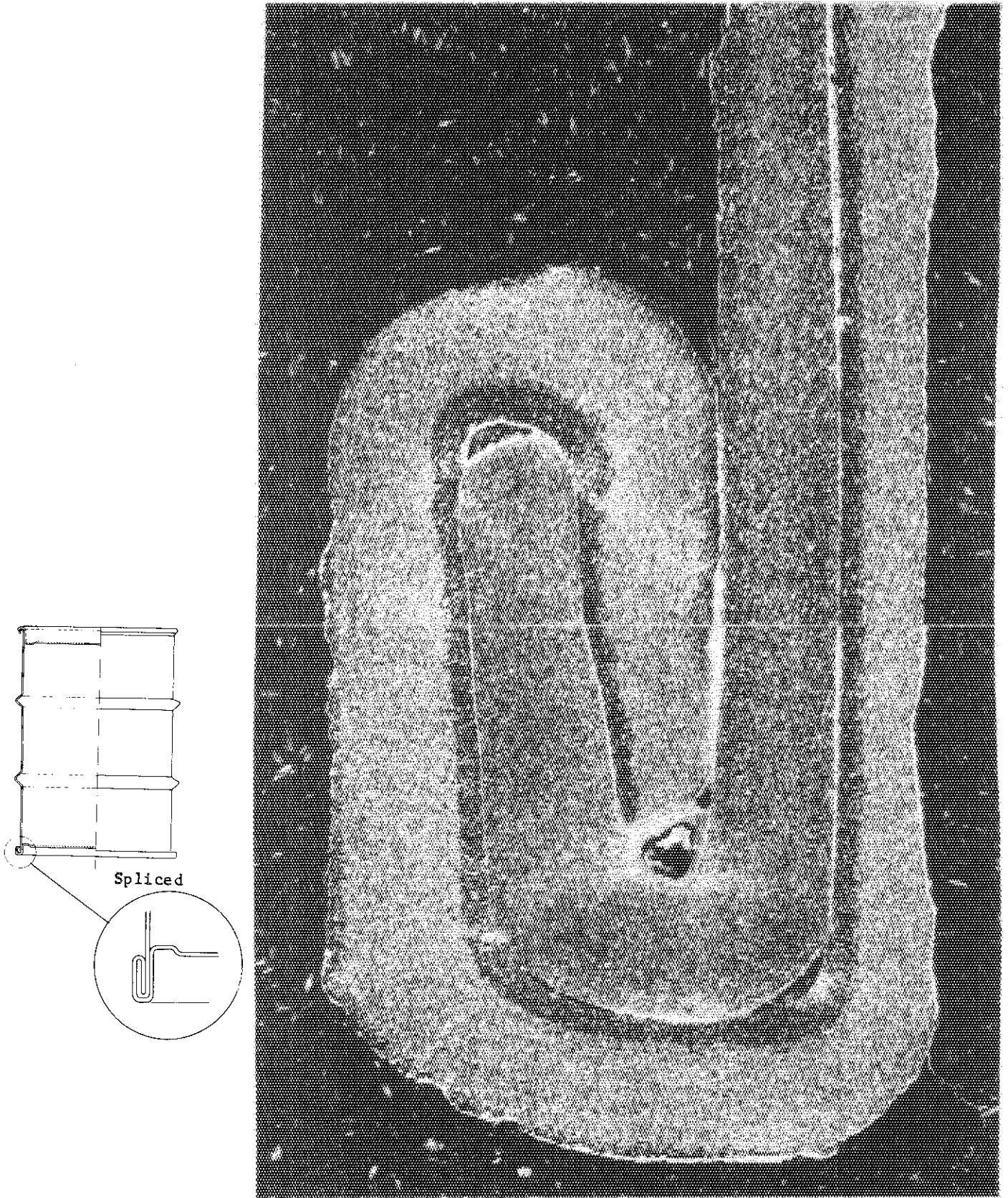


Weld



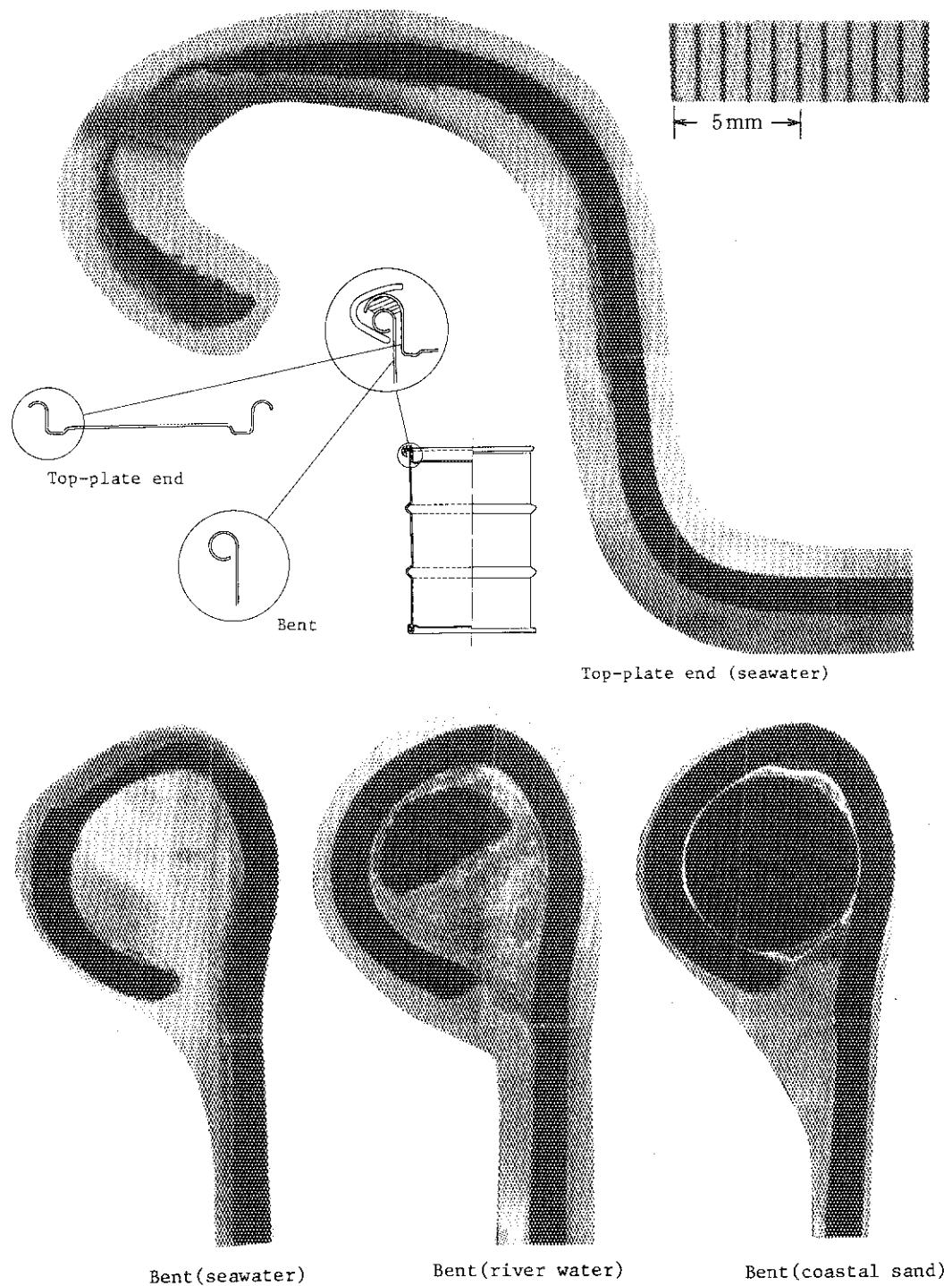
Weld of No.2 test drum(without coating, t-----1.6mm)

Photograph 4-3-(2) Enlarged cross-sectional photograph of each part of a drum after a 55 month seawater corrosion test.



Spliced part of No.2 test drum (without coating, $t \approx 1.6\text{mm}$)

Photograph 4-3-(3) Enlarged cross-sectional photograph of each part of a drum after a 55 month seawater corrosion test.



No.9 drum; Stainless steel (1.5mmt.)

Photograph 4.4 Enlarged cross-sectional photographs of bents and top-plate-to-packing-contacting part of drums after seawater, river water and coastal sand corrosion tests (55 months).

4.1.7 Corrosion Tests for Steel plates

After immersion tests, non-coated drum containers were cut longitudinally, and simulated cemented waste was removed, followed by rust removal by immersing in a 10- % ammonium citrate solution and corrosion tests for steel plates described below.

4.1.7.1 Testing Methods

(1) Average Corrosion Rates

The test pieces shown in Fig. 4-9 were prepared from the rust-removed No. 2 test body (without coating, $t \approx 1.6$ mm), their areas and weights were measured to calculate the average plate thickness, and the average corrosion rate (mm/y) of the steel plate was obtained by comparing them with the thickness before testing.

(2) Local Corrosion Rates

As for the parts of the rust-removed No. 2 test body (without coating, $t \approx 1.6$ mm) shown in Fig. 4-9, the thicknesses of severely corroded portions were measured, the average value of the three thickest was calculated, and the local corrosion rate (mm/y) was obtained by comparing them with the plate thickness before testing. For the measurement of plate thicknesses, a CPM-15 point micrometer fabricated by Sanpo Seisakujo Co., Ltd. was used.

(3) Fixed Point Thickness Measurement

For the purpose of confirming the uneven corrosion over the external and internal surfaces of the drum container, the measuring points were determined for the top plate, shell and bottom plate of the No. 2 test body (without coating, $t \approx 1.6$ mm), and the thickness distribution was calculated. A CPM 15-25 point micrometer was used for the measurement.

4.1.7.2 Results

(1) Average Corrosion Rates

Table 4-5 shows the average plate thicknesses (mm), average changes in plate thicknesses (mm) and average corrosion rates (mm/y) calculated from the measured weights.

(2) Local Corrosion Rates

The plate thicknesses of locally severely affected portions were measured and the local corrosion rates (mm/y), which are shown in Table 4-6, were calculated.

(3) Fixed Point Thickness Measurement

Figure 4-10 shows the results of thickness measurement for the top plate, shell and bottom plate of the No. 2 test body (without coating, $t \approx 1.6$ mm), while Fig. 4-11 the resultant histogram.

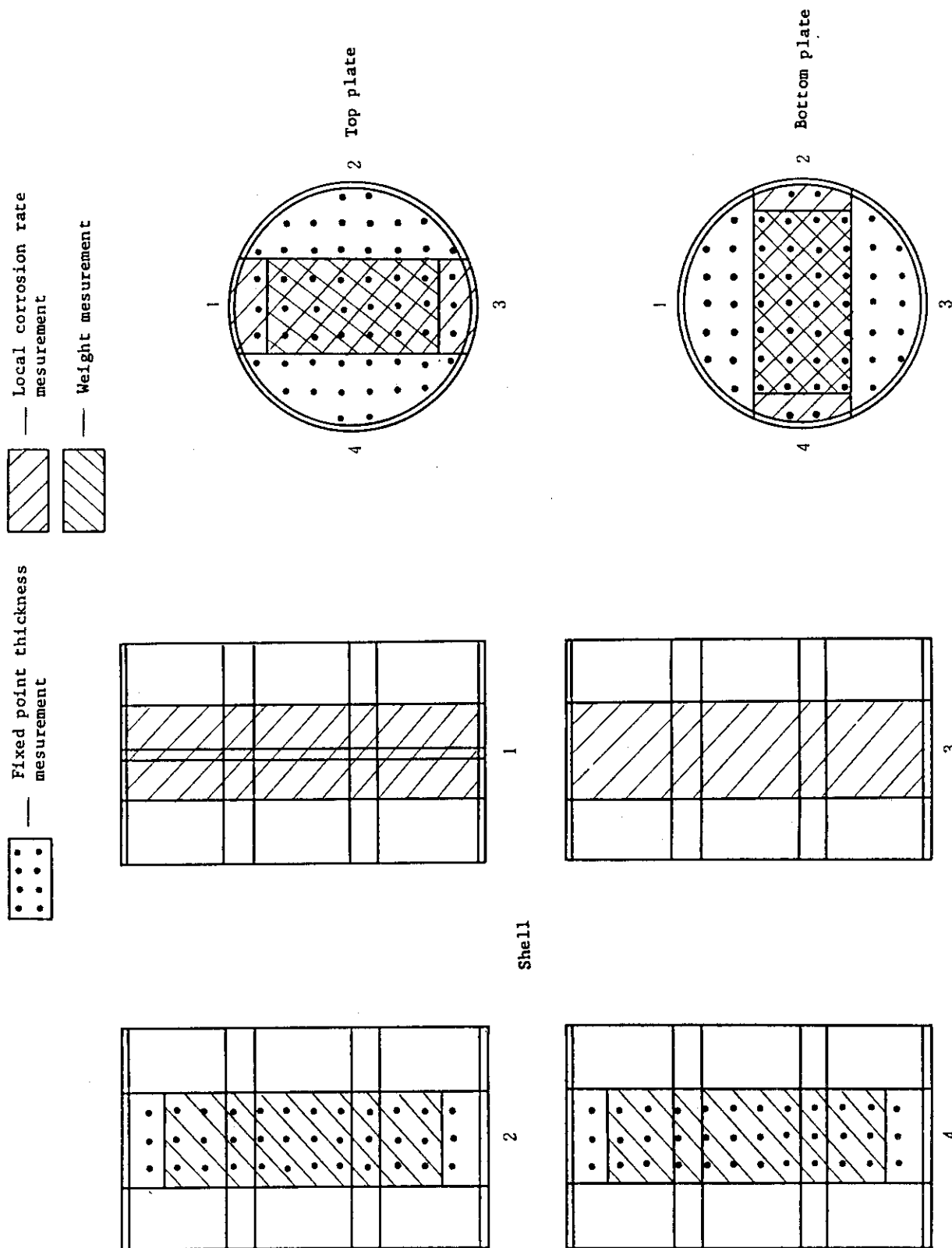


Fig. 4-9 Testing positions of a non-coated drum in steel-plate corrosion tests.

Table 4-5 Average plate thickness and corrosion rates after the seawater corrosion tests obtained from the weight measurement.

No.1 test drum (t ... 1.6 mm), immersion period ... 55 month

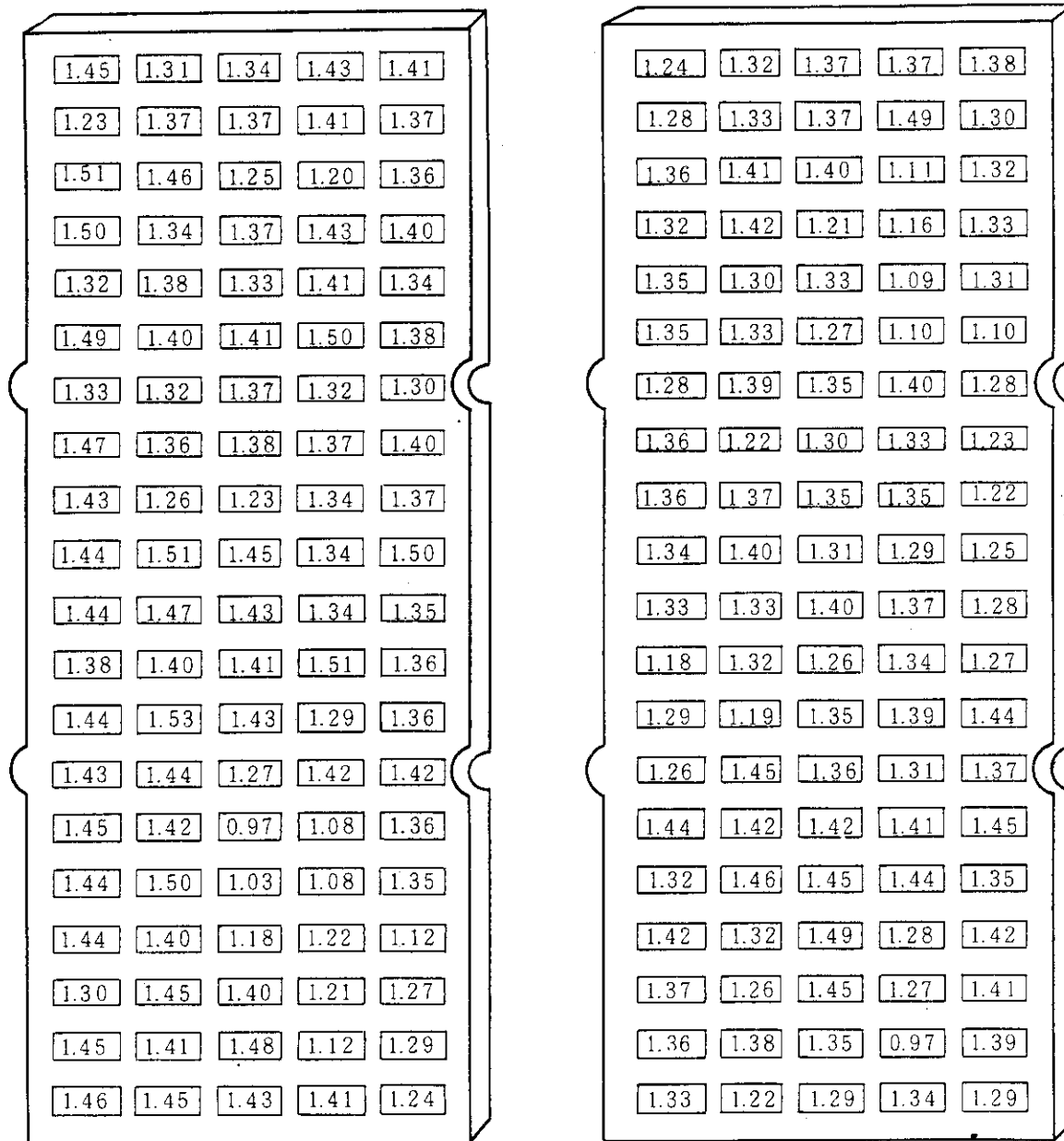
Test object	Weight (g)	Area (cm ²)	Average plate thickness (mm)	Plate thickness before testing (mm)	Thickness change (mm)	Average corrosion rate (mm/Y)
Top plate	769.44	912	1.073	1.190	0.117	0.026
Shell	3506.91	3334	1.340	1.580	0.240	0.052
Bottom plate	1057.36	935	1.439	1.590	0.151	0.033

Table 4-6 Local corrosion rates after the seawater corrosion tests.

No.2 test drum (t ... 1.6 mm), immersion period ... months

Test object	Plate thickness before testing (mm)	Minimum * thickness after testing (mm)	Maximum thickness change (mm)	Local corrosion rate (mm/Y)
Top plate	1.190	0.630	0.560	0.12
Shell	1.580	0.713	0.867	0.19
Bottom plate	1.590	0.837	0.753	0.16

* The minimum plate thickness means the average value of three measured minimum thicknesses.



2

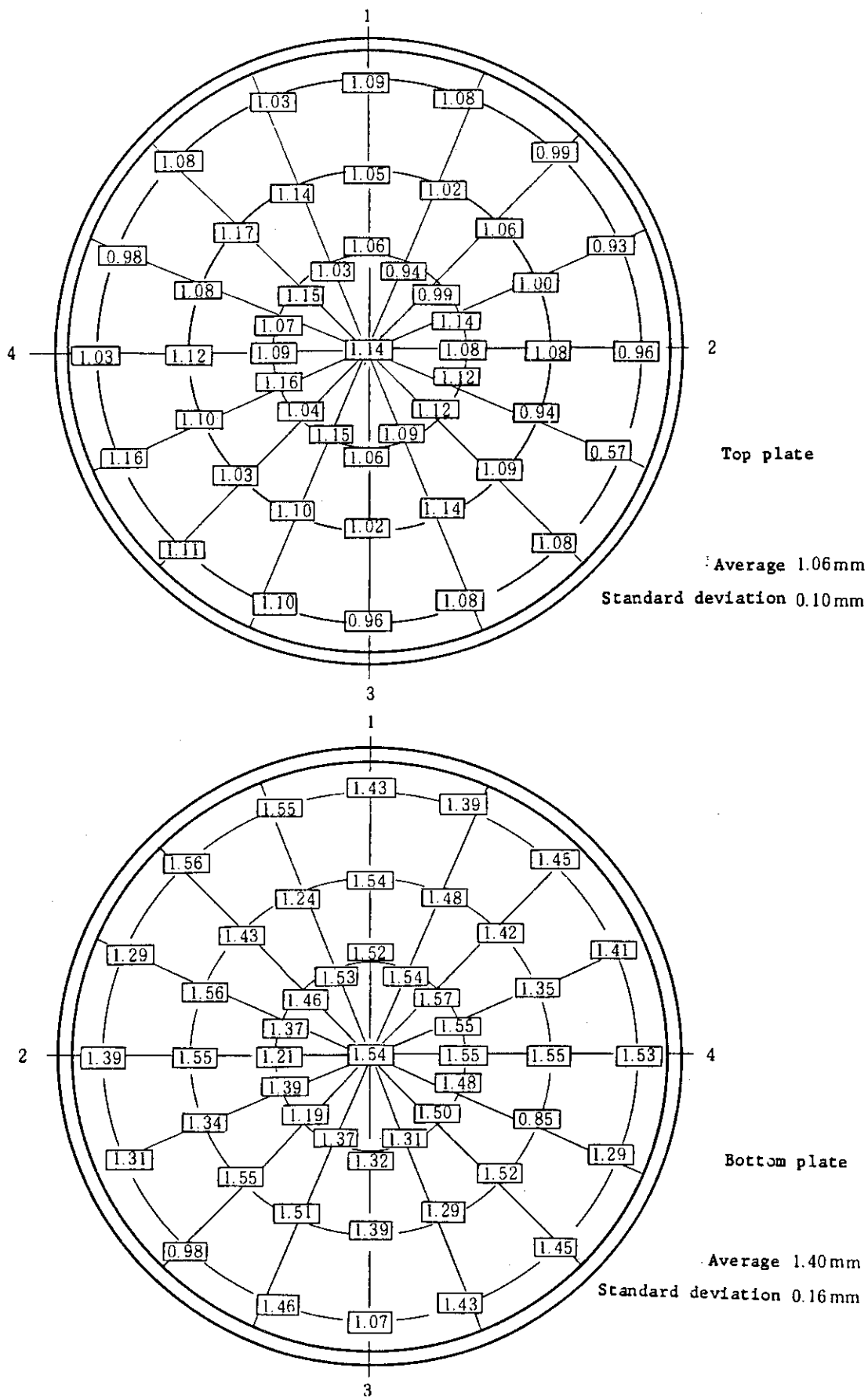
Shell

4

Average 1.35 mm
Standard deviation 0.10 mm

No.2 test drum(without coating, t-----1.6mm)

Fig. 4-10-(1) Fixed-point thickness distribution of drums after the 55-months seawater corrosion tests.



No.2 test drum (without coating, t-----1.6mm)

Fig. 4-10-(2) Fixed-point thickness distribution of drums after the 55-months seawater corrosion tests.

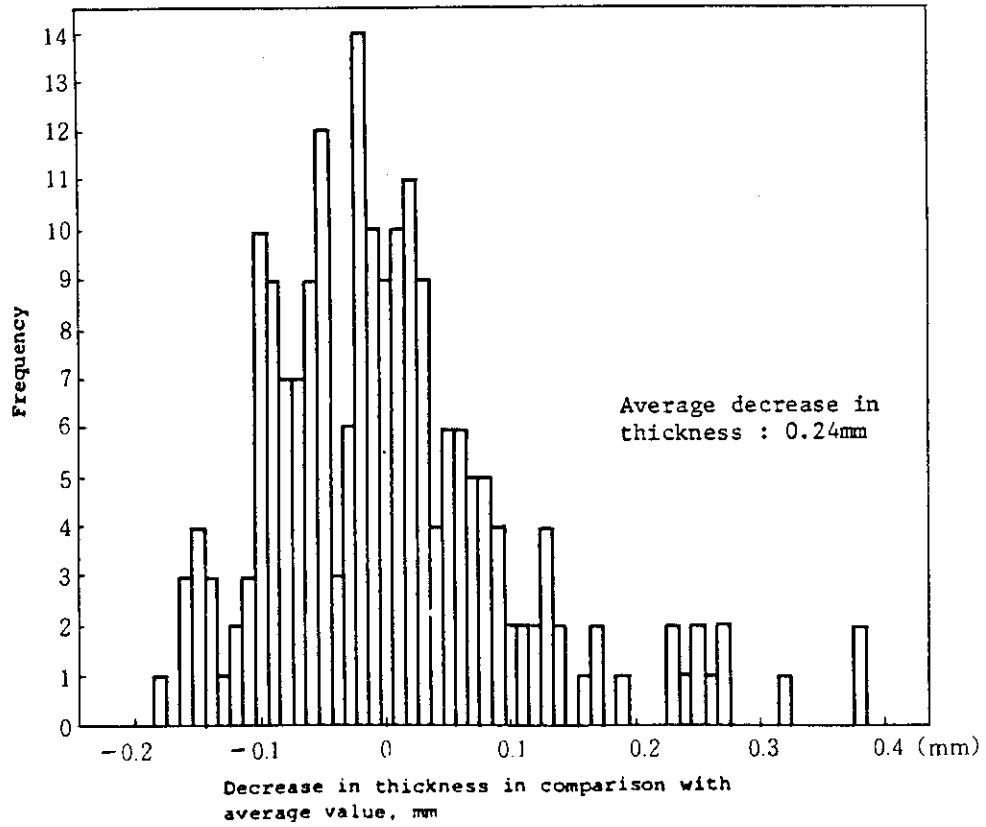


Fig. 4-11 Distribution of decreasing thickness of drum side due to corrosion (An uncoated drum was immersed in seawater at 23°C for 55 months).

4.2 Test Pieces

4.2.1 Testing Methods

The immersion tests under the same environmental conditions as those for the drum containers were carried out in a seawater testing tank; namely, in the each compartment (see Fig. 4-1) where a drum container was immersed a few test pieces with the same kind coating as that of the drum container were suspended.

The immersion tests in low-temperature seawater, on the other hand, were performed in a small testing tank. A 310-mm long, 150-mm wide, 195-mm high acrylic-resin water tank was set in a thermostatic tank, in which the water temperature was kept constant and fresh air was supplied by 5-mm-dia. vinyl tubes to keep the amount of dissolved oxygen constant. The seawater for testing was the same as that in the testing tank in which the drum containers were immersed, and was exchanged once a week.

4.2.2 Environmental Conditions

The various environmental conditions of the seawater in the small testing tank were measured before and after the seawater exchange, with the results shown in Table 4-7. Figures 4-12 and 4-13 show seawater temperatures, amounts of dissolved oxygen, acidities and electric conductivities in terms of secular change. Regarding the temperature, it was set at 5 °C at first, but the average of measured values through the whole testing period was 6.7 °C. The conditions for the corrosion tests for the test pieces suspended in the seawater testing tank were already shown (See 4.1.2).

4.2.3 External Appearance Examination

(1) Examination Methods

The test pieces immersed in the seawater were taken out of the tank

at given intervals, rinsed lightly with the seawater to remove any adhered substances, and then the deterioration of surface coatings together with the state of rust generation were visually observed and photographed.

(2) Results

Tables 4-8 and 4-9 give the results of the visual observation for the seawater immersion tests, but the tests in the small testing tank for the following test pieces were suspended because of the extensive generation of coated blisters and spallings; they are the No. 48 test piece (melamine coating, $t \approx 1.2$ mm), No. 5 (melamine coating, $t \approx 1.6$ mm), No. 56 (electric anticorrosion treatment & melamine coating) and No. 43 (galvanization & melamine coating).

Compared with damage-free test pieces, a great deal of blisters and rust was generated in crosscut test pieces, indicating the influence of the damage to the coatings. With regard to electrically anticorrosion-treated test pieces, no rust was observed in the damage-free and the crosscut test pieces, showing no influence due to any damage to the coatings.

As to galvanized, melamine-coated test pieces, no rust was generated in the crosscut test pieces, showing no influence due to any damage to the coatings, so long as the zinc coating exhibits sacrificial anodic reaction. As a whole, no significant difference was found in test-piece coating deterioration between the results from the small testing tank (average measured temperature ≈ 6.7 °C) and those from the seawater testing tank (average measured temperature ≈ 23 °C).

4.2.4 Corrosion Tests for Steel Plates

(1) Testing Methods

The non-coated test pieces taken out of each seawater testing tank were immersed in 10% ammonium citrate for 24 hr to remove the rust,

washed in water, weighed after drying, and their average corrosion rates (mm/Y) were calculated from the weight losses. In addition, with regard to several portions in which the corrosion of the test pieces remarkably took place, the steel plate thicknesses were measured by a CPM 15-25 point micrometer (Sanpo Seisakujo Co., Ltd.), and an average of three points in which the corrosion rates were largest was looked upon as the minimum thickness of the test pieces, followed by a calculation of the local corrosion rates (mm/Y) from a comparison with the thickness before testing. The test pieces that had rust removed from them were not used again in subsequent test.

(2) Results

The average corrosion rates (mm/Y) are given in Table 4-10 and Fig. 4-14, while the local ones (mm/Y) can be seen in Table 4-11.

Table 4-7 Environmental conditions for test pieces
(in small testing teank)

Environmental condition	Unit	Mrasurement value	Remarks
Temperature	°C	Average 6.7	Secular change in Figure 4-12
Dissolved oxygen	ppm	Average 13	"
pH	-	Average 7.8	Secular change in Figure 4-13
Electric conductivity	m Ω/cm	Average 33	"
Flow rate	cm/sec	Agitation by circulation	"

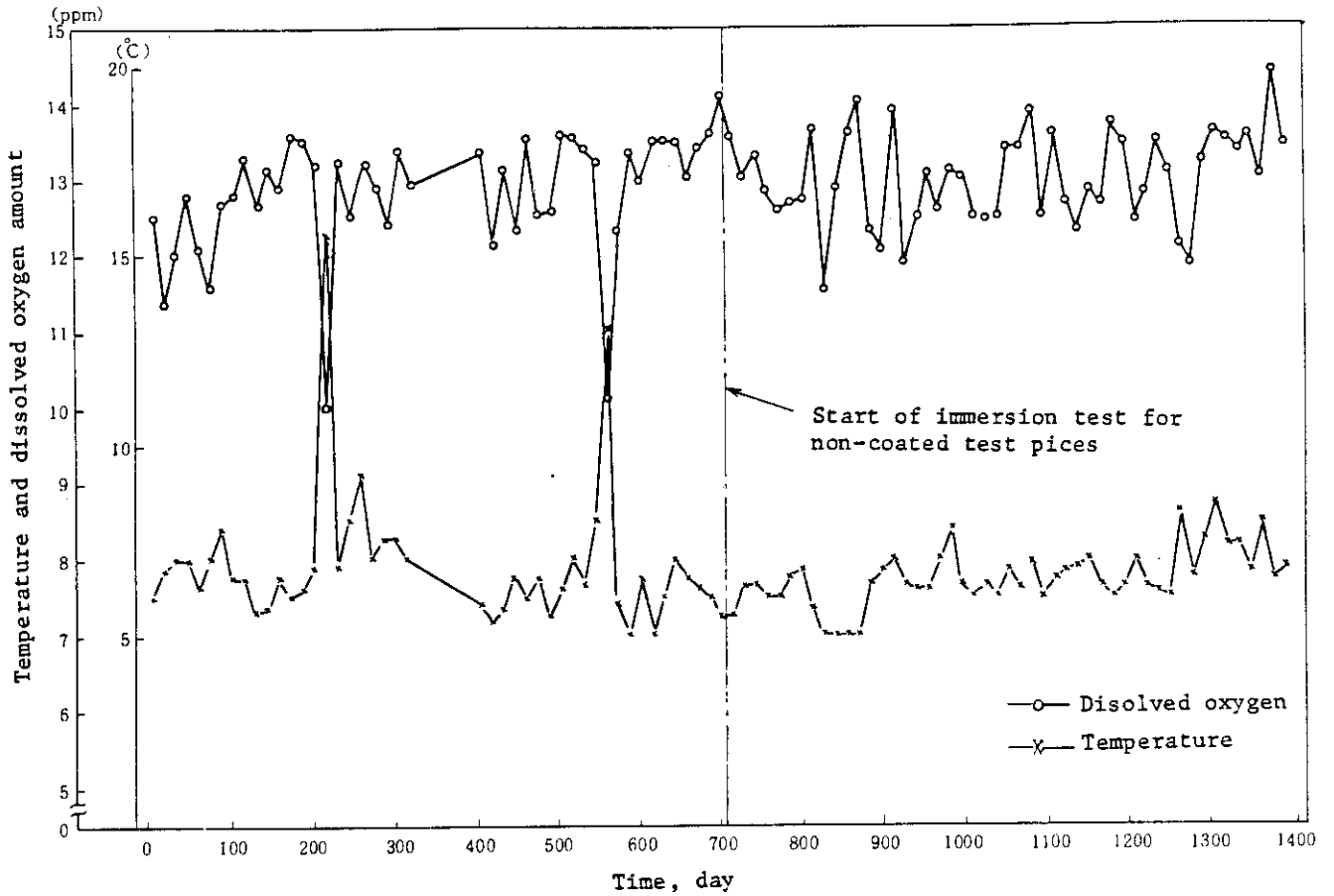


Fig. 4-12 Secular change in dissolved oxygen amounts and seawater temperatures in the small testing tank.

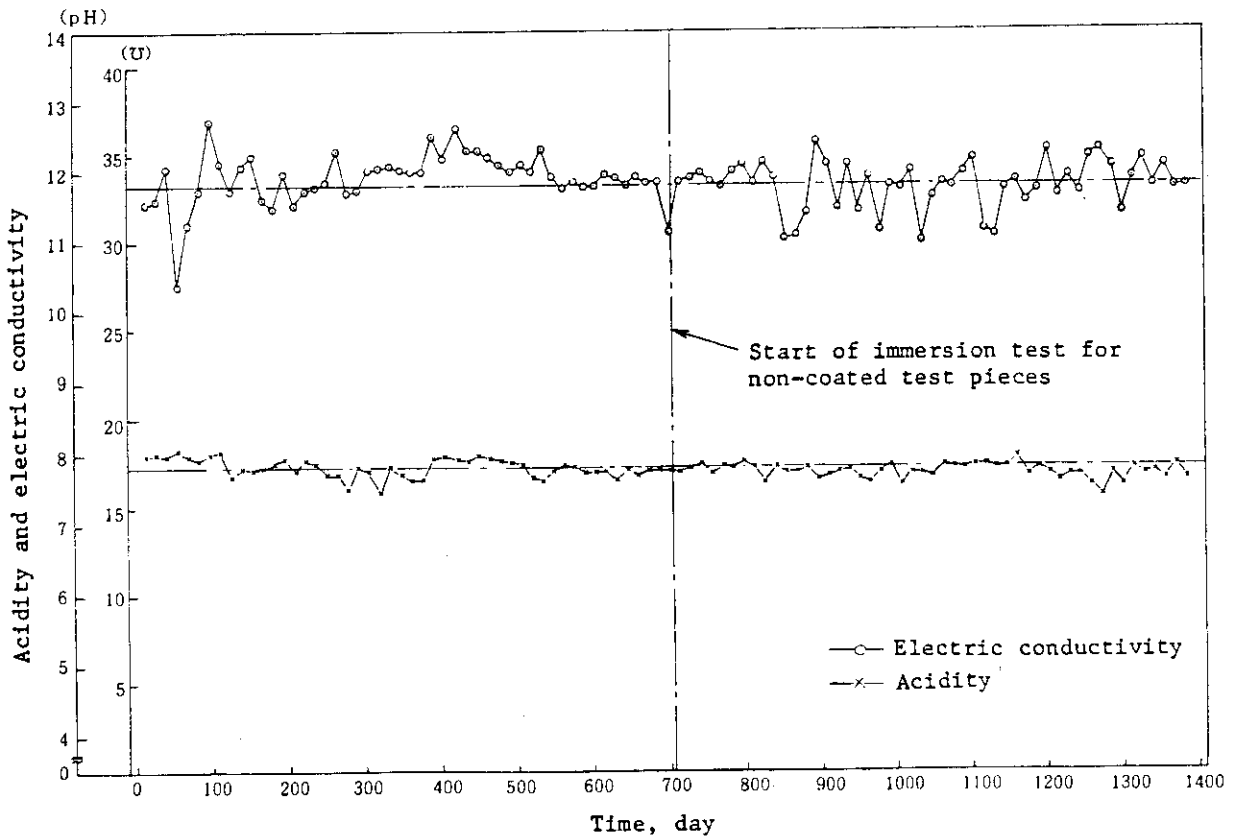


Fig. 4-13 Secular change in acidities and electric conductivities in the small testing tank.

Table 4-8 Results of test-piece observation after the seawater corrosion tests (6.7 °C)

Test piece No.	Corresponding test drum No.	Classification of anticorrosion treatment	Steel plate thickness	Immersion test period	Results of surface observation
2	2	Non-coating	1.6	24 months	The whole surface is covered with blown rust under which black rust covered the steel plate.
48	3	Melamine coating	1.2	18 months	A small amount of coated blisters were generated in the damage-free test pieces with a trace amount of red brown rust existing partly. Regarding the crosscut test pieces, coated blisters were generated over the whole surface, and red-brown rust was generated in part of the broken coatings.
5	4	Melamine coating	1.6	18 months	Coated blisters were generated over the whole surface of the damage-free test pieces, in which spallings and cracks were partly generated with red-brown rust. In the crosscut test pieces, rust covered about 80-% of steel plate.
54	5	Zinc-bearing anticorrosion Primer and epoxy coating	1.6	40 months	The damage-free test pieces were sound. As to the crosscut test piece, 3-to 5-mm dia. blisters and rust due to corrosion were generated along the crosscut.
56	6	Electric anticorrosion treatment and melamine coating	1.6	18 months	Spallings were observed over the whole surface of the coating over the damage-free test pieces, but steel plates were sound. In the crosscut test pieces, spallings occurred over the whole surface with a trace amount of thin red-brown rust being generated in the crosscut and around it.
43	8	Galvanization and melamine coating	1.6	18 months	Coated spallings occur over the whole surfaces of both the damage-free test pieces and crosscut ones, but the steel plates themselves were sound.
41	9	Stainless steel (SUS 304)	1.5	40 months	The test pieces were sound except for the generation of a trace amount of thin brown rust.

Table 4-9 Results of test-piece observation after the seawater corrosion tests (25 °C)

Test piece No.	Corresponding test drum No.	Classification of anticorrosion treatment	Steel plate thickness	Immersion test period	Results of surface observation
2	2	Non-coating	1.6	4 years	The whole surface of the test piece were covered with dark-brown rust with dark rust being generated underneath.
48	3	Melamine coating	1.2	4 years	Over the whole surface of the damage-free test piece 1-to 5-mm-dia. coated blisters were generated with about 3-mm-dia. brown rust existing in several portions. Corrosion under the coating also occurred due to cracks in the coating (about 10 %). As for the crosscut test piece, dark-brown rust was generated over the crosscut, around which 3-to 10-mm-dia. brown rust was generated in several places with a concentration (30 %) of 2-to 15-mm-dia. blisters.
47	4	Melamine coating	1.6	4 years	Over about 20-% of the surface of the damage-free test piece there existed 3-to 10-mm-dia. coated blisters with 10-mm-dia. brown rust existing in two places, with pimples being found on the edges of the coating with dark rust beneath. Dark brown rust was generated over the whole area of the crosscut with 5-mm-dia. brown rust being generated in several other areas. In addition, 3-to 5-mm-dia. blisters were found on about 15 % of the coating, and dark rust was generated partly over the steel plate because of the pimples on the surface of the coating.
54	5	Zinc-bearing anticorrosion primer and epoxy coating	1.6	4 years	Over 50 % of the surface of the damage-free test piece was found coated blisters having a diameter size of 10-mm or less, with 5-mm-dia. brown rust existing in one place. As for the crosscut test piece, thin brown rust was generated over it with large blisters surrounding it. Over the crosscut and on its edges rust and thin one due to corrosion under the coatings were found with blisters occupying about 70% of the surface.
34	6	Electric anticorrosion treatment and melamine coating	1.6	4 years	Coated blisters cover the whole surface of both the damage-free and crosscut test pieces, and consequently had no protective function. Over the surface of spalled coatings and beneath them white zinc oxides were formed.
43	8	Galvanization and melamine coating	1.6	4 years	The coating-protecting function was lost in both the damage-free and crosscut test pieces, and in 30-% of the surface of the damage-free test piece and in 40-% of the surface of the crosscut test piece brown and dark rust were generated, respectively. In places where there was no rust white zinc oxides were formed.
41	9	Stainless steel (SUS 304)	1.5	4 years	The test piece was sound except for a trace amount of brown rust and a different colored rust on part of the edges.

Table 4-10 Corrosion amounts, corrosion depth and average corrosion rates for seawater calculated from weight changes of non-coated test pieces.

Time (day)	Corrosion amounts ** (mg/cm ²)		Corrosion depth (mm)		Average corrosion rate (mm/Y)	
	6.7°C	23°C	6.7°C	23°C	6.7°C	23°C
90	18.33	-	0.023	-	0.095	-
182(187)	20.55	26.05	0.026	0.033	0.053	0.065
365	25.98	51.67	0.033	0.066	0.033	0.066
549	39.78	-	0.051	-	0.034	-
748(730)	36.82	95.32	0.047	0.121	0.023	0.061
849	-	109.37	-	0.139	-	0.060
1679	-	178.32	-	0.227	-	0.050

* The time is the value at 23°C.

** The average value for two test pieces except for values for 1679 days for which one test piece was used.

Table 4-11 Local thickness change and local corrosion rates for non-coated test pieces.

Time (day)	Thickness before testing (mm)		Minimum thickness after testing** (mm)		Maximum thickness change (mm)		Local corrosion rate (mm/Y)	
	6.7°C	23°C	6.7°C	23°C	6.7°C	23°C	6.7°C	23°C
90	1.494	-	1.367	-	0.127	-	0.26	-
182(187)	1.488	1.487	1.324	1.314	0.164	0.173	0.16	0.17
365	1.489	1.487	1.134	0.994	0.355	0.493	0.18	0.25
549	1.492	-	1.059	-	0.433	-	0.14	-
748(730)	1.483	1.492	1.020	0.710	0.463	0.782	0.11	0.20
847	-	1.480	-	0.515	-	0.965	-	0.21
1679	-	1.912	-	0.368	-	1.544	-	0.17

* The time is the value at 23°C.

** The average value for the three points in which thickness change was largest: The values for 1679 days were obtained by using one test piece, other ones were the average of two test pieces.

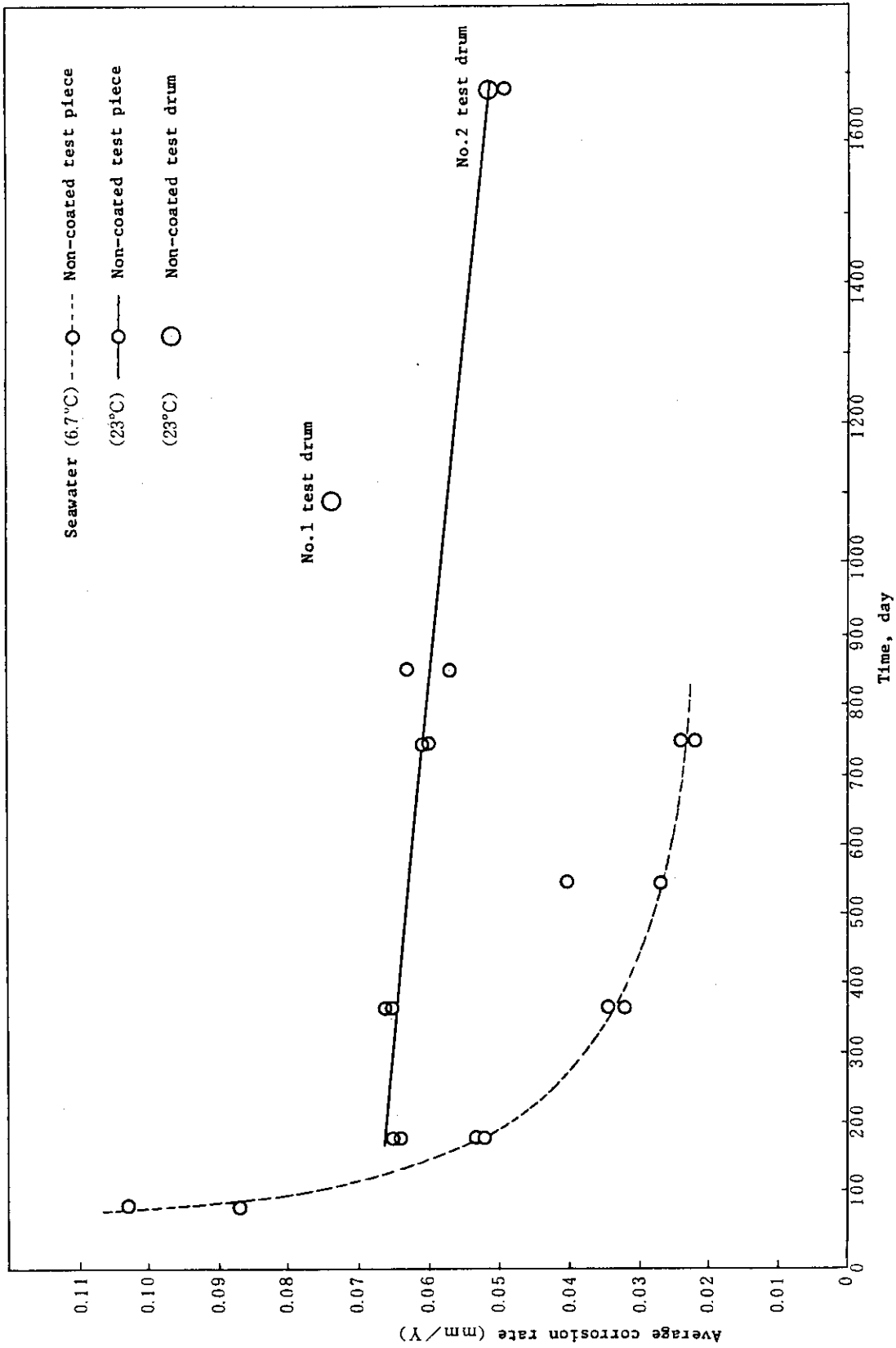


Fig. 4-14 Average corrosion rates for seawater calculated from weight changes of non-coated test pieces.

4.3 Internal Surface Corrosion Tests

When the drum container enclosing low-level radioactive waste is disposed of at sea, seawater may seep into the drum container through the gaps between the container and clamp because of the high pressures existing at the bottom of the sea. The corrosion resistance of the drum container, therefore, must be estimated by taking into consideration seawater seepage into the drum along with internal corrosion.

In the seawater immersion tests for drum containers, seawater seepage into the container cannot be imagined because the top plate and shell are tightly clamped. Consequently, the internal surface corrosion of the drum container was tested here by using buckets and test pieces consisting of the same material as those of the drum container with seawater being allowed to seep into the cans in advance.

4.3.1 Testing Methods

4.3.1.1 Internal Surface Corrosion Tests for Non-coated Buckets

Each of the three 18-l buckets (steel plate thickness ... 0.5 mm) was filled with simulated cemented waste having the composition shown in Fig. 3-1 with the upper 20 mm left unfilled. The top plate was then closed, clamped by a 6 and, a 5-mm-dia. hole was drilled in the center of the top plate, and the bucket was immersed in the seawater of the testing tank. After immersion, the top plate was lightly tapped to confirm whether or not the bucket was filled with water. The seawater tank used in a series of these tests was part of the tank for the drum-container immersion tests (See Fig. 4-1); accordingly, the environmental conditions in the former tank were the same as those in the seawater immersion tests for the drum containers (See 4.1.2).

After immersing in seawater for 6, 12 and 18 months, the buckets

were taken one by one out of the seawater tank, the top plates were removed, and the shells were cut into two parts using a welder to remove the cemented waste, followed by a visual observation of corrosion in the external and internal surfaces of the buckets. The seawater that had seeped into the buckets, on the other hand, were subjected to a corrosion measurement for such things as the dissolved oxygen amount, acidities and electric conductivities.

4.3.1.2 Internal surface Corrosion Tests for coated buckets

The simulated low-level radioactive waste was poured into three 18-l buckets coated with the same coatings (external surface ... melamine, internal surface ... epoxy resin) as those for the drum containers, with the upper 20 mm left unfilled. After closing and clamping the top plates, a 5-mm-dia. hole was drilled into the center of each bucket; then, the buckets were immersed in the seawater tank, and confirmation was made on whether or not the seawater had seeped into the buckets through the hole drilled into the buckets. The environmental conditions of the seawater in the tank were the same as those in 5.1.1. After seawater immersion for 6, 12 and 18 months, the buckets were taken out of the tank, cut longitudinally by a gas welder, and visually observed after removing the cemented waste from the buckets. In addition the corrosion factors of the seawater that had seeped into the buckets were measured, with such things as the dissolved oxygen amount, acidities and electric conductivities being measured.

4.3.1.3 Corrosion Tests for In-buckets Test Pieces

Two 150-mm-long, 75-mm-wide, 1.6-mm-high non-coated test pieces made of the same material as those of the drum container (SPHC) were set both within and on the cemented waste enclosed in the coated buckets described in 4.3.1.2. After immersing for 6, 12 and 18 months, they

were taken one by one out of the buckets in the testing tank, visually observed, immersed in 10- % ammonium citrate for 5 hr to remove the rust from the surface for weight measurement, followed by a calculation of the average corrosion rates (mm/Y) from the weight losses obtained by a comparison with the measured values before testing.

4.3.2 Test Results

4.3.2.1 Internal Surface Corrosion Tests for Non-coated Buckets

Table 4-12 and Photo. 4-5 show the results of visual observation of the non-coated buckets after the 18-month seawater immersion test. Although the external surfaces were covered with red rust, a little rust was found on the internal surface and near the band with other portions being sound. In particular, the portions with which the cemented waste was in contact with had an unchanged metal luster.

4.3.2.2 Internal surface Corrosion Tests for Coated Buckets

Table 4-13 and Photo. 4-6 give the results of visual observation for the coated buckets after the 18-month seawater immersion test. The spallations of the coating and rust were partly found the bents, welds and in the spliced parts except for the bands in which rust was generated over the whole external surface. No rust, however, was generated in the internal surface, and judged therform to be sound.

4.3.2.3 Corrosion Tests for In-bucket Test Pieces

(1) Results of Visual Observation

Table 4-14 and Photo. 4-7 show the results of the visual observation for the in-coated-bucket, non-coated test pieces after the 18-month seawater immersion test. No visual change was seen, compared with the condition before testing, in the test pieces buried in the cemented waste. A minor amount of rust was partly found on the test pieces which

were placed in the upper part of the buckets and were always in contact with seawater and land water that had seeped in.

(2) Corrosion Rates of Steel Plates

Table 4-15 shows the average corrosion rates (mm/Y) calculated from the weight change of the non-coated test pieces placed in the coated buckets for the 6-, 12- and 18-month immersion tests. Table 4-16 gives the dissolved oxygen, acidities and electric conductivities of the seawater that had seeped into the buckets in which the test pieces were immersed. Table 4-17 shows the weight losses of the non-coated test pieces after being immersed in 10- % ammonium citrate for 5 hr; namely, this was, as it were, a blank test for acid rinsing to remove the rust, and the corrosion amounts of test pieces listed in Table 4-18 were corrected values by using the results of the blank test.

Compared with an average corrosion rate of 0.063 mm/Y of the non-coated test pieces immersed for 18 months in the seawater in the same tank as that in which the buckets were dipped (seawater outside the buckets), the average corrosion rate of steel plates in the seawater that had seeped into the buckets was extremely small and show a value of 0.0006 mm/Y. This directly corresponds to the fact that the amount of dissolved oxygen in the seawater outside the buckets was 8 ppm but that inside the buckets was 1 ppm. The above phenomena indicate that the dissolved oxygen that had seeped into the buckets through the small hole in the top plate was not from the outside; in other words, the exchange of seawater between the inside and outside of the buckets was extremely difficult. In addition, a pH value of 9.8 shown by the seawater intruded into the pail can, which is smaller than that of 8.1 shown by the seawater outside the can, gives rise to a small amount of steel plate corrosion inside the buckets.

4.3.3 Conclusions

With regard to ocean disposal of low-leveled radioactive waste enclosed in drum containers, for the purpose of evaluating the corrosion resistance of drum-container steel plates at the time of seawater seepage, a series of corrosion tests were carried out for steel plates immersed in the seawater that had seeped buckets using 18-l buckets and test pieces made of the same material as those of the drum containers. The results are summarized as follows:

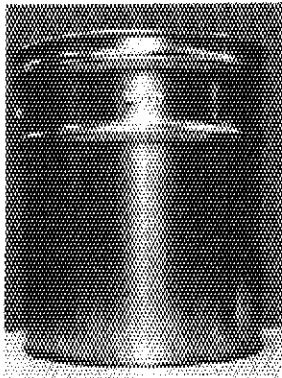
- (1) The corrosion amount of steel plates immersed in the seawater that had seeped into buckets was 1/50 or less for that outside the buckets. In particular, no visually observable rust was generated, after the 18-month immersion test, for the steel plates which were in contact with the cemented waste.
- (2) The amount of dissolved oxygen in the seawater that had seeped into the buckets was 0.9 ppm, whereas that outside was 8 ppm corresponding to about 1/8 of the former, showing that a smaller amount of steel plate corrosion occurred inside the buckets.
- (3) The pH value of the seawater inside the buckets was 9.8, and was higher than the pH value of 8.1 outside, presumably demonstrating that a smaller amount of steel plate corrosion occurred inside the buckets.

Table 4-12 Results of visual observation for non-coated buckets after the 18-month immersion in seawater.

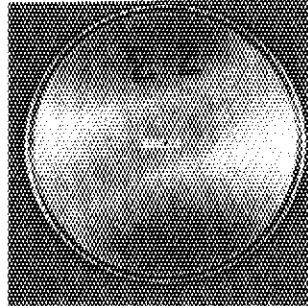
Classification of anticorrosion treatment	Steel plate thickness (mm)	Test zone	External surface	Internal surface
Non-coating	0.5	Top plate	The whole surface was covered with thick red-brown rust, in which 5-mm-dia. swollen rust (black rust beneath) was found.	About 80% of the whole surface was covered with a thin green precipitate which may be due to the dissolution of the rubber packing, but no rust was found on the surface of the steel plate indicating its soundness. There existed, moreover, the traces of seawater seepage through the packing.
		Shell	The whole surface was covered with thick red-brown rust, especially the swollen rust along the weld (black rust beneath).	Thin green precipitates were found from the bent to the boundary of the simulated cemented waste with thick green precipitates existing partly. No rust, however, was generated on the surface of the steel plate, indicating its soundness.
		Bottom plate	Black rust was observed along the peripheral part of the bottom plate (corresponding to about 20% of the whole surface), and a trace amount of thin brown rust in other parts.	The bottom plate was sound.
		Band	The whole surface was covered with thick red-brown rust (black rust beneath).	The whole surface was covered with thin green precipitates with black rust generated partly.

Table 4-13 Results of visual observation of coated buckets after the 18-month immersion in seawater.

Classification of anticorrosion treatment	Shell plate thickness (mm)	Test zone	External surface	Internal surface
External surface ... melamine coating Internal surface ... epoxy coating Band ... galvanization	0.5	Top plate	Pimples of 1-to 5-mm diameter were found on about 80% of the whole surface with 1% of them being coating spallings.	A part of the rubber packing adhered to about 1% of the periphery, and thin white precipitates were observed there, but the coating itself was almost completely sound.
		Shell	About 2-to 10-mm-dia. blisters were generated over the 70% of the whole surface with pimples existing near the bent. In addition, a minor trace of rust generation and coating swelling due to corrosion under the coating were observed.	About 65% of coating was spalled when the cemented waste was removed. No rust was generated on the spalled surface of the steel plate.
		Bottom plate	A minor amount of 2-to 4-mm-dia. blisters were generated, together with black rust due to corrosion under the coating. The other places were sound.	About 20% of the coating was spalled when the cemented waste was removed. No rust was found on the spalled surface.
		Band	Nearly all of the surface was covered with brown rust with thick rust swellings in several areas (black rust beneath).	Nearly all surface was covered with white precipitates with black rust generated on 1% of the area. A trace of thin brown rust was found on the rest of the area.

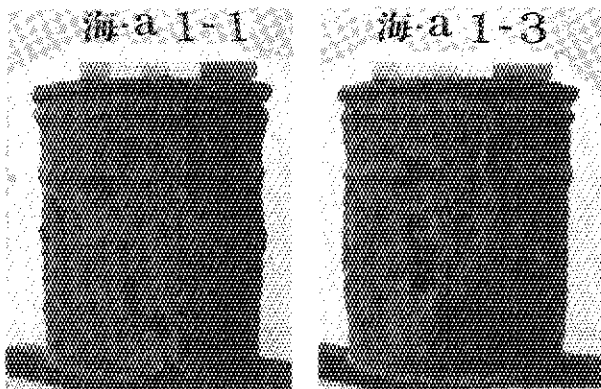


External surface of shell

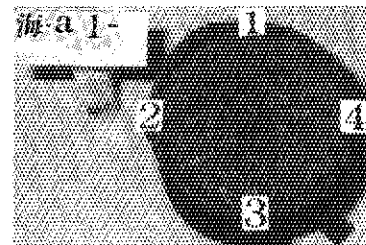


External surface of top plate

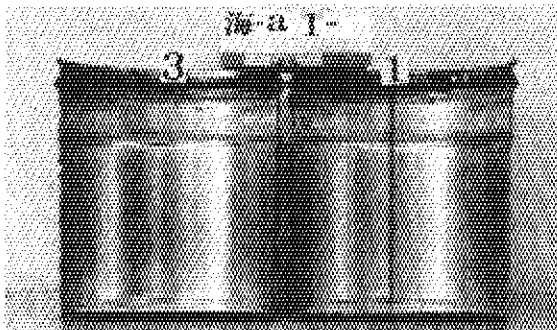
1) Bucket before testing



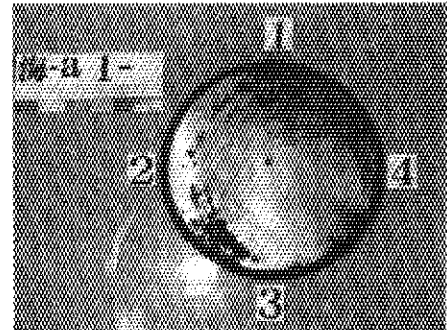
External surface of shell



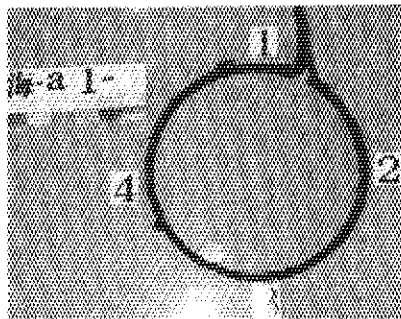
Internal surface of top plate



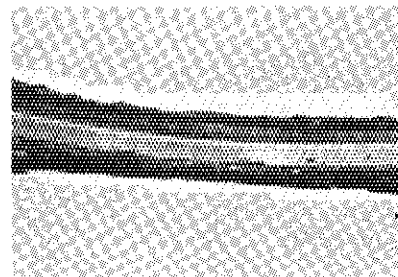
Internal surface of shell



Internal surface of top plate



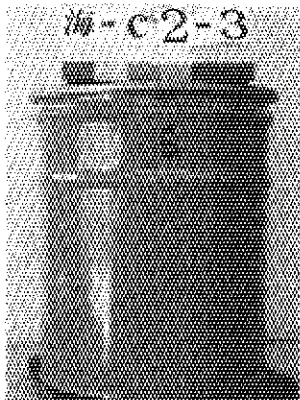
External surface of band



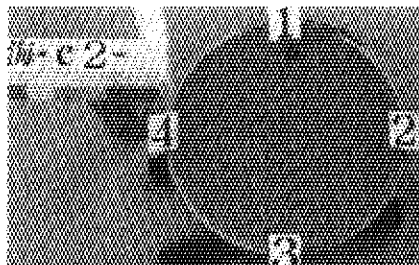
Internal surface of band

2) Bucket after testing

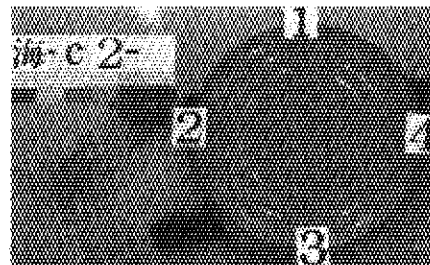
Photograph 4.5 Non-coated bucket after the 18-months immersion in seawater (24°C).



External surface of shell

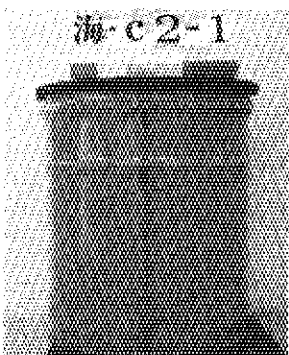


External surface of top plate

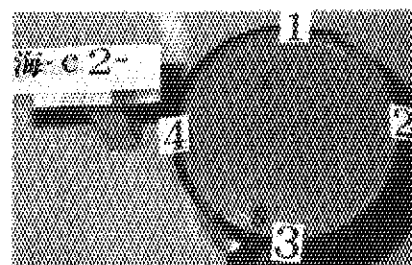
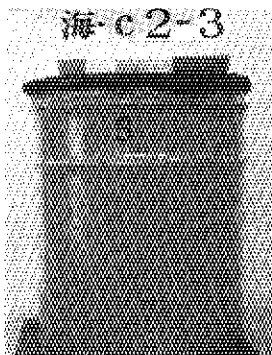


External surface of bottom plate

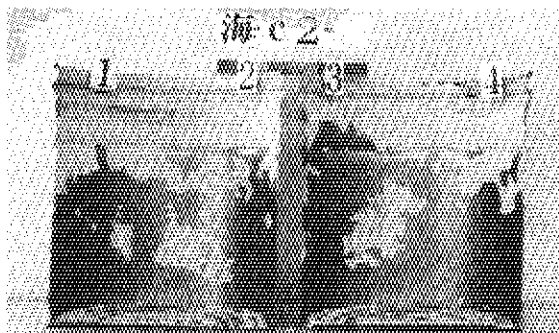
1) Bucket before testing



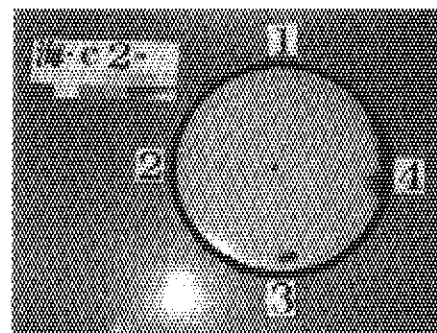
External surface of shell



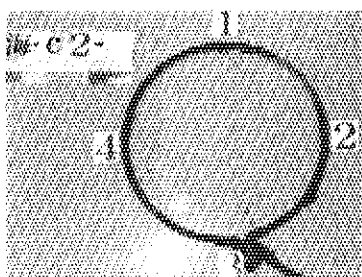
External surface of top plate



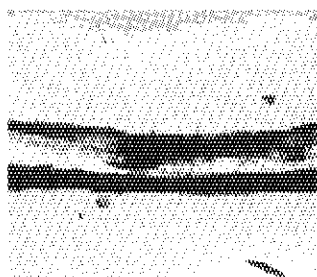
Internal surface of shell



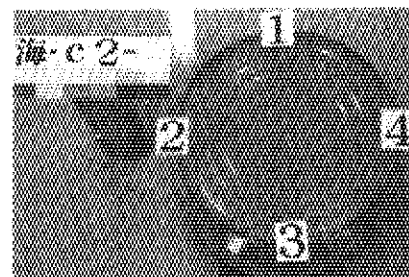
Internal surface of top plate



External surface of band



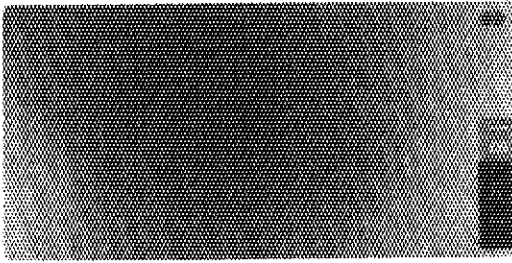
Internal surface of band



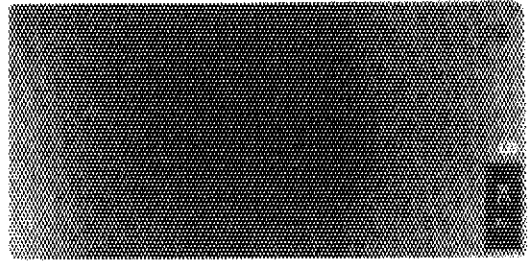
External surface of bottom plate

2) Bucket after testing

Photograph 4.6 Melamine-coated bucket after the 18-months immersion in seawater (24°C).

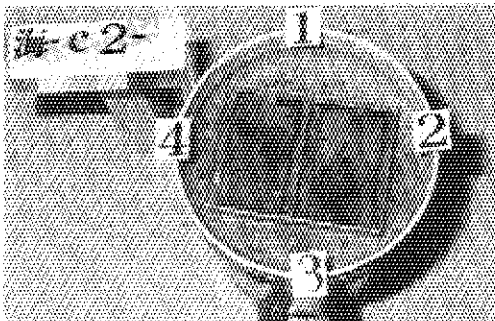


Test piece in in-bucket liquid

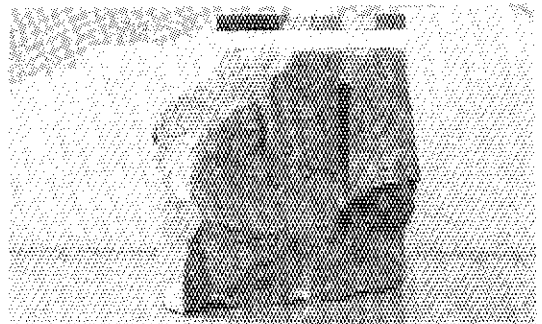


Test piece buried in cemented waste

1) Test pieces before testing

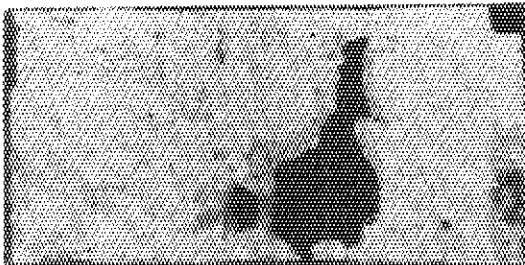


Position in in-bucket liquid

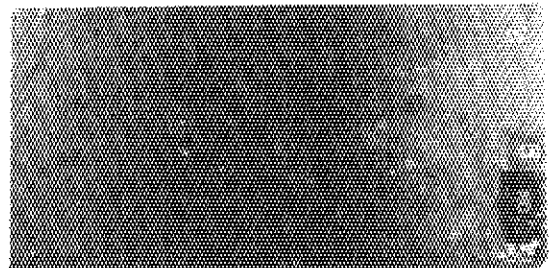


Position in cemented waste

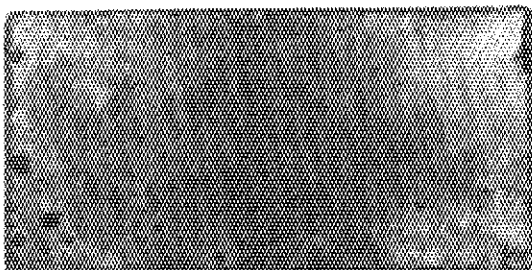
2) Positions of test pieces in bucket



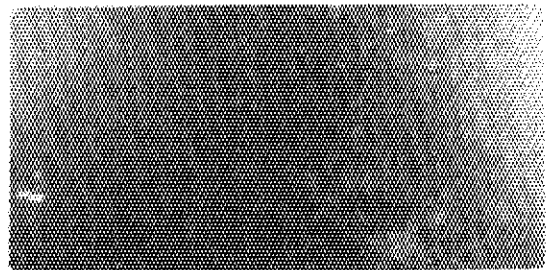
Test piece in in-bucket liquid (surface)



Test piece buried in cemented waste (surface)



Test piece in in-bucket liquid (reverse)



Test piece buried in cemented waste (reverse)

3) Test pieces after testing

Photograph 4.7 Non-coated test pieces in bucket after the 18-months immersion in seawater (24°C).

Table 4-14 Results of visual observation for non-coated buckets after the 18-month immersion in seawater.

Classification of anticorrosion treatment	Steel plate thickness (mm)	In-bucket environment	Test piece surface
non-coating	1.6	In bucket liquid	About 85 % of the front area of the test piece was covered with a mixture of white precipitates and brown rust, whereas the back area had a thin grey flilm.
		Burid in cemented waste	No rust was generated, and therefore sound.

Table 4-15 Average corrosion rate calculated from weight changes of non-coated test pieces in buckets immersed in seawater.

Time (day)	Testing environment	Weight change (g)	Corrosion amount (mg/cm ²)	Average corrosion rate (mm/Y)
194	In bucket liquid	0.2265	1.007	0.0024
		0.2392	1.063	0.0026 (0.0025)*
365	Buried in cemented waste	0.2755	1.224	0.0029
		0.2643	1.175	0.0028 (0.0029)*
547	In bucket liquid	0.2545	1.131	0.0014
		0.2408	1.070	0.0013 (0.0014)*
547	Buried in cemented waste	0.2424	1.077	0.0014
		0.2520	1.120	0.0014 (0.0014)*
547	In bucket liquid	0.1272	0.565	0.0005
		0.1507	0.670	0.0006 (0.0006)*
547	Buried in cemented waste	0.2154	0.957	0.0008
		0.2466	1.096	0.0009 (0.0009)*

* Average value for two test pieces

Table 4-16 Dissolved oxygen, acidities and electric conductivities of liquids in buckets immersed in seawater.

Testing environment	Time (day)	Acidity (pH)	Electric conductivity (Ω /cm)	Dissolved oxygen (ppm)
Seawater	Seawater in tank (outside bucket)	8.1 (21°C)	49 (21°C)	8.0 (21°C)
	194	12.4 (21°C)	49 (21°C)	1.0 (21°C)
	365	13.0 (21°C)	54 (21°C)	1.0 (21°C)
	547	9.8 (22°C)	46 (22°C)	0.9 (21°C)

Table 4-17 Weight losses of non-coated test pieces in 5-hour immersion in 10% ammonium citrate solution.

Test piece No.	Weight before immersion in 10-% ammonium citrate solution	Weight after 5-hour immersion in 10-% ammonium citrate Solution	Weight loss (g)
1	140.3625	139.8779	0.4846
2	138.1914	137.6220	0.5694
			Average 0.5270

5. Integrity Evaluation for Drum Containers

5.1 Evaluation Methods

5.1.1 Evaluation Philosophy

The objective of this paper lies in evaluating the corrosion resistances of drum containers for low-level radioactive waste under a seawater environment, so the evaluation philosophy must be made clear in advance.

The minimum role required for the low-level radioactive waste drum containers is to prevent the radioactive material from leaking out or dispersing and to transport them to a place for safe disposal. The drum containers, however, still have the capabilities of suppressing the release of radioactive nuclides contained in the low-level radioactive waste to the environment and of reducing environmental exposure. If the radioactive nuclides in the cemented waste are assumed to be released mainly by seawater leaching, the information on the leached-out rates of the radioactive nuclides from the cemented waste contained in the drum container is necessary to evaluate its barrier capacity for preventing the release of radioactive nuclides into the environment. In this paper, the corrosion resistances of the drum containers were evaluated so that their barrier capacities against the release of radioactive nuclides to the environment might be calculated when the information on the leached-out rates of the radioactive nuclides in the cemented waste is available.

5.1.2 Evaluation Methods

The corrosion of the drum container under a disposal environment may proceed as follows from the long-term point of view: At first,

the coating deteriorates and partly loses its protective function for a steel plate. The deterioration of the coating then extends to the whole surface, followed by overwhelming corrosion of the steel plate. In the subsequent stage, corrosion through-holes were generated partly on the steel plate, extended to the whole surface, and finally the drum container lost its protective function for the cemented waste in it.

Thus, the corrosion evaluation means here to clarify the process of the generation of a corrosion through-hole in the drum container and of its growth as time passes. From this point of view, the corrosion resistances of the drum containers were carried out under various testing conditions.

It is difficult, however, to complete in a few years the above-mentioned tests in which the process of corrosion through-hole generation and its growth is to be clarified. In a series of the tests, therefore, the long-term durabilities of coatings and steel plates were first estimated through the former's deterioration and the latter's corrosion by using separate data, followed by an overall estimation by combining the two results.

Generally, the top plate and shell of a drum container are clamped by a band by using packing, so that water barely ever seeps into the container. In the case of ocean disposal, seawater may seep into the container because of hydraulic pressure; accordingly, seawater seepage was assumed in this evaluation.

If corrosion is assumed to proceed uniformly and at a constant rate for a non-coated steel plate, time t at which the plate thickness becomes zero due to corrosion is obtained from Eq. (1).

$$t = H/m \quad (1)$$

where H represents the original plate thickness (mm), and m the corrosion rate (mm/y) of the steel plate. However, as indicated in the example in Fig. 4-11, the corrosion of the steel plate proceeds unevenly. In other words, the surface of the steel plate and corrosion environment are not uniform microscopically, and show, as a whole, distribution $q(m)$ given in Fig. 5-1. This trend is looked upon as the corrosion probability of the steel plate, and probability distribution $q(m)$ of corrosion rate m is assumed to be constant in this evaluation.

Corrosion rate m (mm/y) in the axis of abscissa in Fig. 5-1 corresponds, from Eq. (1), to the time required for initial steel plate thickness H to become zero due to corrosion at a rate of m; namely, distribution $q(m)$ represents the ratio of the surface area in which the thickness becomes zero in time t to the whole surface area of the steel plate. Consequently, the integration of distribution $q(m)$ from larger m (from smaller t) as shown in Eq. (2) gives curve $g(t)$ which indicates the secular change in the total corrosion-through-hole area over the steel plate (Fig. 5-2).

$$g(t) = - \int_{\infty}^{H/t} q(m) dm \quad (2)$$

The area of deteriorated coating, which has lost its ability to protect the steel plate from corrosion, is represented by $P(t)$ as a function of time (Fig. 5-3). If the corrosion of a newly-formed area of deteriorated coating is assumed to be given by equation (2), whenever it is generated, the function $G(t)$ showing the area of perforation per unit area of the coated drum plate can be written as equation (3) (Fig. 5-4).

$$G(t) = \int_0^b g(t-\tau) \frac{dP(\tau)}{d\tau} d\tau \quad (3)$$

where b means the time required for $P(t)$ to reach 1.

The above discussions concern the same material, but as shown in Fig. 5-5, the drum container consists of a top plate, a shell, a band and a bottom plate, and all of them differ in their corrosion resistances. When the secular changes in the through-hole-area ratios of a whole drum container and each of its component part are represented by $G(t)$, $G_1(t)$, $G_2(t)$, ..., $G_6(t)$, $G(t)$ can be expressed as the weighted average of $G_1(t)$, $G_2(t)$, ..., $G_6(t)$ as shown below.

$$G(t) = \frac{\sum_i W_i G_i(t)}{\sum_i W_i} \quad (4)$$

where W_i means the surface area ratio of each part of the drum container (Table 5-1).

To determine corrosion-through-hole-increasing curve $G(t)$ in terms of time elapsed by means of the above-mentioned method, it is necessary to determine probability distribution $q(m)$ for the steel plate with a corrosion rate of m together with the coating's deterioration-area-increasing curve $P(t)$.

5.1.2.1 Determination of Corrosion Rates of Steel Plates

(1) Corrosion Rates of External Surface of Steel Plates

As to the corrosion rate of the external surface of a steel plate, probability distribution $q(m)$ on the same steel plate, which is

estimated from the results of fixed-point thickness measurement for the container shell, is assumed to be effective hereafter.

Moreover, since the observation in seawater of each part of the drum container showed no significant corrosion difference between them, the corrosion rates in each part of the container are assumed to be equal.

In general, the decrease in the steel plate thickness tends to be smaller as time passes (Fig. 5-6). This is because the increased layer of rust originated from corrosion may in turn play the role of a protective coating against the corrosion of steel plates. Accordingly, if the steel plate thickness is represented by steel plate thickness h , corrosion rate μ defined by Eq. (5) decreases as time passes.

$$\mu = - \frac{dh}{dt} \quad (5)$$

The corrosion rate of a steel plate obtained in this testing is not μ defined in Eq. (5), but m integrated from 0 to t defined by Eq. (6)

$$m = \frac{1}{t} \int_0^t \mu dt \quad (6)$$

(2) Corrosion Rates of Internal Surface of Steel Plate

As shown in the schematic figure of the state of corrosion (Fig. 5-7), the seawater corrosion conditions for the external surface of the drum container differ from those for the internal surface. As for the internal surface which is in contact with the cemented waste, its corrosion rate is about 1/50 or less that of the external surface, as explained in the results of the corrosion tests for

internal surfaces (Table 4-15), even if seawater seepage is assumed, and no visual corrosion is observed. In addition, the durability of the coating is strong, so its corrosion rate is assumed to be zero in the evaluation. Regarding the top plate and bent which are not in contact with the cemented waste, the corrosion amounts of their internal surfaces are 1/50 or less of those of the external ones, and, in fact, extremely small. However, minor corrosion was found visually, so that the corrosion rate of the internal surface of this part is assumed to be 1/10 of that of the external one. In short, with regard to the top plate and bent, the probability distribution of the corrosion rate of internal-plus-external surface of the steel plate is $q(m/1.1)$ with that of external surface being $q(m)$.

5.1.2.2 Determination of Coating's Deterioration-area-increasing Curves

(1) External Coatings

Curve $P_1(t)$ which indicates the secular change in the coating's deterioration-area of the drum container's shell is determined on the basis of the data obtained during the 4.7-year testing period. Curves $P_2(t)$ to $P_6(t)$ for the other parts of the container, on the other hand, are determined, as shown in Table 5-2, on the basis of the results of the visual observation with $P_1(t)$ for the shell being the standard. In other words, comparing the durabilities of each part with that of the shell, the values for the top plate and bent, the weld and spliced part, and the bottom plate are 1/2, 1/3 and 1, respectively.

(2) Internal Coatings

According to the results of tests, the deterioration of the internal coating is less severe than that of the external one. In particular, little coating deterioration is observed in the parts which are in contact with the cemented waste. Since the deterioration of the internal coating is not quantitatively understood, its durability is assumed to be equal to that of the external one upon evaluation; namely, the secular increase curve $P'(t)$ for the internal coating's deterioration-area is assumed to be equal to that of the external one $P(t)$ (Table 5-2). $P(t)$ (Table 13-2).

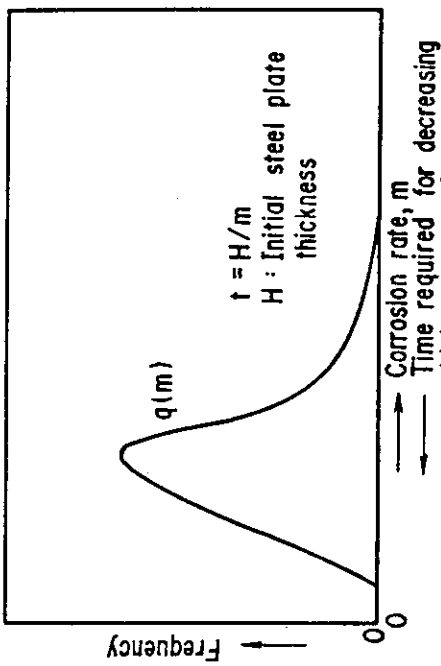


Fig. 5-1

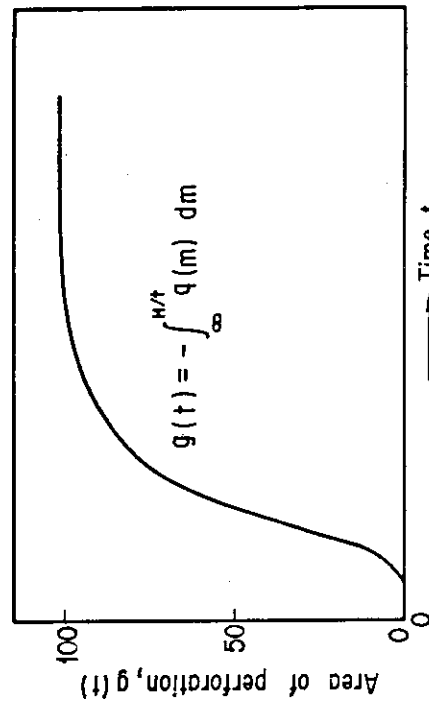


Fig. 5-2 Area of perforation of steel plate due to corrosion as a function of time.

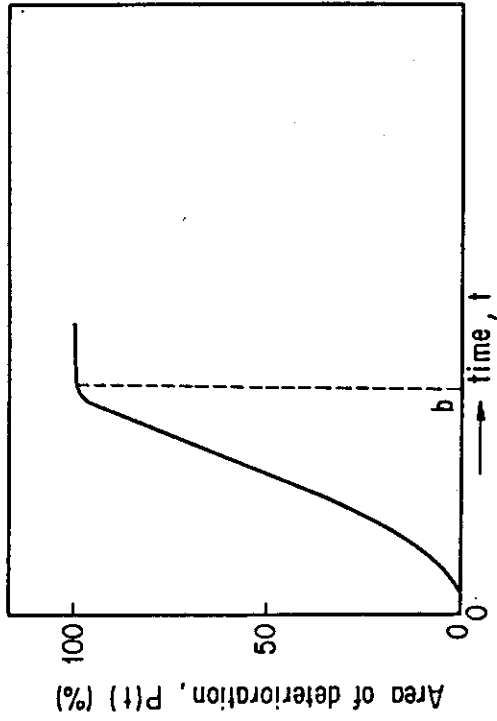


Fig. 5-3 Deteriorated area of drum coating as a function of time.

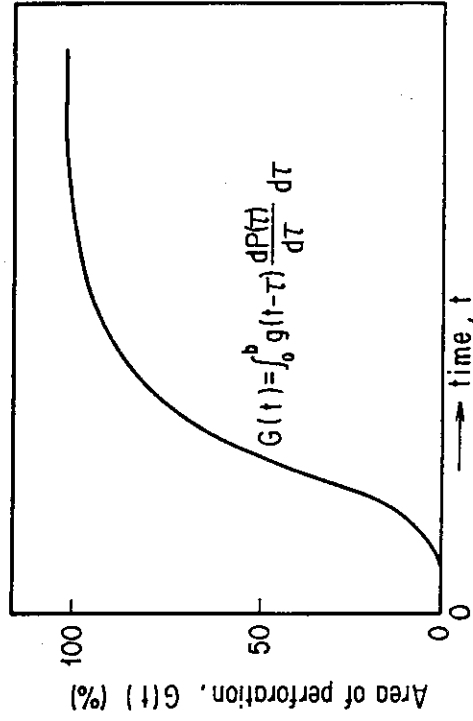


Fig. 5-4 Area of perforation of coated drum due to corrosion as a function of time.

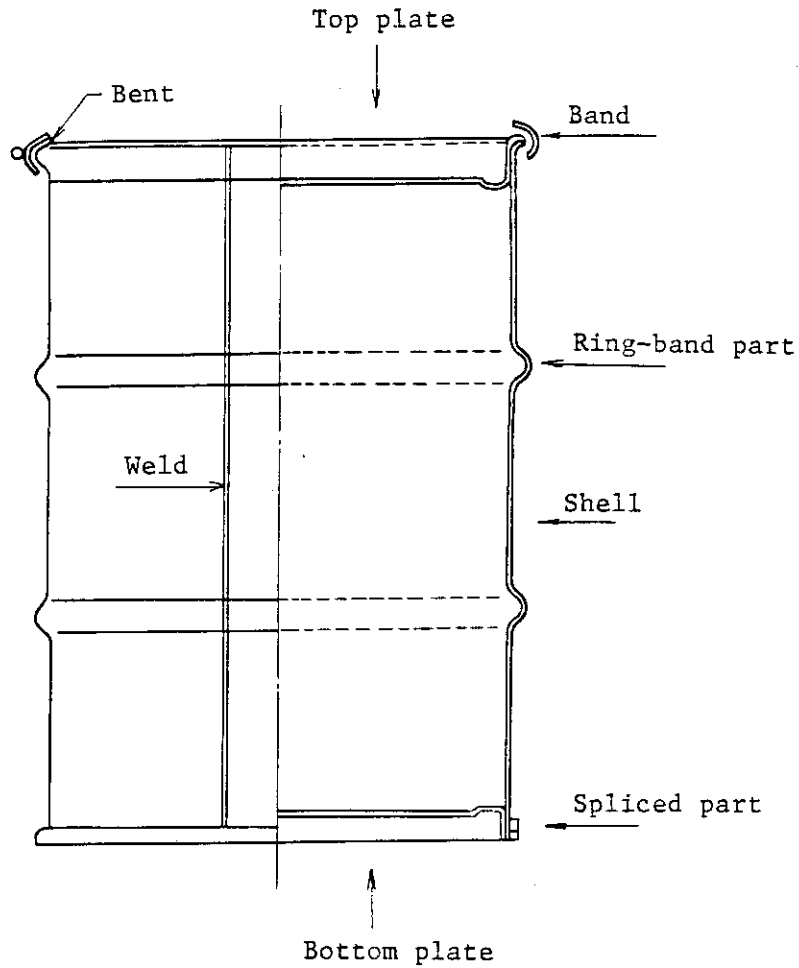


Fig. 5-5 Profile of drum

Table 5-1 Area ratio of each part of drum.

Part of drum	Area ratio W_i
Shell(Exclusive of bent, weld and spliced part)	0.60
Top plate	0.13
Bent	0.05
Weld	0.03
Spliced part	0.06
Bottom plate	0.13

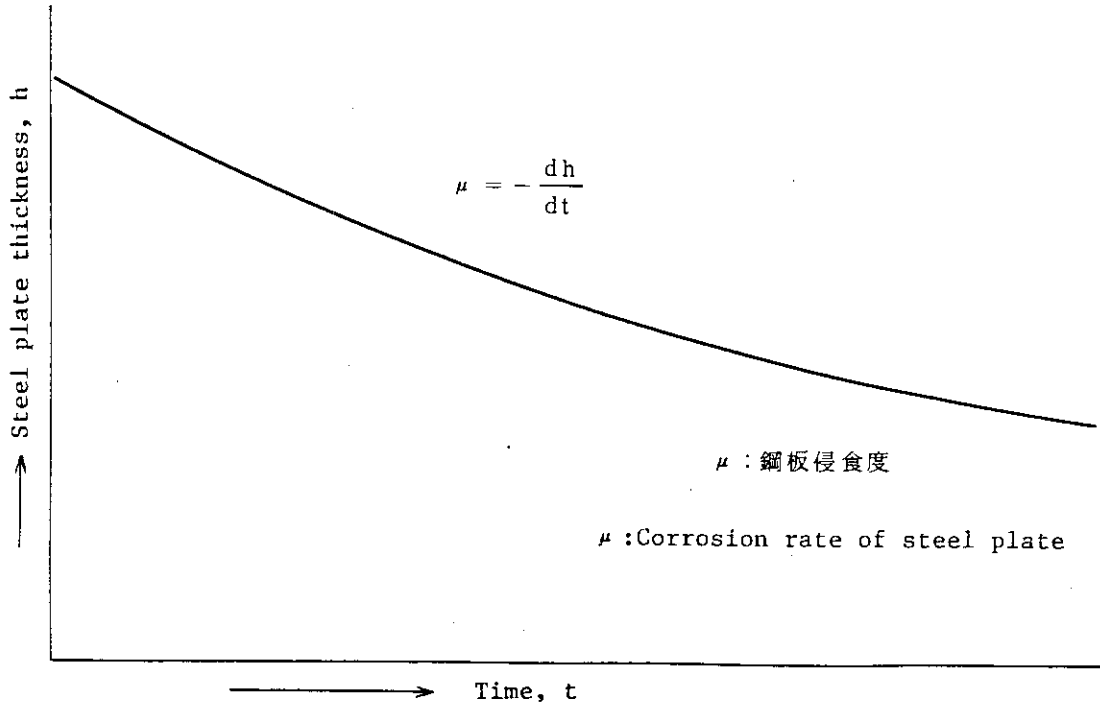


Fig. 5-6 Steel plate thicknesses under a corrosion environment.

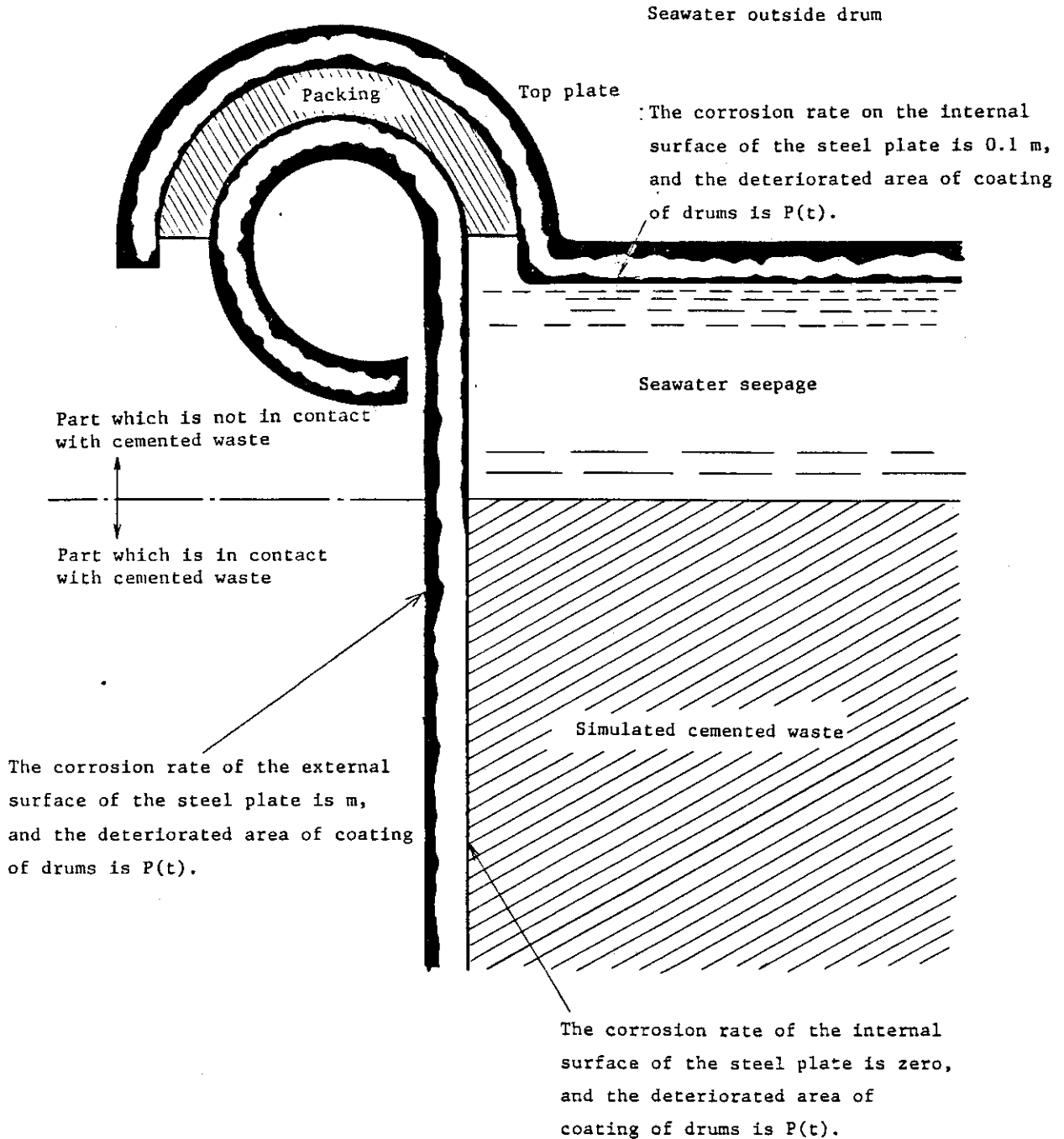


Fig. 5-7 Schematic figure for seawater corrosion of drums.

Table 5-2 Steel-plate corrosion rates and $P(t)$ in each part with the shell being the standard.

Part of drum	Steel-plate corrosion rate (mm/Y)		P(t)	
	External surface	Internal surface	External surface	Internal surface
Shell (exclusive of bent, weld and spliced part)	m	0	$P_1(t)$	$P'_1(t)=P_1(t)$
Top plate	m	0.1 m	$P_2(t)=P_1(2t)$	$P'_2(t)=P_1(2t)$
Bent	m	0.1 m	$P_3(t)=P_1(2t)$	$P'_3(t)=P_1(2t)$
Weld	m	0	$P_4(t)=P_1(3t)$	$P'_4(t)=P_1(3t)$
Spliced part	m	0	$P_5(t)=P_1(3t)$	$P'_5(t)=P_1(3t)$
Bottom plate	m	0	$P_6(t)=P_1(t)$	$P'_6(t)=P_1(t)$

5.2 Corrosion Resistance Evaluation for Drum Containers in a Seawater Environment

The corrosion resistances of various drum containers were evaluated in a sea water environment by the method described in 5.1, with the following conditions assumed:

- (1) The drum containers to be evaluated were a 200-l H-class open-type containers made of 1.6-mm-thick steel plates as specified in JIS 1600-1977 (See Table 3-1), with enclosed simulated cemented waste.
- (2) Upon evaluation the seawater was assumed to have seeped into the drum containers.
- (3) If an estimation is difficult because of insufficient test data, the corrosion amounts should be estimated as conservatively as possible.

5.2.1 Results of Evaluation

The seawater corrosion tests were carried out at 25°C (for drum containers and test pieces) and 5°C (for test pieces), and the average temperatures for the above two conditions throughout the testing periods were 23 and 6.7°C, respectively.

- (1) No. 2 Test Bodies (without coating)
 - 1) At a Seawater Temperature of 23°C

The frequency distribution of the corrosion amounts (mm) (Fig. 4-11), which were obtained from the results of fixed-point-thickness measurement for the non-coated external surfaces of the shell plates immersed in seawater for 55 months, was mathematically smoothed out, and then converted into a distribution in which the axis of abscissa represents corrosion rates (mm/y). The resultant probability distribution on the steel plate whose external-surface corrosion rate was m (Fig. 5-8(a)) was used for this evaluation.

As for the average value of the distribution, the value 0.052 mm/y (See Table 4-5) that was obtained by weight-loss measurements was used. (Although there were slight differences in the average thicknesses between the values of the weight measurements obtained and those due to thickness measurements, the former being employed as the average thickness.) Figure 5-9(a) gives the corrosion-through-hole-increasing curve $G(t)$ for the external surface of a non-coated steel plate. In addition, the corrosion rates of each part of the drum container (See 4.1.6) are assumed to be equal. Regarding the corrosion rates of the internal surfaces of steel plates, in accordance with the test results (See 4.3), the rates of the shell, weld, spliced part and bottom plate with which the cemented waste was in contact with were assumed to be zero, whereas those of the top plate and bent with which the cemented waste was not in contact with were 1/10 of those of the external surfaces of the steel plates.

Figure 5-10(a) shows the corrosion-through-hole-increasing curves for each part of the non-coated drum container $G_1(t)$, $G_2(t)$... $G_6(t)$, together with that of the drum container as a whole $G(t)$. All of them were calculated from that of $g(t)$ for non-coated plates.

2) At a Seawater Temperature of 6.7°C

An average corrosion rate of 0.023 mm/y, which was obtained by the on-the-same-steel-plate test pieces immersed in 6.7-°C seawater for 2 years (See Table 4-10), was used for investigating the corrosion rates of the external surfaces of the steel plates, whereas the distribution determined by the 25-°C seawater immersion tests for the "form" of the probability distribution on the steel plate; in other words, if the probability distribution of the external surface corrosion rate determined by the 23-°C seawater tests was $q(m)$, that

in the 6.7-°C seawater was equal to q (0.052 m/0.023) (Fig. 5-8(b)).

Figure 5-9(b) demonstrates the corrosion-through-hole-increasing curve $g(t)$ for the external surfaces of non-painted steel plates calculated from the above distribution by the same method as that described in 1).

The corrosion rates of the steel plates for each part of a drum container are completely the same as those in 1). Regarding the corrosion rates of the internal surfaces of steel plates, like in 1), the corrosion rates of the shell, weld, spliced part and bottom plate with which the cemented waste was in contact with were zero, whereas those of the top plate and bent with which the cemented waste was not in contact with were 1/10 of the corrosion rates of the external surfaces of the steel plates.

Figure 5-10(b) gives the corrosion-through-hole-increasing curves of each part of the non-coated drum container $G_1(t)$, $G_2(t)$, ..., $G_6(t)$ and of the container itself $G(t)$. All of them were calculated from the corrosion-through-hole-increasing curve for the external surface of the non-coated steel plate $g(t)$.

(2) No. 4 Test Bodies (melamine coating)

1) At a Seawater Temperature of 23°C

As to the external surface of the shell, the coating's deterioration-area-increasing curve $P(t)$ to be used for this evaluation was determined as shown in Fig. 5-11 on the basis of the 55-month test data. Regarding the other parts of the container, the $P(t)$ curve were determined from Table 5-2 by comparing each $P(t)$ with $P_1(t)$ for the shell. According to the test results (See 4.1.5), the degree of deterioration of the internal coating was smaller than that of the external one, but was not measured quantita-

tively, so that $P(t)$ of the internal coating was assumed to be equal to that of the external one.

The curve $P(t)$ thus determined and $g(t)$ defined in (1)-1), together with the method explained in 5.1 led to the determination of the corrosion-through-hole-increasing curves for each part of the drum container $G_1(t)$, $G_2(t)$, ... $G_6(t)$, followed by that of the drum container as a whole $G(t)$ in accordance Eq. (4) (Fig. 5-12(a)).

2) At a Seawater Temperature of 6.7°C

The results of a series of coating deterioration tests (See 4.2.3) in 23- and 6.7-°C seawater made clear that the temperature difference did not give rise to any significant change in coating deterioration, and so the coating's deterioration-area-increasing curve $P(t)$ of the external coating in 6.7-°C seawater was assumed to be equal to that (Fig. 5-11) in 23-°C seawater. As for the other parts, their $P(t)$ s were determined from Table 5-2 by comparing them with that of the shell $P_1(t)$. Like in 1), $P'(t)$ of the internal coating was assumed to be equal to $P(t)$ of the external one.

The curves $P(t)$ s thus obtained and $g(t)$ in 1) led to the determination of the corrosion-through-hole-increasing curves of each part of the container $G_1(t)$, $G_2(t)$, ... $G_6(t)$ as well as that of the container as a whole $G(t)$ (Fig. 5-12(b)).

(3) No. 5 Test Bodies (Epoxy-resin Coating)

Although the deterioration of coating during the 55-month seawater immersion test was too short to determine their coating's deterioration-area-increasing curves $P(t)$ s, the curve $P_1(t)$ shown in Fig. 5-13 was determined for the external coating of the shell by using the same method as described in (2). The $P(t)$ curves for the other parts of the drum container were obtained from

Table 5-2 by comparing them with the $P_1(t)$ of the shell. Like in (2), $P'(t)$ of the internal coating was assumed to be equal to the $P(t)$ of the external one.

Figure 5-14 shows the corrosion-through-hole-increasing curves of the each part of the drum container $G_1(t)$, $G_2(t)$, ... $G_6(t)$ obtained by using the $P(t)$ determined above and $g(t)$ defined in (1), along with that of the container as a whole $G(t)$.

(4) NO. 6 Test Bodies (Electric Anticorrosion Treatment and Melamine Coating)

In the electric anticorrosion treatment using zinc, the pH value over the surface of the steel plate shifted gradually to the alkaline side, and tended to cause spalling of the melamine coating because of its vulnerability to alkalis. As indicated in the results of the coating deterioration tests (Table 4-3), the blisters and spallings of the coating over the surface of the No. 6 test body were remarkable, so the average effective term of the coating was looked upon as a year in this evaluation.

The amount of zinc added initially to the test body was 6.300 g, but 1.005 g were consumed during the 55-month seawater immersion test. If the effective period of the coating is one year with the zinc-consuming period being 43 months, the annual zinc consumption is 281 g. If the electric anticorrosion efficiency is 85 % and zinc is consumed at a constant rate, the electric anticorrosion effect is expected to continue for 19 years. Therefore, if the coating's effective period and zinc's effective anticorrosion period are 1 and 19 years, respectively, the corrosion-through-hole-increasing curve $G(t)$ for the test body corresponds to the 20-year-shifted curve $G(t)$ (Fig. 5-10 (a)) for the non-coated drum container shown in Fig. 5-15.

(5) No. 8 Test Bodies (Galvanization and Melamine Coating)

As shown in the corrosion potentials of the No. 8 test body, the potential of galvanized zinc shifted to that of iron after about 1.5 years, and then rust began to generate. These test results led to the assumption that the effective anticorrosion period was one year once galvanized zinc was exposed to seawater.

In this evaluation, the blister-plus-spallation-increasing curve (dotted line in Fig. 5-16) for the external coating of the shell was determined by the test data, and the one-year steel-plate-protecting period thanks to galvanized zinc was added to the above curve to form the coating's deterioration-area-increasing curve $P_1(t)$ for the external coating of the shell (full line in Fig. 5-16).

Figure 5-17, by using the $P(t)$ thus obtained and $g(t)$ determined by (1), gives corrosion-through-hole-area-increasing curves of each part of the container $G_1(t)$, $G_2(t)$, ... $G_6(t)$, along with that of the container as a whole $G(t)$.

(6) No. 9 Test Bodies (Stainless Steel)

The amounts of corrosion of stainless steel (SUS 304), except for a part of the drum container, were less than the limit of quantitative measurement even after the 55-month testing period; consequently, the corrosion rates of steel plates could not be determined. It was found, however, that local corrosion proceeded to some extent near the 6.5-mm iron core wire used for the bent (Photo. 4-5), and that remarkable gap corrosion was observed around the rubber packing between the top plate and shell (Photo. 4-5).

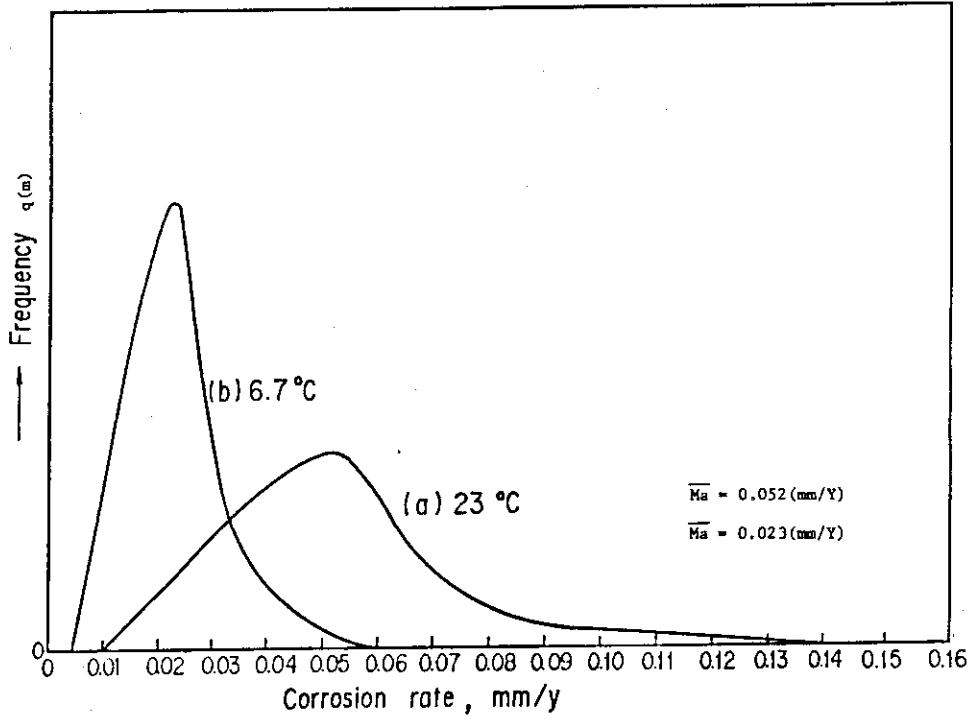


Fig. 5-8 Corrosion rate distributions of drum steel plates in seawater.

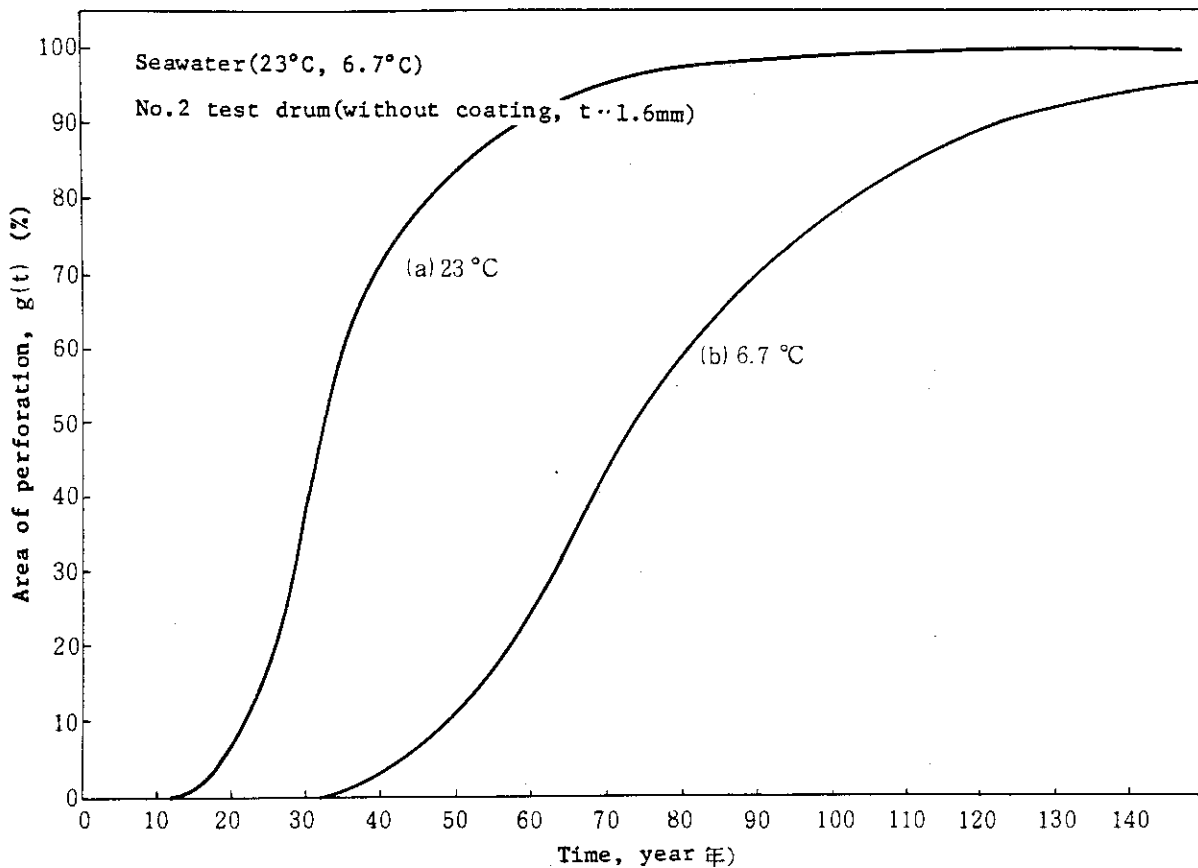


Fig. 5-9 Area of perforation of drum steel plates due to corrosion in seawater.

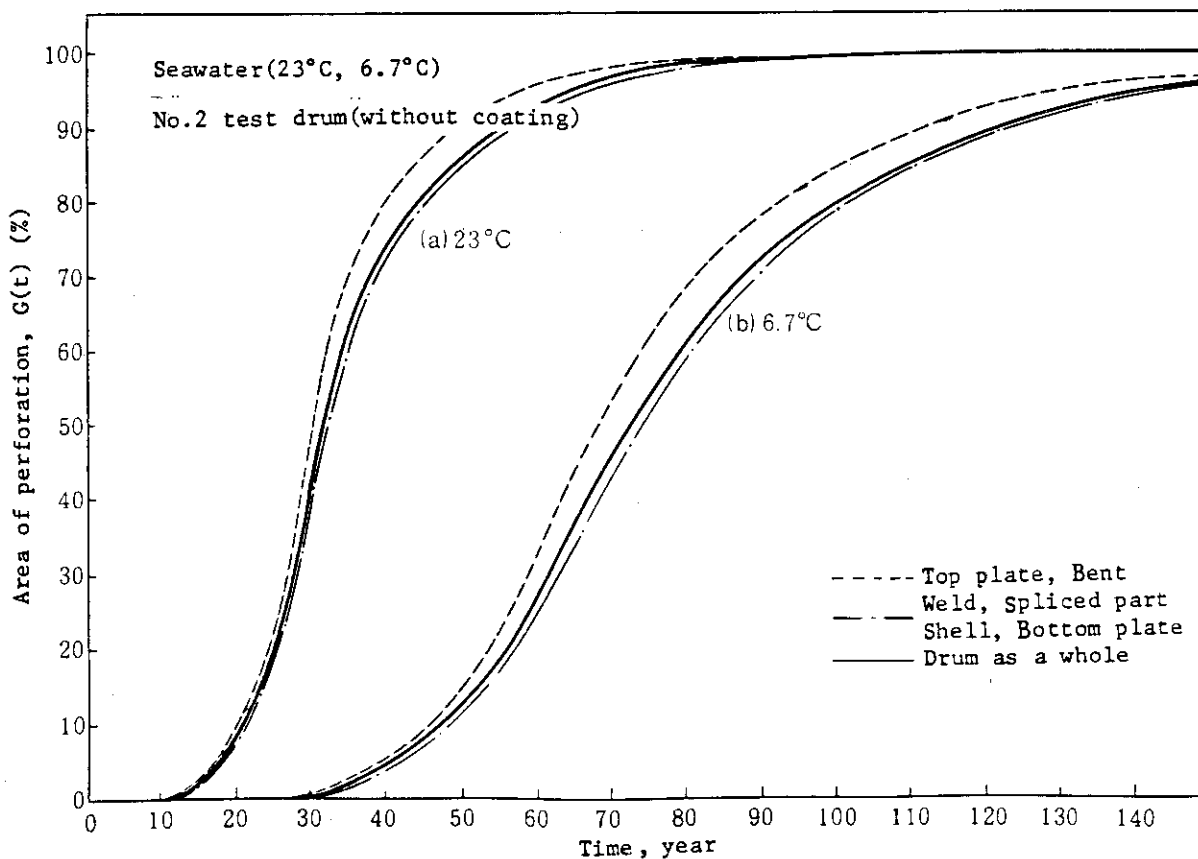


Fig. 5-10 Area of perforation of drums due to corrosion in seawater

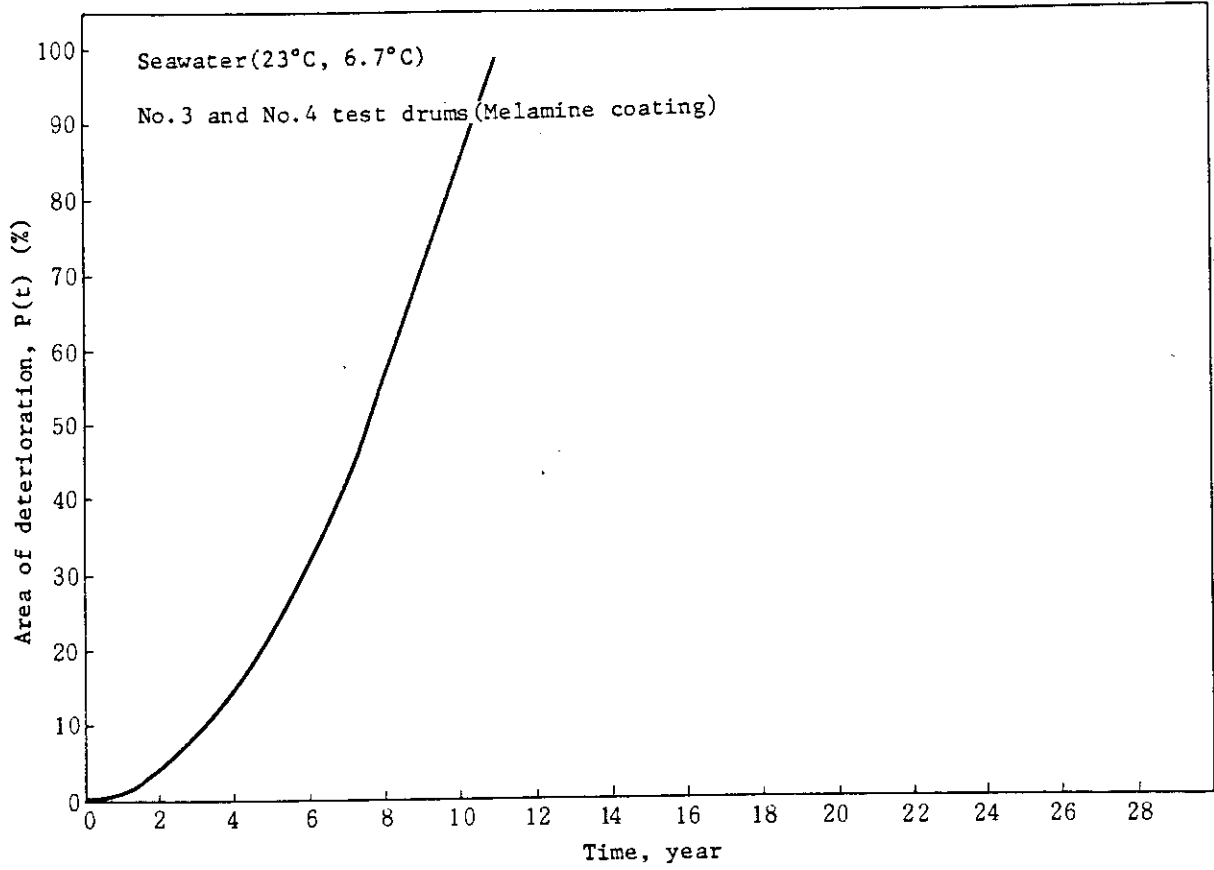


Fig. 5-11 Deteriorated area of coating of drums in seawater.

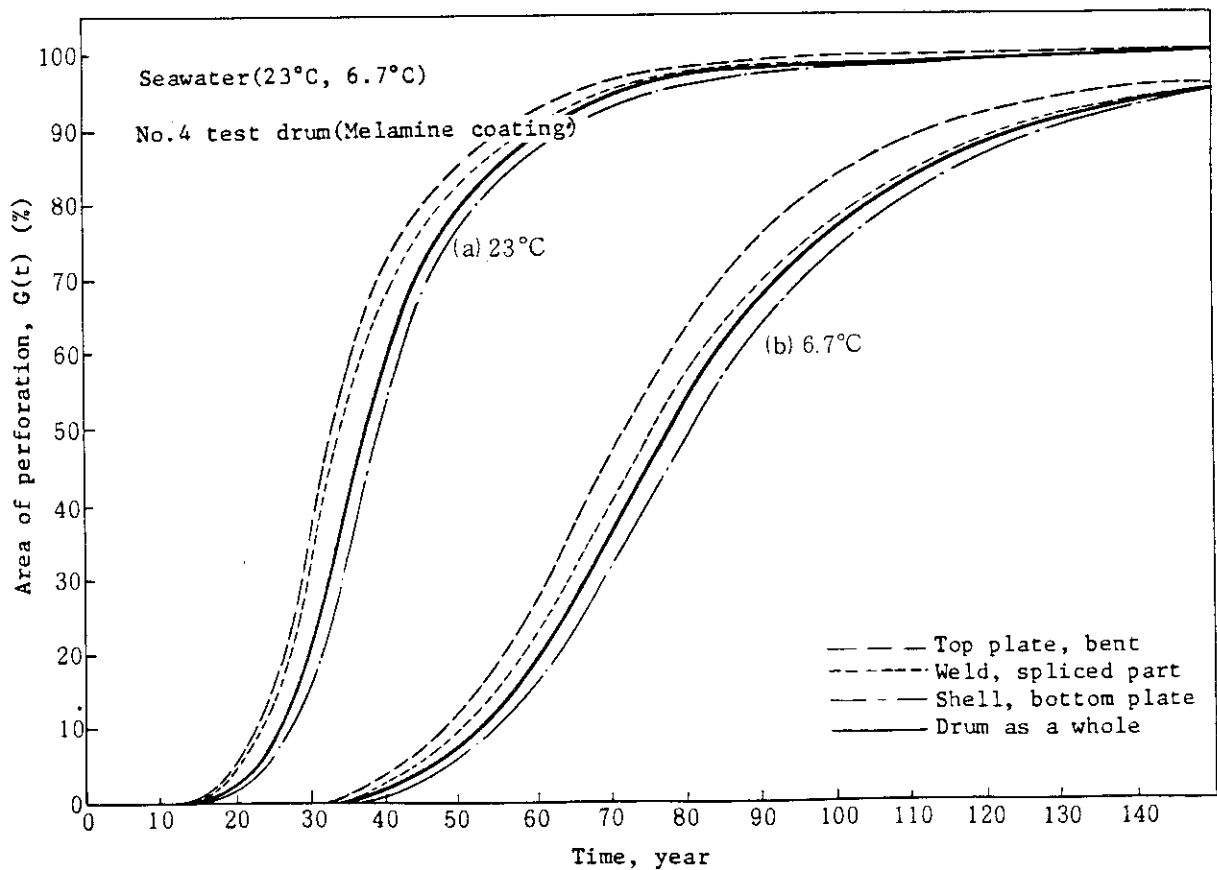


Fig. 5-12 Area of perforation of drums due to corrosion in seawater.

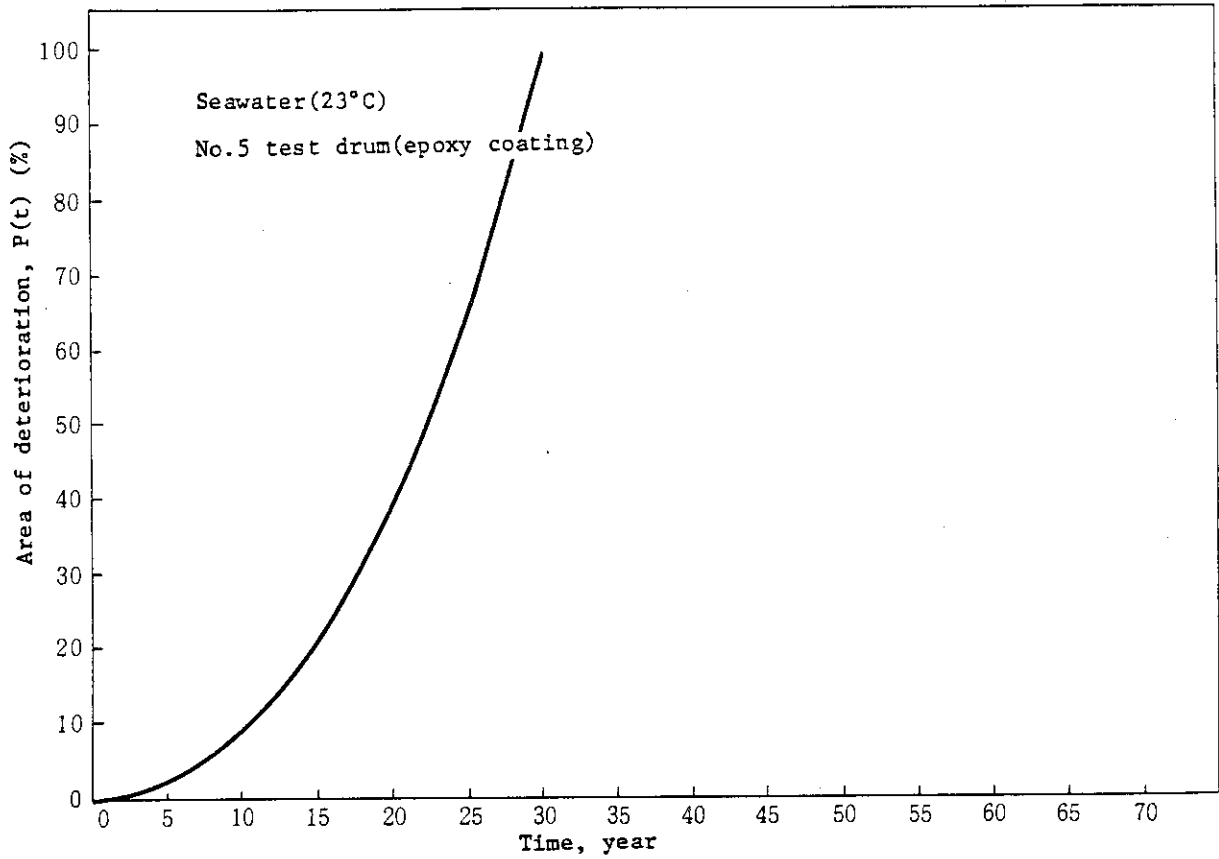


Fig. 5-13 Deterioration area of coating of drum in seawater.

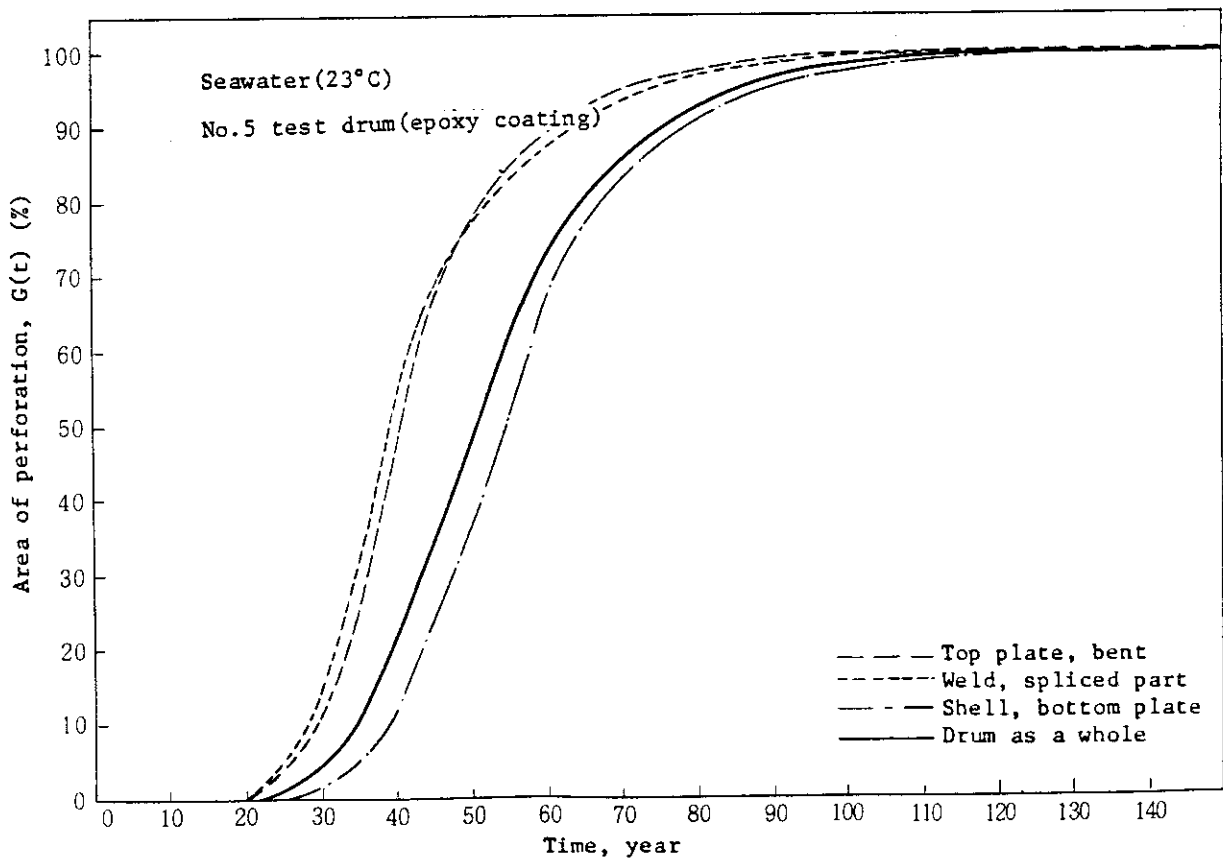


Fig. 5-14 Area of perforation of drum due to corrosion in seawater.

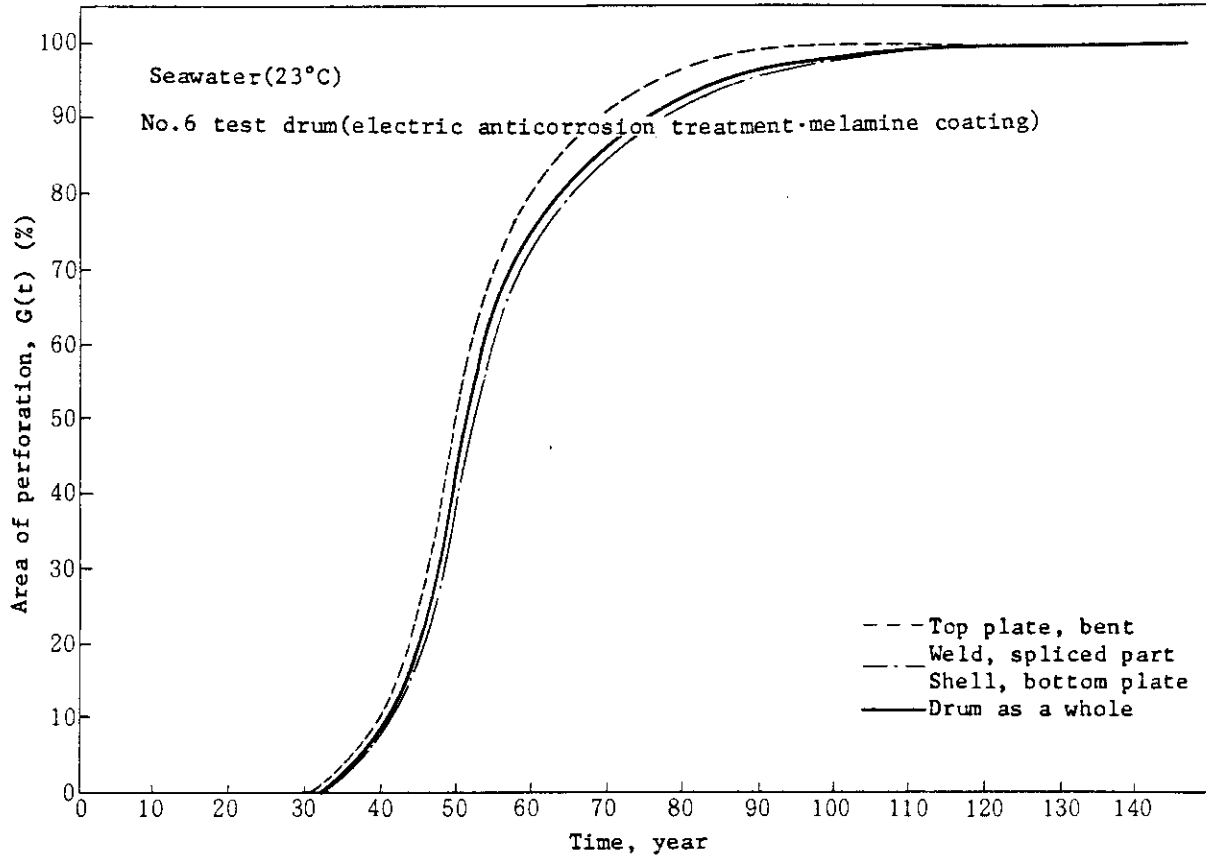


Fig. 5-15 Area of perforation of drum due to corrosion in seawater.

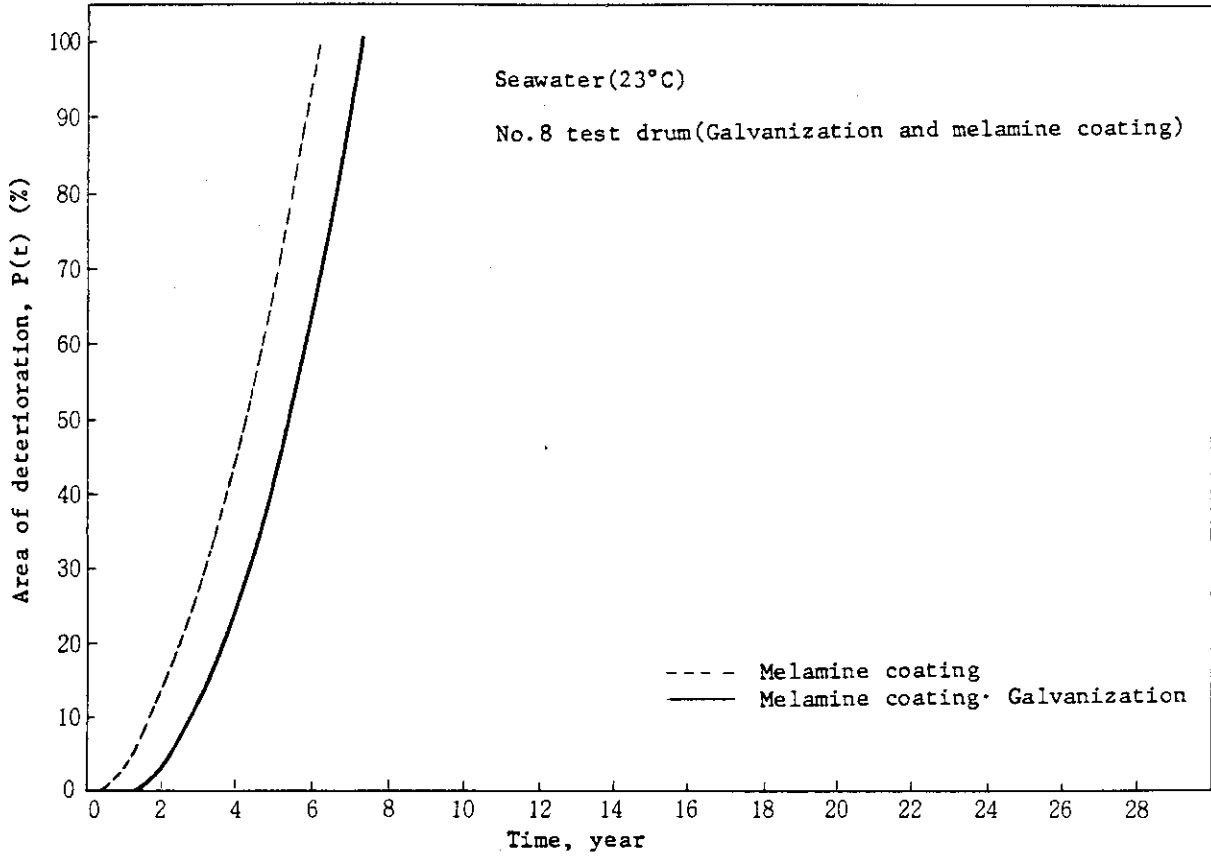


Fig. 5-16 Deterioration area of coating of drum in seawater.

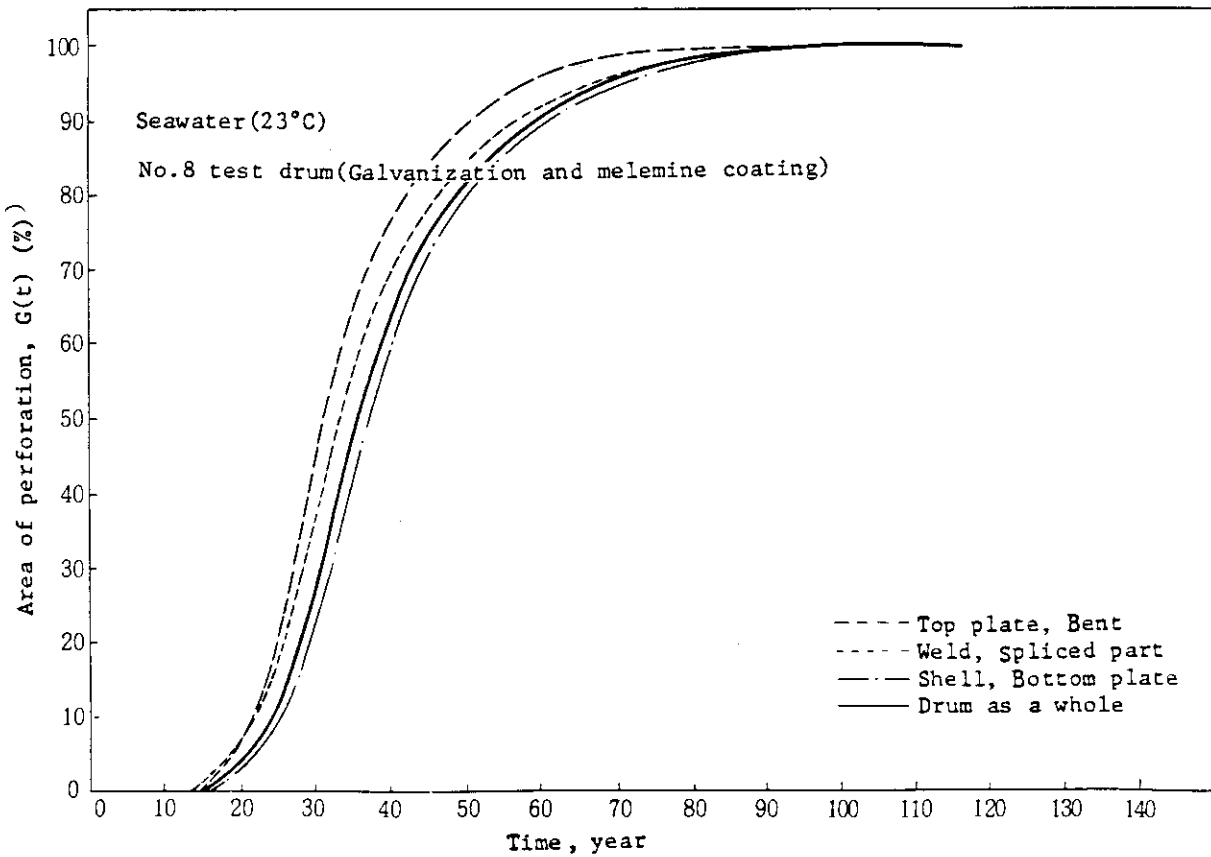


Fig. 5-17 Area of perforation of drum due to corrosion in seawater.

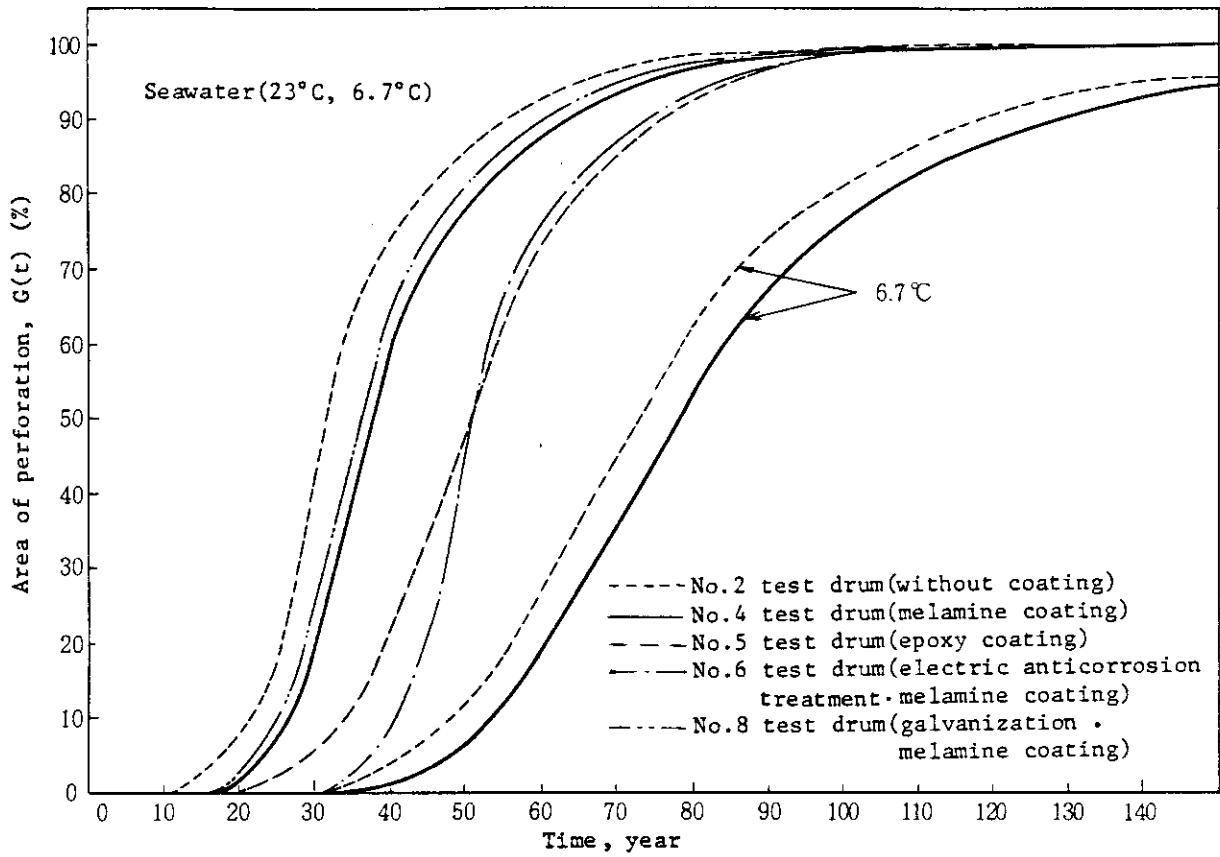


Fig. 5-18 Area of perforation of various drums due to corrosion in seawater.

5.2.2 Discussions

(1) Evaluation Accuracy

The evaluation results in the estimation of the long-term drum container's corrosion-through-hole-area-increasing curve $G(t)$ on the basis of only several years of testing, so that the accuracy of the evaluation should be discussed. The accuracy, therefore, is discussed hereafter concerning an ordinary drum container (melamine coating, $t \dots$ 1.6 mm).

1) Corrosion-through-hole-area-increasing Curves $g(t)$ s for Steel Plates

The drum container's corrosion-through-hole-area-increasing curve $G(t)$, as indicated in Eq. (3), can be obtained from both the steel plate's corrosion-through-hole-area-increasing curve $g(t)$ and the coating's deterioration-area-increasing curve $P(t)$, and $g(t)$ gives several times greater an influence on the results of $G(t)$ than $P(t)$ does (the durability of a steel plate was several times longer than that of a coating). Therefore, the estimation accuracy of $g(t)$ virtually determines that of $G(t)$. In addition, since $g(t)$ can be written by the integral of probability distribution $g(m)$ of the corrosion rate m on the steel plate as given by Eq. (2), the estimation accuracy of $g(m)$ eventually determines that of $G(t)$.

As shown in Figs 5-19 and 5-20, probability distributions $g_1(m)$, $g_2(m)$ and $g_1'(m)$ of the corrosion rates on the steel plate correspond respectively to the steel plate's corrosion-through-hole-area-ratio-increasing curves $g_1(t)$, $g_2(t)$ and $g_1'(t)$. As for $g(t)$, the corrosion

resistance of the steel plate is improved as $g(t)$ shifts in the right direction along the axis of abscissa; namely, average value \bar{m} of $q(m)$ directly dominates the evaluation of $g(t)$. With regard to average corrosion rate \bar{m} of the steel plate, the following two kinds of values are employed and the estimation accuracy of \bar{m} was excellent because of a comparatively greater amount of data; one set of data was obtained from the weight loss measurement for non-coated drum container's shells after the 55-month corrosion tests (exclusive of 6.7-°C seawater environment), and the other set of data was obtained from that for test pieces. To use the time-averaged corrosion rate during this testing period for a long-term evaluation tended to overestimate corrosion rates, in other words, to estimate them conservatively concerning the safety evaluation (Corrosion rates gradually decrease as time passed.). Although the "form" of probability distribution $g(m)$ of corrosion rate m over the steel plate was obtained from the results of fixed-point thickness-distribution measurement for the non-coated container shell after the 55-month corrosion tests, the long-term change in the "form" of the distribution cannot be estimated at present. As shown in Figs 5-19 and 5-20, however, the "form" of $q(m)$ had an effect only on the "form" of $g(t)$, which was not so important because the essential role of the drum container for radioactive waste was determined by where $g(t)$ moved in the direction of t by value \bar{m} . of course, the "form" of $g(m)$ becomes very problematical if the local corrosion itself of the drum container was to be evaluated (for example, when corrosion through-hole occurred on the drum container), such a maximum corrosion rate was not evaluated here.

The above discussions suggest that $g(t)$ estimated in this paper was relatively high in its accuracy, and that the corrosion rate was not

underestimated, compared with at least actual $g(t)$.

2) Coating's Deterioration-area-increasing Curve $P(t)$

Although the coating's deterioration-area-increasing curve $P(t)$ showing its durability was obtained on the basis of the secular measurement of the rust areas generated over coatings during the 55-month testing period, $P(t)$ was so determined as to overestimate the deterioration of the coating. Here, the rust area was looked upon as the steel-plate-protecting-function-lost area; namely, in this evaluation, even if blisters occurred on the surface of a coating, it was looked upon as having a steel-plate-protecting function if no rust was generated. For radioactive waste which was disposed of in a stationary state and not subjected to an external force, the blister of a coating did not always mean spallation. The test results clarify that if a simple coating blister contains sea or land water in it, it does not proceed to steel plate corrosion at once because of a lack of dissolved oxygen.

(2) Corrosion Evaluation for Internal Surfaces of Drum Containers

1) Corrosion Rates of Steel Plates

The top plate and shell of the drum container are clamped by the band through a packing, and, therefore, sea water hardly intrude into the container. In seawater disposal, however, seawater may sometimes intrude into the drum container, so seawater intrusion was assumed in this evaluation. According to the test results of this case, the corrosion rate of the internal surface of the steel plate was 1/50 or less of that of the external surface (See Table 4-18), so that, for the purpose of evaluation, the corrosion rate of the steel-plate's internal surface which was not in contact with the simulated cemented waste was assumed to be 1/10 of that of the external surface. Since no corrosion was observed on the steel-plate's internal surface which was in contact with the cemented waste, the corrosion rate was assumed to be zero upon evaluation.

In all the cases, the corrosion rates of the steel-plate's internal surfaces were assumed to be smaller than those of the external surfaces, but these corrosion rates originated from the results of the corrosion tests, and so the corrosion rates were not underestimated from the viewpoint of the overall corrosion-resistance evaluation.

2) Durabilities of Coatings

According to the visual observation of disassembled drum containers after the corrosion tests, the deterioration of internal coatings were smaller than that of the external ones. In particular, no deterioration of coatings were recognized for their parts which were in contact with the cemented waste. However, since the corrosion rates of the steel-plate's internal surfaces were satisfactorily smaller than those of the external surfaces and were 0 to 1/5 of the latter, the durability of the internal coating was assumed to be equal to that of the external one.

(3) Evaluation of High-draft Special Parts

As for the bent, weld and spliced part to which high draft is applied, the adhesion of the coatings and steel plates seemed to be poor, resulting in 2 to 3 times greater coating deterioration in most parts of the container (exclusive of the shell) than that in the shell for all the environments. No difference was found, however, between the corrosion resistance of the shell steel plate and those of other parts of the container (See 4.1.6). The effects of these special parts on the corrosion resistance of the drum container as a whole were smaller.

(4) Evaluation of the Bands

The thickness of the steel band was 2 mm, and a little thicker than those of the other parts of the drum container which were usually 1.6 mm. Since the band was not coated (Despite of the presence of a 5-to 6- μ galvanized layer, it was dissolved by seawater in about 3 months.), its

corrosion resistance as a whole will be nearly equal to that of the shell. According to the results of about 5 year corrosion tests, the top plate and shell were so strongly adhere to each other that it was necessary to remove them using tools. Therefore, the role of the band for the drum container is filled when the container reaches a sea bottom, and there is no need of a corrosion-resistance evaluation for it after being disposed of.

(5) Evaluation of Differences between Individual Drum Containers

Since the drum containers for low-level radioactive waste had been mass-produced, little dispersion was found between individual drum containers, so the differences between individual drum containers may not be problematic as a whole. In addition, the individual differences may be smaller than the overestimated corrosion amounts in the long-term evaluation; consequently, the differences in corrosion resistances between individual drum containers, except for specially poor-quality products, may be ignored.

5.2.3 Conclusions

The results of corrosion-resistance evaluation for various drum containers under a seawater environment (Fig. 5-18) can be summarized as follows;

1) Ordinary Coated Drum Containers

- ① As for No. 4 test bodies (melamine coating) in 23 and 6.7 °C seawater, the times when their corrosion-through-hole-area ratios reach 1 % will be about 18 and 38 years, respectively, and further, those when the ratios reach 10 % will be about 26 and 53 years, respectively.
- ② The durability of melamine coatings is small, and is about 1/4 and 1/10 of that of 1.6-mm-thick steel plate in 23- and 6.7-°C seawater

(comparison of the times when the coating's steel-plate-protecting-function-lost area ratio and corrosion-through-hole-area ratio reach 50 %); consequently, flaws of the coatings are not necessarily a big defect.

③ The durabilities of the coatings covering the weld, bent and spliced part are small, and correspond to 1/2 to 1/3 of the shell, but no significant differences are observed in steel-plate corrosion during the testing period between all the parts of the drum containers.

④ If seawater seeps into the drum container, the corrosion from its internal surface is extremely short, corresponding to 1/50 or less of that from the external surface.

2) Anticorrosion-treated Drum containers

① The durability of the epoxy-resin coating in combination with the zinc-bearing primer which are applied to No. 5 test body is longer, and will be 3 times or more as long as the melamine coating applied to No. 4 test body.

② The zinc-utilized electric anticorrosion treatment applied to No. 6 test body is effective, and its most outstanding advantage is to be able to calculate its anticorrosion effect in advance, enabling a suitable amount of anticorrosive zinc necessary for an expected durability to be applied to the drum containers. However, the zinc-bearing primer and electric anticorrosion treatment are inadequate for melamine coatings because of their greater spallation.

③ With regard to the galvanization and melamine coating applied to No. 8 test body, the adhesion of a zinc layer and melamine coating is poor, leading there by to a great deal of melamine coating spallation; in other words, this combination does not permit the galvanized zinc

and melamine coating to show their anti-corrosion effect fully. When the zinc-galvanized steel plate is to be used after being coated, the kind of coating and the method of coating should be fully investigated.

④ The corrosion amount of stainless steel plate, exclusive of part of a drum container, could not be quantitatively measured after the 55-month corrosion test, indicating a great corrosion resistance. However, corrosion-through-holes appear locally because of the gap corrosion occurring in a part of the stainless steel drum container with which the rubber packing is in contact. Since the steel wire is used for the bent as the core wire, remarkable corrosion is found in and around the core wire, necessitating countermeasures to cope with these problems when the stainless steel drum containers are to be used.

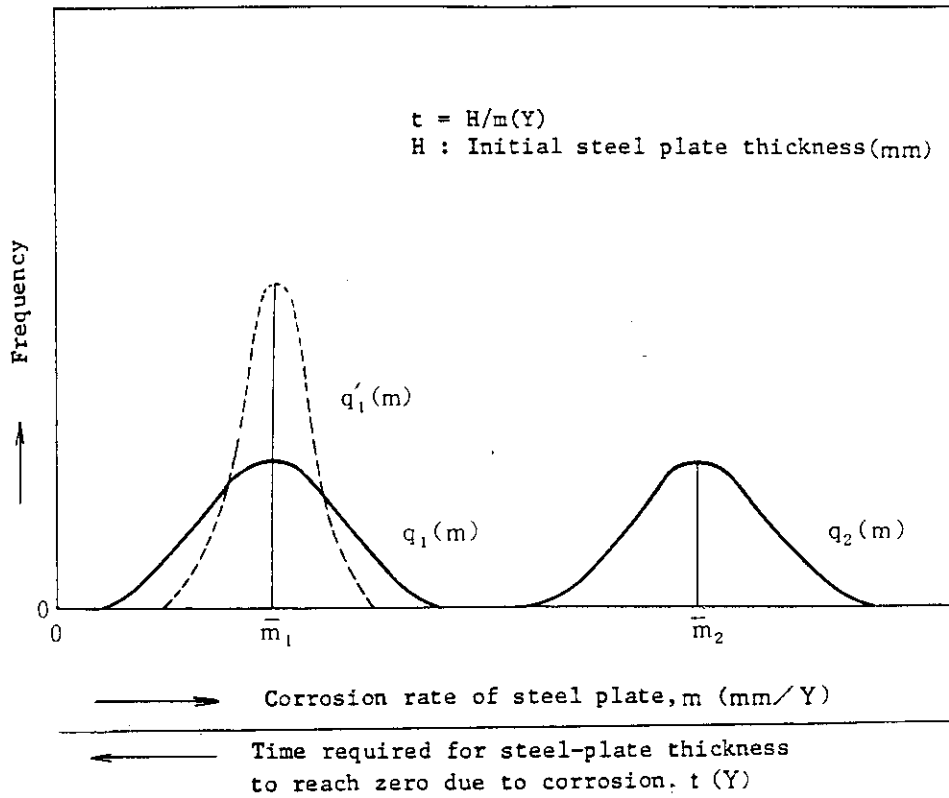


Fig. 5-19 Probability distribution of corrosion rates on the same steel plate.

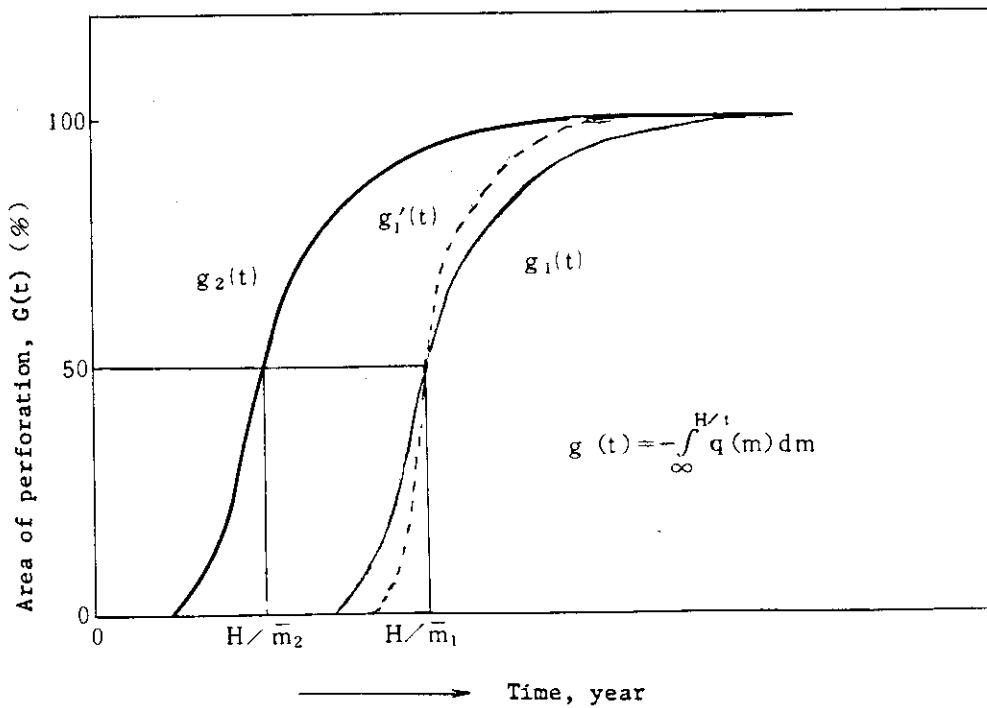


Fig. 5-20 Area of perforation of drum steel plate due to corrosion in seawater.

6. Postface

The "Corrosion Safety Demonstration Tests" started in November 1978 to clarify the process of corrosion under various environments of low-level radioactive waste containers — the drum containers enclosing cemented waste — and to demonstrate their great capacity of reducing the release of radioactive nuclides into the environment of the time of waste disposal. Since then tests have been performed for about 5 years to complete the "final report". This paper is part of the "final report", describing the corrosion tests under a seawater environment. Since the method of and the criteria for ocean disposal has already been specified, the disposal environment was easily thought out, corresponding to a series of 6.7 °C tests under a seawater environment in this paper. The temperature of the actual sea-disposal environments was considered to be 2 °C, so that the test results obtained at 6.7 °C will give conservative corrosion resistances.

Figure 5-18 gives the test results, in which all the drum containers showed greater corrosion resistances, achieving the aim expected initially.

Finally, the authors would like to express hearty thanks to the members of the Corrosion Safety Demonstration Test Committee headed by Prof. Kiyose and the officers concerned of the Science and Technology Agency for the wonderful guidance and encouragement, together with the persons concerned inside and outside of JAERI for their skill and cooperation.

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Appendix A Safety Evaluation of Packages of Low-Level Radioactive
Waste for Ocean Disposal

The present environmental exposure evaluation for the ocean disposal of low-level radioactive waste was carried and conclusions were drawn on the assumption that all the radioactive nuclides were released into the seawater as soon as the dumped waste reached the bottom of the sea; in other words, the conclusion that the packages were safe was drawn without taking into consideration the barrier capacity of the packages (cemented waste plus drum container). The packages, however, were provided with a barrier capacity against the release of radioactive nuclides into the environment; accordingly, the evaluation of the barrier capacity will improve the accuracy of the results of the safety evaluation completed already for ocean disposal. In the ocean disposal of low-level radioactive waste, the waste shall be solidified in cement in such a way as to have a given specific weight and uniaxial compressive strength, and will be disposed of at a sea bottom depth of 5.000 m or more. Figure A-1 gives the safety evaluation items necessary for environmental exposure for this case.

This paper provides an evaluation method for the barrier capacity — the capacity of preventing radioactive nuclides from being released into seawater — of a waste package in ocean disposal, and thereby evaluated their barrier capacities by using results of "immersion tests for cemented waste" and "corrosion tests for drum containers".

1. Evaluation Methods

Figure A-2 schematically shows the secular change in the amounts of leach-out radionuclides into seawater when low-level

radioactive packages are disposed of at sea. Curve (1), $f(t)e^{-\lambda t}$, represents the secular change in leached-out rates of the radioactive nuclides from the cemented waste into seawater, curve (2), $G(t)$, that in the corrosion-through-hole-area ratios of the drum container, and curve (3), $f'(t)e^{-\lambda t}$, that in the leached-out rates of the radioactive nuclides from the waste package including the drum container. Curves (4), $F(t)$, and (5), $F'(t)$, show the secular changes in the leached-out amounts of radioactive nuclides in the cemented waste without and with the enclosing drum container, respectively, with the leached-out amount written in terms of the leached-out ratio (ratio of the amount of the leached-out radioactive nuclides up to time t to the total amount of the ones which existed initially in the cemented waste). They can be written as follows:

$$F(t) = \int_0^t f(t)e^{-\lambda t} dt \quad \dots (1)$$

$$F'(t) = \int_0^t f'(t)e^{-\lambda t} dt \quad \dots (2)$$

If R and Q are defined as Eq.s (3) and (4), respectively, R , Q and PQ represent the reduction rates in the radionuclide-releasing amount of the cemented waste itself, the drum container and of the package, respectively; namely, these three give the barrier capacities of the cemented waste, drum container and of the package against the release of radioactive nuclides into the environment.

$$\lim_{t \rightarrow \infty} F(t) \equiv R \quad \dots (3)$$

$$\lim_{t \rightarrow \infty} [F'(t) / F(t)] \equiv Q \quad \dots (4)$$

To calculate the reduction rate RQ of the radionuclide-

releasing amount from the packages, therefore, the leached-out rate of the radioactive nuclides from the packages, $f'(t)e^{-\lambda t}$, (system consisting of a drum container and cemented waste) must be first obtained. To calculate $f'(t)e^{-\lambda t}$ in the case of a drum container in which a corrosion-through-hole generates and grows, the following model is assumed for the process in which the radioactive nuclides are leached out of the surface of the cemented waste into seawater, resulting in a decreased in the radioactive nuclide concentration.

Figure A-3-(1) shows a hypothetical process in which the radionuclide-concentration change on the surface of the cemented waste will actually follow when the corrosion-through-hole area grows. Figure A-3-(2), on the other hand, shows a hypothetical model to make the calculation of $f'(t)e^{-\lambda t}$ easier, and assumes that as soon as a through-hole increment appears in the course of its growth on the drum container, the radionuclide concentration there becomes equal to the initial concentration over the surface of the cemented waste; namely, no movement of the radionuclides is assumed except in the direction perpendicular to the surface of the cemented waste. Under these conditions, $f'(t)$ is represented by Eq. (5) as a convolution integral of the differential values of $G(\tau)$ from 0 to 1 (the increment of $G(\tau)$) and $f(t-\tau)$.

$$f'(t) = \int_c^b f(t-\tau) \frac{d}{d\tau} G(\tau) d\tau \quad \dots (5)$$

where b means the time required for the corrosion-through-hole-area ratio to reach 1. In addition, $G(t)$ can be differentiated, and

$$0 \leq \frac{d}{dt} G(t)$$

$$0 \leq G(t) \leq 1$$

2. Results of Evaluation

By using the method described in 1, a calculation was carried out on the barrier capacities against radionuclide release of cemented waste itself R, drum container Q and of package RQ in the ocean disposal of low-level radioactive waste.

The cemented waste with 150-kg/cm^2 uniaxial compressive strength was evaluated as a solidified waste, while the 200- ℓ , 1.6-mm-thick, melamine-coated open-type steel drum as a drum container. As for radioactive nuclides, ^{60}Co , ^{90}Sr and ^{137}Cs were chosen for the evaluation (According to the estimation¹⁾ of the amount of the environmental exposure due to radioactive waste to undergo ocean disposal, 90 % of it is ascribed to the three nuclides.), and the values²⁾, which are estimated from the results of the seawater leaching tests for radionuclides contained in the cemented waste, are used for $f(t)$ which gives the secular change in the leached-out rates of the radionuclides from the cemented waste in seawater. In other words, $f_{\text{Co}}(t)$, $f_{\text{Sr}}(t)$ and $f_{\text{Cs}}(t)$, which represent the secular changes in the leached-out rates of ^{60}Co , ^{90}Sr and ^{137}Cs contained in the cemented waste, respectively, are determined as follows:

$$f_{\text{Co}}(t) = 3.6 \times 10^{-5} \frac{1}{\sqrt{t}} \quad \dots (6)$$

$$f_{\text{Sr}}(t) = 4.0 \times 10^{-4} \frac{1}{\sqrt{t}} \quad \dots (7)$$

$$f_{\text{Cs}}(t) = 1.3 \times 10^{-2} \frac{1}{\sqrt{t}} \quad \dots (8)$$

where the unit for t is year.

In addition, curve $G(t)$ which was estimated in $6.7\text{-}^\circ\text{C}$ seawater and is shown in Figure A-4 is employed as corrosion-through-hole-area-increasing curve $G(t)$ for the drum container under the disposal environment.

Substitution of $f_{Co}(t)$ defined by Eq. (6) into Eq. (1) leads to the leached-out ratio of ^{60}Co , $F_{Co}(t)$, from the cemented waste immersed in seawater as shown by the dotted line in Figure A-5. In addition the solid line in the figure shows $F'_{Co}(t)$ calculated from Eq.s (1) and (5), together with from $G(t)$ in Fig. 4. From the resultants $F_{Co}(t)$ and $F'_{Co}(t)$, the reduction rate of the amount of ^{60}Co released from the cemented waste, drum container and package are $R_{Co} = 1.5 \times 10^{-4}$, $Q_{Co} = 3.5 \times 10^{-4}$ and $R_{Co}Q_{Co} = 5.2 \times 10^{-8}$, respectively.

Likewise, the secular changes in the leached-out ratios of ^{90}Sr and ^{137}Cs from the 200-l cemented waste buckets be obtained by using $f_{Sr}(t)$ defined by Eq. (7), $f_{Cs}(t)$ defined by Eq. (8), $G(t)$ in Figure A-4 and Eq. (5); the resultant $F_{Sr}(t)$ and $F_{Cs}(t)$ for the cemented waste without the drum container, and $F'_{Sr}(t)$ $F'_{Cs}(t)$ for one with the drum container are shown by the dotted lines (2) and (3), and by the solid lines (2) and (3) in Fig. 5. Table 1 shows the results of calculation on the releasing-amount reduction rates R_{is} , Q_{is} and R_iQ_{is} for ^{60}Co , ^{90}Sr and ^{137}Cs by the cemented waste itself, drum container and package which are obtained on the basis of Figure A-5.

3. Conclusions

The authors proposed an evaluation method for the barrier capacities of packages (including cemented waste and drum containers) against the release of radioactive nuclides from the radioactive cemented waste into the environment at the time of ocean disposal, and thereby evaluated their barrier capacities for example by using results of "immersion tests for cemented waste" and "corrosion tests for drum containers". When test data was insufficient, calculation was carried out so that the leached-out amounts might be some what overestimated.

The barrier capacities of the packages vary with the kinds of radioactive nuclides. As indicated in Table A-1, the barrier effect of the package is relatively small for high-leached-out-rate, long-half-life ^{137}Cs , but great for low-leached-out-rate, short-half-life ^{60}Co .

Sofar, upon evaluating the environmental exposure due to ocean disposal of radioactive waste, the barrier effect of the packages has been looked upon as zero, and it has been therefore to be safe. To take into account the barrier effect of the packages, therefore, allows the evaluation of environmental exposure to shift greatly to the safety side.

Finally, the main objective of this paper is to clarify the evaluation philosophy and method for low-level radio-active waste disposal. The authors, therefore, would greatly appreciate it if they could receive some constructive comments on the evaluation method rather than on the results.

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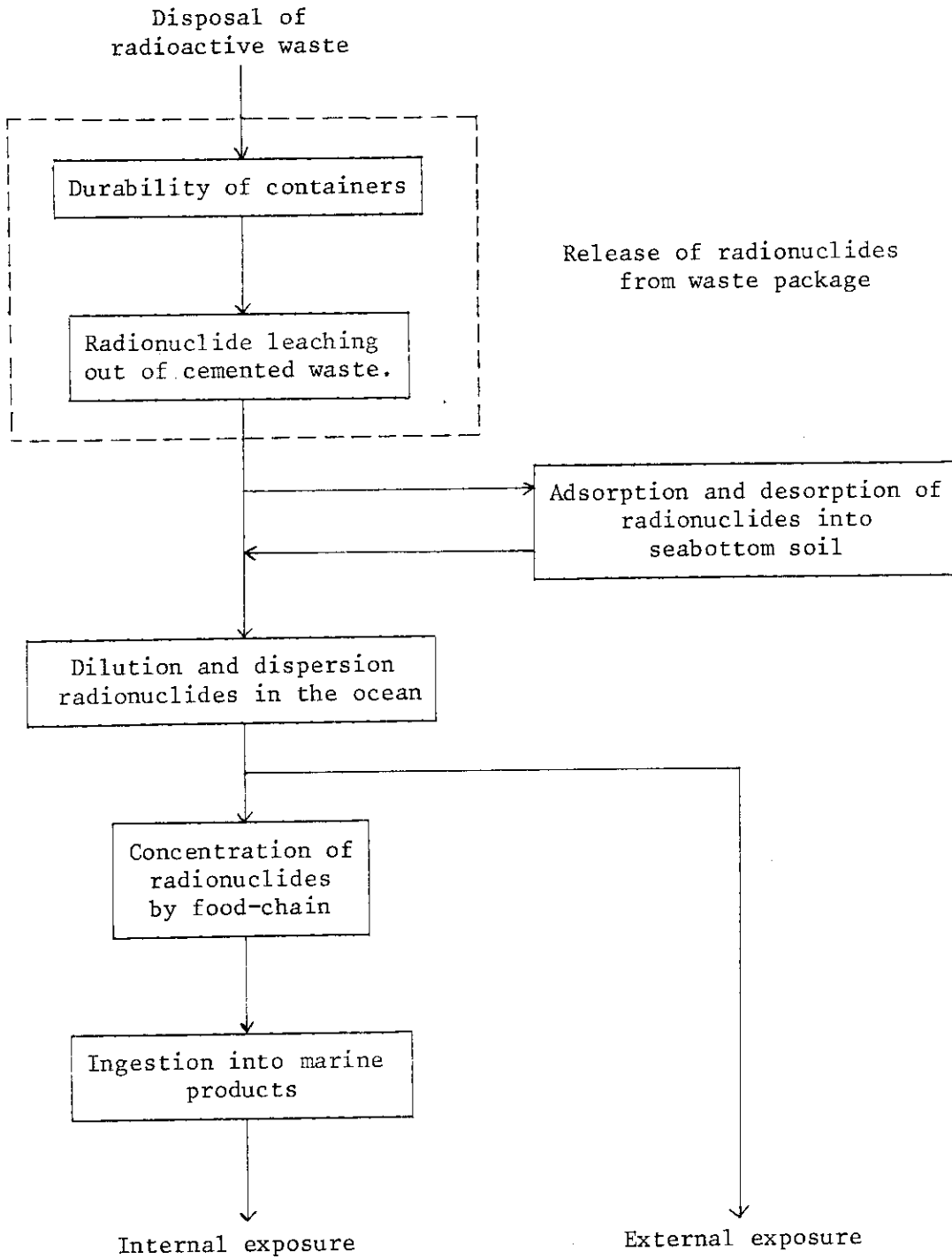


Fig. A-1 Safety evaluation items for environmental exposure due to low-level radioactive waste during ocean disposal.

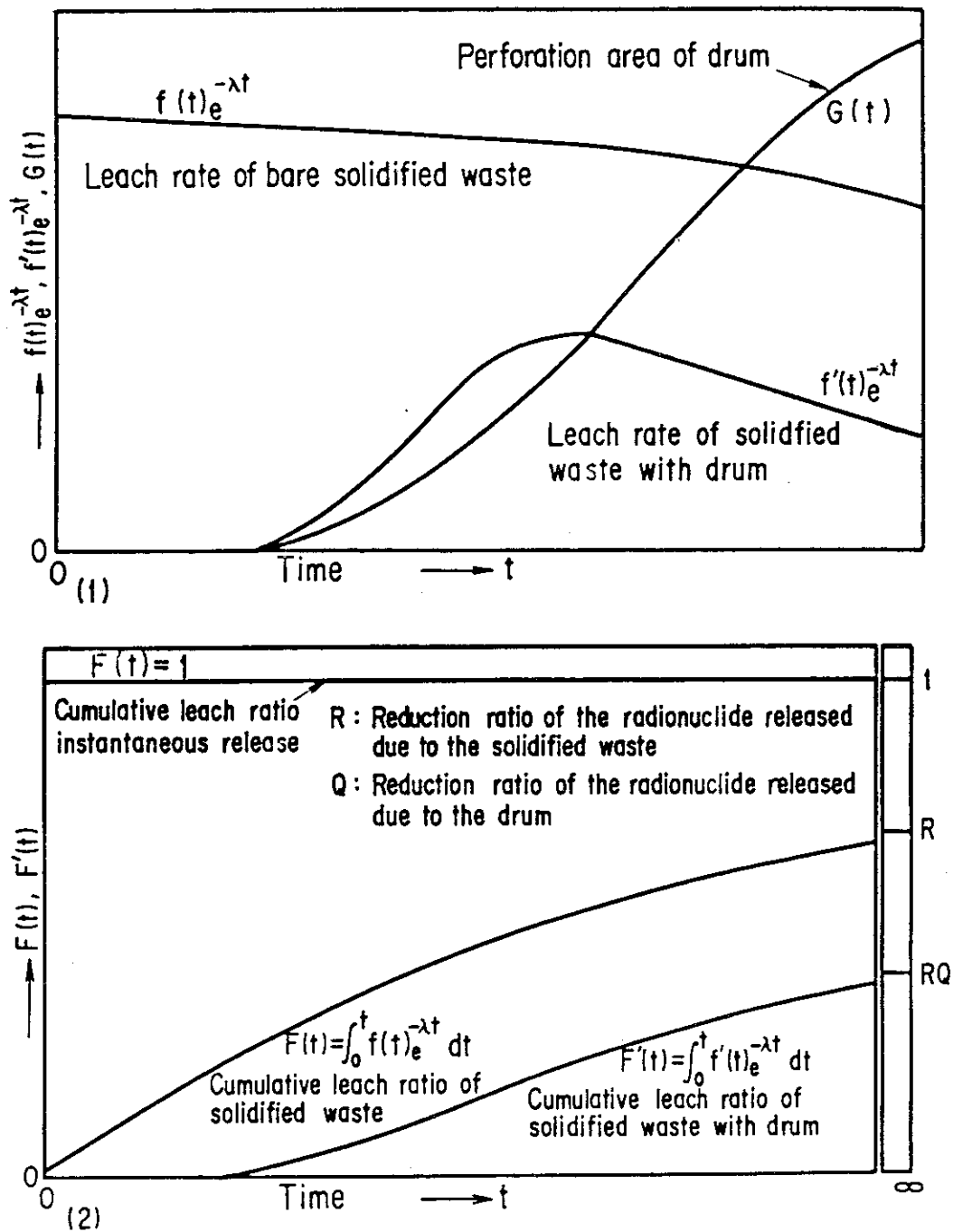
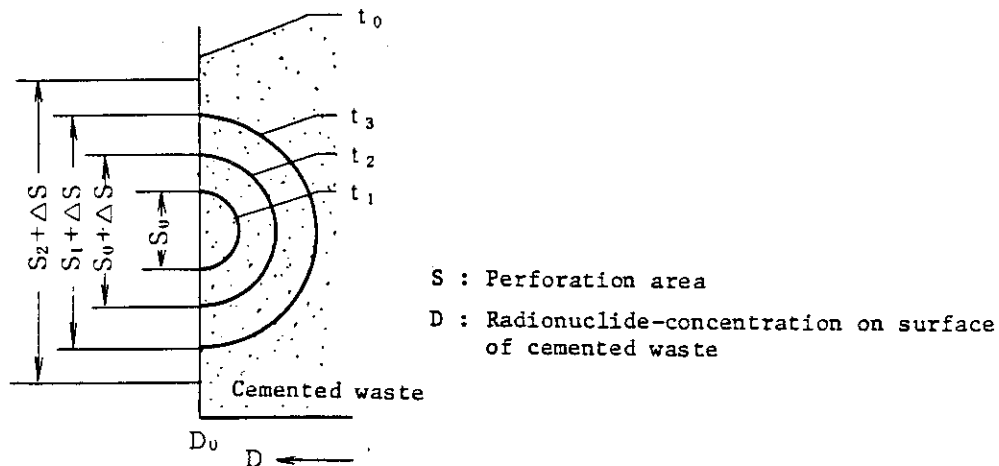
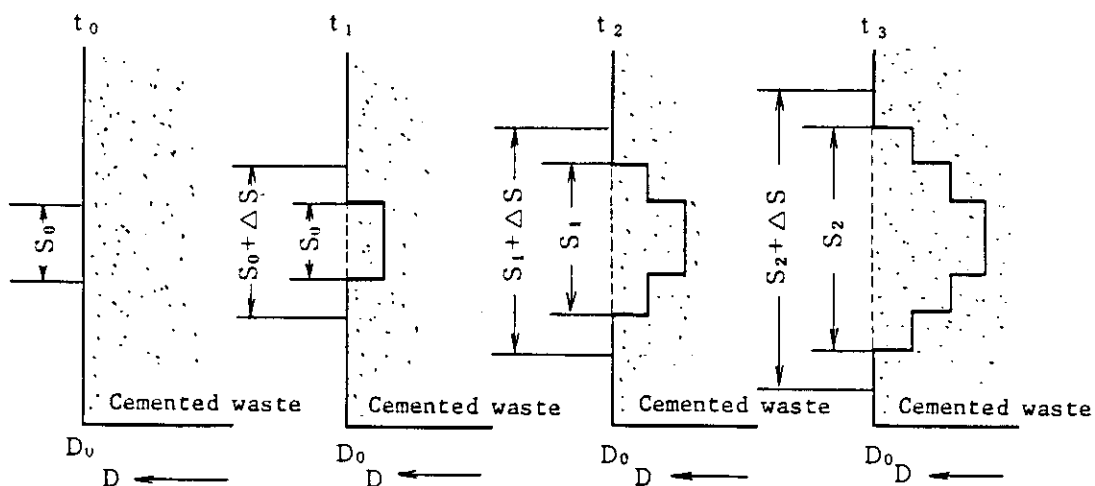


Fig. A-2 Schematic release curves of radionuclides in the case of ocean disposal of low-level radioactive wastes.



(1) Hypothetical actual corrosion process



(2) Corrosion process in which the radionuclide-concentration in the perforation area increment ΔS is assumed to be always equal to the initial concentration D_0

Fig. A-3 Schematic radionuclide-concentration change in the plate where perforation area of drum due to corrosion is increasing.

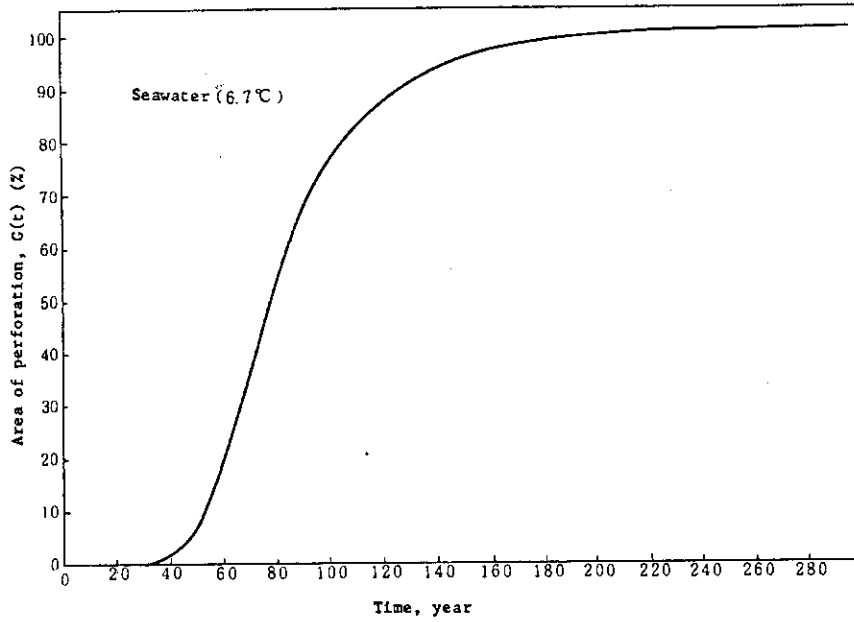


Fig. A-4 Secular perforation area of H-class drum due to corrosion in seawater.

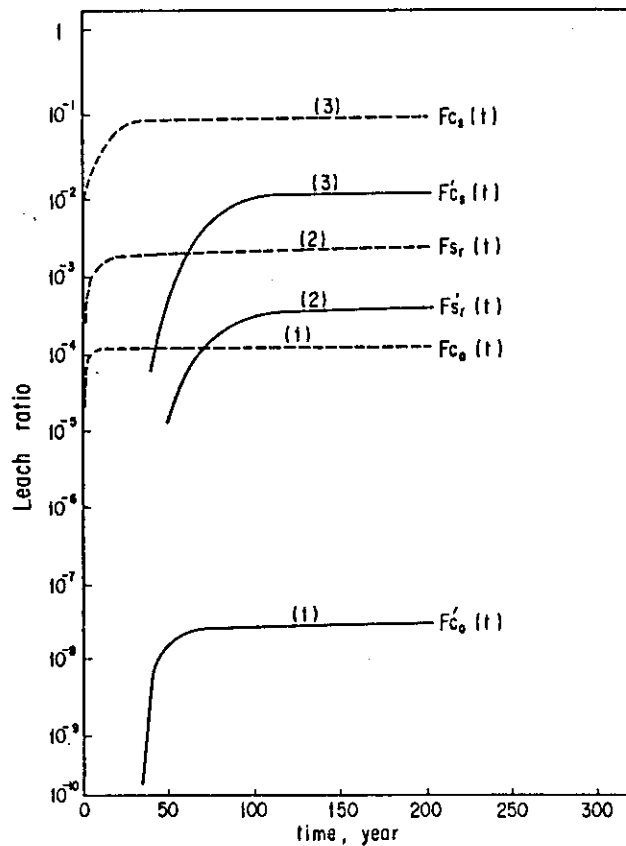


Fig. A-5 Cumulative leach ratio of radionuclides from 200ℓ cement-solidified waste without and with drum calculated using values in Figure A-4.

Table A-1 Reduction rate of release radionuclides contained in 200-1 cemented waste due to "cemented waste itself" "drum" and "package".

Radionuclide	Half-life (Year)	Reduction rate due to cemented waste itself R_I	Reduction rate due to drum Q_I	Reduction rate due to package $R_I Q_I$
Co-60	5.27	1.5×10^{-4}	3.5×10^{-4}	5.2×10^{-8}
Sr-90	28.5	4.3×10^{-3}	1.5×10^{-1}	6.4×10^{-4}
Cs-137	30.0	1.3×10^{-1}	1.2×10^{-1}	1.6×10^{-1}