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THE STUDY ON THE CORROSION OF POOL COMPONENTS BY THE EVALUATION OF MICRO-ELEMENTS VARIATION IN FUEL STORAGE WATER, 1983

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Abstract

As a part of the IAEA coordinated programme on BEFAST, behavior of spent fuel assemblies during extended storage, corrosion of pool components has been investigated. In this study the concentrations of micro-elements of corrosion products (Fe, Cr, Ni) in pool water have been periodically measured and the evaluation of corrosion has been made from the results of analysis. The concentrations of micro-elements of corrosion products (Fe, Cr, Ni) was almost constant during this study. Corrosion rate was estimated to be only a little as compared with the pool lining in thickness of 3 mm (the side) or 4 mm (the base).

Contents

1. Introduction

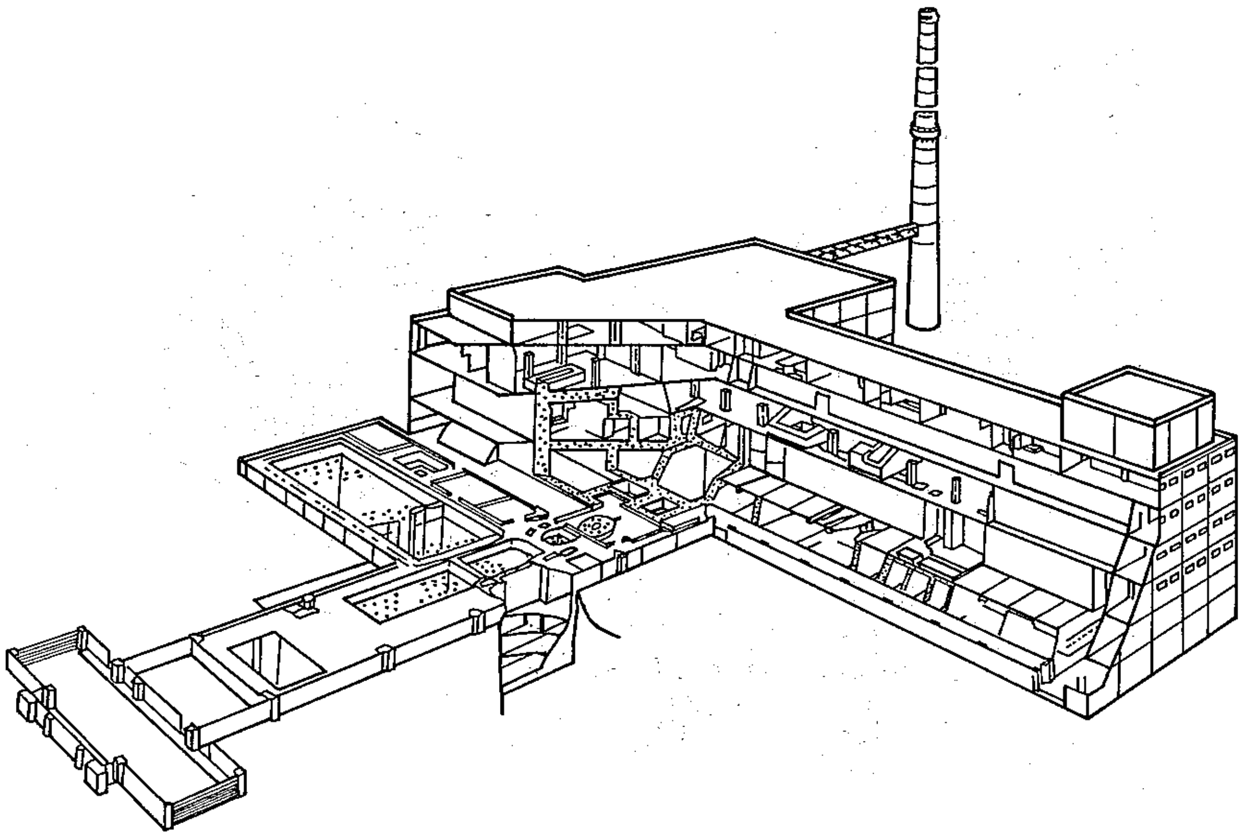
IAEA Coordinated Research Programme on BEFAST is aiming to provide statistically significant data for extended storage of spent fuels. As a part of this Programme, this study has been carried out utilizing the fuel storage pool of PNC Tokai reprocessing plant. This plant has been in hot operation since 1977, and 255 tU of spent fuels have been received and stored in the fuel storage pool for reprocessing. Based on some experience on receiving and storage technique, it is recognized to be important to keep the integrity of pool components in long term to ensure the safe storage of spent fuels. In this context, corrosion of pool components has been investigated on the basis of the quantitative way to make clear the problems for extended storage. In this study, as the concentration of micro-elements of corrosion products (Fe, Cr, Ni) in pool water is considered to be a factor which relates directly to corrosion of pool components, then the measurements have been periodically performed, and the evaluation of corrosion has been made from the results of analysis.

2. Spent fuel receiving and storage facility

The spent fuel receiving and storage facility is located on the East side of the main plant as shown in Fig.1. The facility has three principal pools, which are cask unloading pool, fuel storage pool and fuel transfer pool. Those are separated by the tight doors to avoid mixing the fuel storage pool water with other pools. It has also two other pools which are preparatory. The layout of the facility is given in Fig.2 and Fig.3. The dimension of the fuel storage pool is 30 m in length, 10 m in width, 10 m in height and 9 m in depth of water.

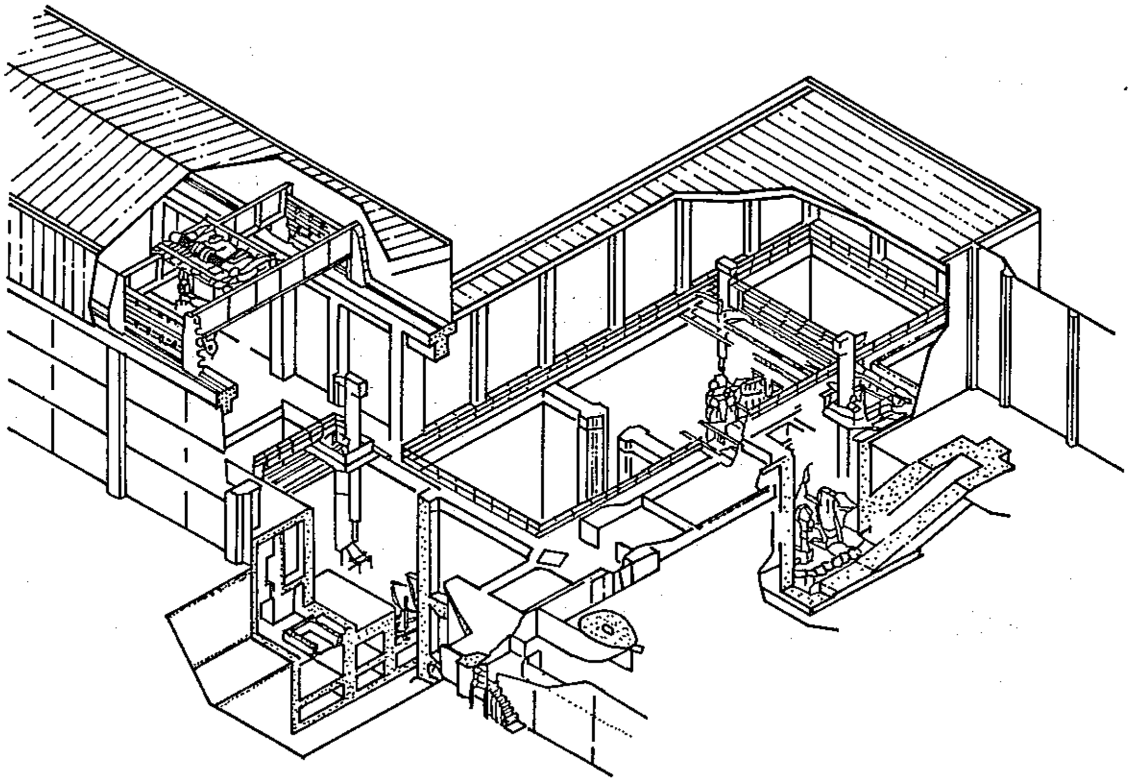
The pool is filled with demineralized water and lined with welded stainless steel plates. The storage pool can hold 97 tons of spent fuels as uranium metal weight. Fuels are stored in movable fuel storage baskets. A basket holds 4 or 8 containers, and one container holds one fuel assembly as shown in Fig.4. The baskets are placed in storage racks of pipe framed structure. Those are made of stainless steel.

The fuels are cooled for more than 100 days before received in the plant. Casks, shipped from reactor, are moved through the truck airlock to the decontamination room by cask crane, where the internal water of the caks is replaced by demineralized water. The caks is then placed in the cask unloading pool, where the spent fuels are taken out of the cask, and put into a container of the basket to keep the storage pool away from contamination. The basket is placed on a transfer cart which is used for the under water transfer to the storage pool. In the storage pool, the fuel storage crane having a telescopic up-down moving grappler are provided for basket handling from the transfer cart to the storage rack. The racks are set out in six longitudinal and nineteen transversal rows. Two longitudinal and one central area in the pool is free of storage to allow the moving of the grappler of the fuel storage crane. After the spent fuels are stored for a certain period of time, a basket is taken out of the storage rack by the fuel storage crane, and moved to the fuel transfer pool by transfer cart. In the fuel transfer pool, the internal water of the container is replaced by demineralized water, then the fuels are taken out of the container and sent to the mechanical processing cell. A list of the main equipment of fuel storage pool is shown in Table 1.



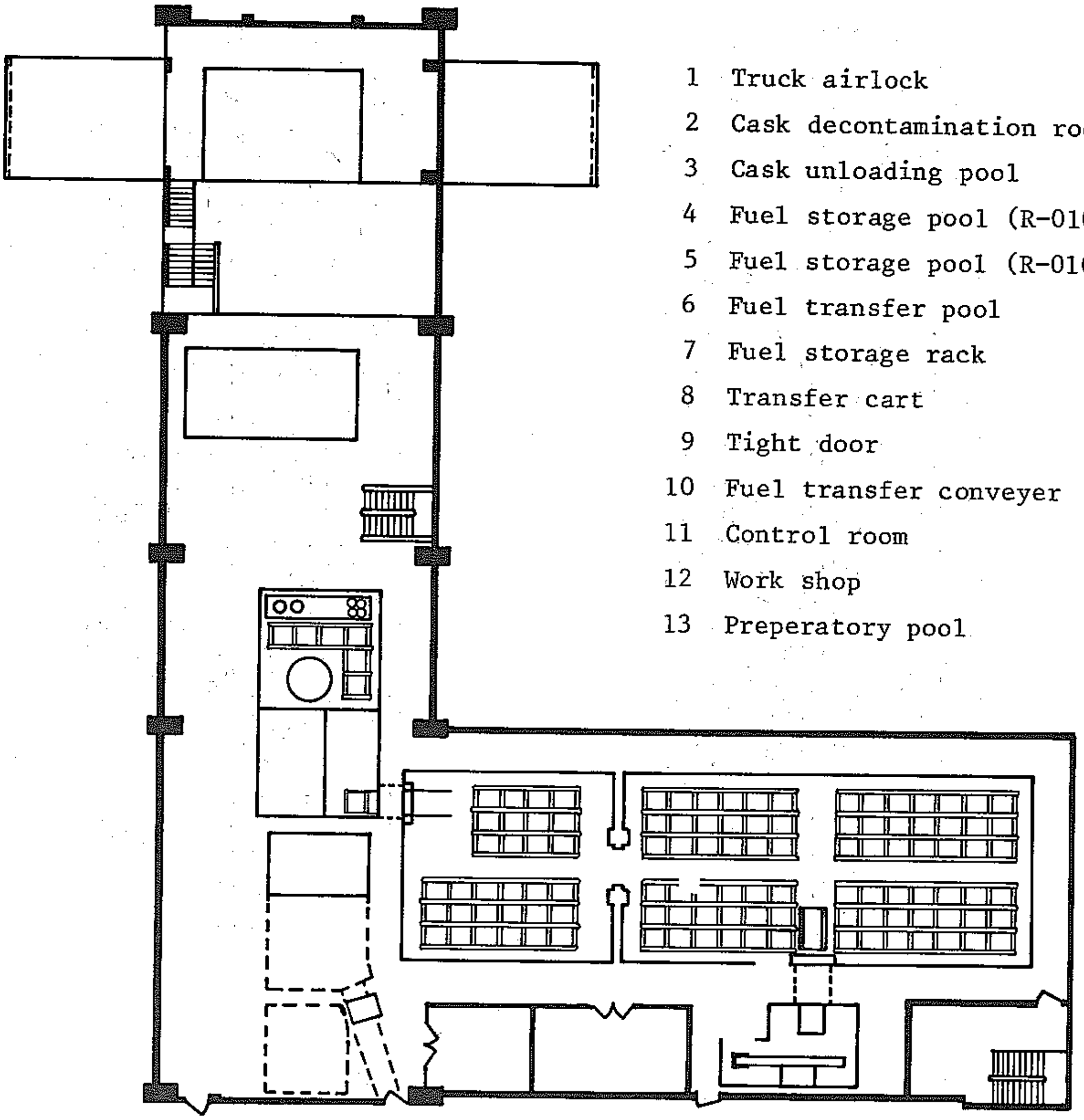
1 Truck airlock
2 Cask decontamination room
3 Cask unloading pool
4 Fuel storage pool
5 Mechanical treatment cell
6 Dissolver loading cell
7 Feed adjustment cell
8 Separation cells
9 U purification cell
10 Pu purification cell
11 Utility room
12 Control room

Fig.1 Perspective cutaway of Main plant



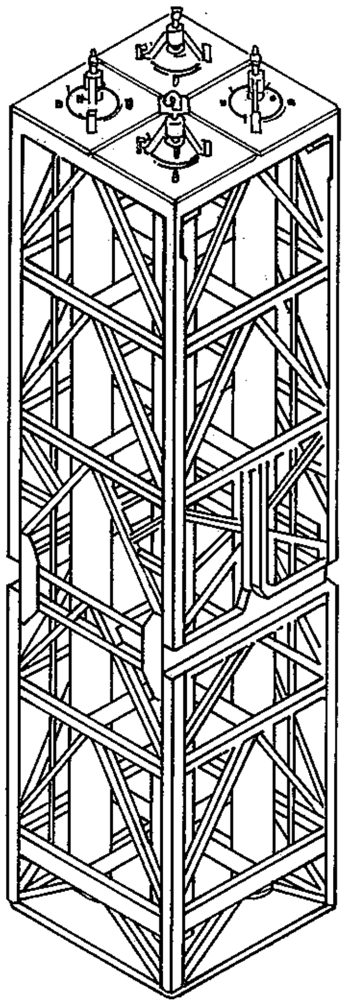
1	110 tons cask crane	CUP	Cask unloading pool
2	Pool crane for cask unloading pool	FSP	Fuel storage pool
3	Fuel storage crane	ETP	Fuel transfer pool
4	Pool crane for fuel transfer pool		
5	Transfer cart		
6	Fuel storage basket		
7	Fuel storage rack		
8	Locker arm		
9	Fuel transfer conveyer		

Fig.2 General layout of spent fuel receiving and storage facility

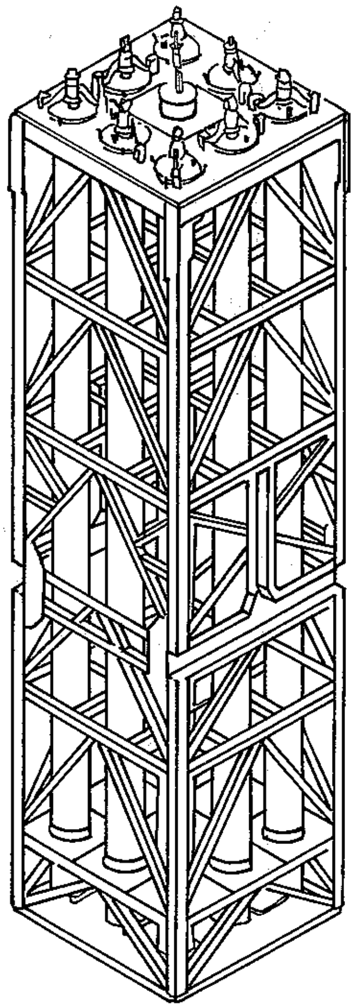


1 Truck airlock
2 Cask decontamination room
3 Cask unloading pool
4 Fuel storage pool (R-0101)
5 Fuel storage pool (R-0107)
6 Fuel transfer pool
7 Fuel storage rack
8 Transfer cart
9 Tight door
10 Fuel transfer conveyer
11 Control room
12 Work shop
13 Preperatory pool

Fig.3 Spent fuel receiving and storage facility arrangement



Basket for 4 BWR
or PWR fuel assemblies



Basket for 8 BWR
fuel assemblies

Fig.4 Fuel storage basket

Table 1 List of the main equipment of fuel storage pool

Name	Number	Characteristics
Fuel storage basket	98	Length 1.1 m Width 1.1 m Height 4.6 m Material SUS 304, SUS 304L
Fuel storage rack	108	Length 1.3 m Width 1.2 m Height 2.7 m Material SUS 304
Pool crane	1	Load capacity 7 ton Grappler 4 fingers type Material SUS 304
Transfer cart	1	Length 2.4 m Width 1.52 m Height 2.32 m Maximum load capacity 7 ton Hydraulic drived type Material SUS 304
Tight door	2	Width 2.0 m Height 6.0 m Hydraulic drived type Material epoxy painted steel
Pool lining	―	Surface area side 930 m^2 bottom 320 m^2 Thickness side 3 mm bottom 4 mm Material SUS 304L
Piping	―	Diameter 165 mm, 216 mm, 89 mm Length (total) 107 m Material SUS 304L

3. Water treatment

1) Water treatment circuit

An outline of the pool water treatment circuit for the fuel storage pool is shown in Fig.5.

The water of the storage pool is drawn off by the pump of an output of 140 m^3/hr from dispersors disposed along the length of the pool about 2 m under the surface level. The water runs through the sand filter of 5.5 m^3 filtering bed. The filtered water crosses the heat exchanger and flows back to the pool by another dispersors about 7 m under the surface level. A part of the filtered water can be pumped at a rate of 30 m^3/hr

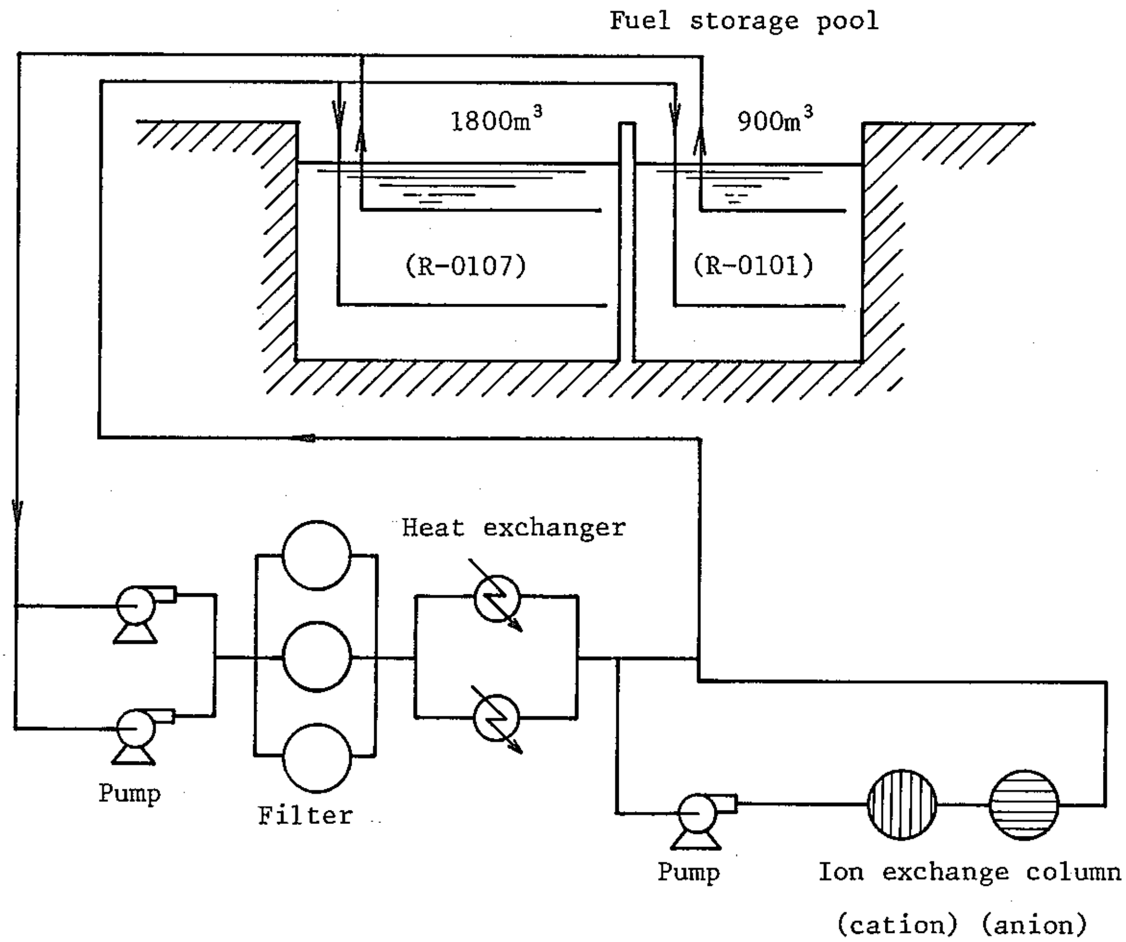


Fig.5 Pool water treatment circuit

to the ion exchanging line, which belongs to the circuit for the cask unloading pool and the fuel transfer pool, for removing dissolved impurities. The purified and decontaminated water comes back into the main circuit. A list of the main equipment of the pool water treatment for the fuel storage pool is shown in Table 2.

Table 2 List of the main equipment of the pool water treatment for the fuel storage pool

Name	Number	Characteristics	
Filter	3	Vertical cylindrical tank	
		Hold up	16 m^3
		Filtering sand volume	5.5 m^3
		Outside diameter	3 m
		Height	2.7 m
		Sand diameter	approx. 1 mm
		Material	SUS 304L
Heat exchanger	2	Vertical plate type	
		Cooling capacity	1.22×10^6 kcal/hr
		Material	Titanium
Pump	2	Horizontal centrifugal	
		Output	140 m^3/hr
		Discharge pressure	3.5 kg/cm^2
		Material	SCS 14
Cation exchange column	1	Vertical cylindrical tank	
		Hold-up	2.8 m^3
		Strong cationic resin (DIAION SKN 1)	1.1 m^3
		Outside diameter	1.2 m
		Height	2.8 m
		Material	SUS 304L
Anion exchange column	1	Vertical cylindrical tank	
		Hold-up	2.8 m^3
		Strong anionic resin (DIAION SAN 1)	1.1 m^3
		Outside diameter	1.2 m
		Height	2.8 m
		Material	SUS 304L
Pump	1	Horizontal centrifugal	
		Output	30 m^3/hr
		Discharge pressure	3.5 kg/cm^2
		Material	SCS 14

2) Water control

The temperature, conductivity, PH value and radioactivity of the water are measured to maintain safe operation. The range of present storage pool water conditions is shown in Table 3.

Table 3 Water control data

Temperature	20 ~ 30 °C
Conductivity	< 3 µ℧/cm
PH value	5.0 ~ 6.0
Total γ	10^{5} ~ 10^{4} µCi/cm^3

When the pressure drop of the filters indicates more than 0.4 kg/cm^2, a filter washing with air and water feeding is necessary. When the conductivity becomes more than 10 µ℧/cm, or the radioactivity becomes more than 0.3 m Ci/cm^3, ion exchange regeneration is carried out by 1 N HNO_3 for cation resins and 1 n NaOH for anion resins.

4. Cumulative sum of fuels received

Since the first reception of JPDR fuel in 1977, Tokai reprocessing plant has received 255 tU of spent fuel assemblies, and already reprocessed about 172 tU. Cumulative sum of received fuels is shown in Table 4. The transition of the amount of fuels stored is shown in Fig.6.

Table 4 Cumulative sum of fuels received

Type of fuel assembly	Number of assemblies received	Fuel weight (tU)	Burn-up (MWD/T)	Cooling Time (days)
JPDR	71	4	110 ~ 5641	2970 ~ 4960
BWR	787	150	6273 ~ 28246	284 ~ 2034
PWR	264	101	1953 ~ 34497	316 ~ 2938
Total	1122	255	——	——

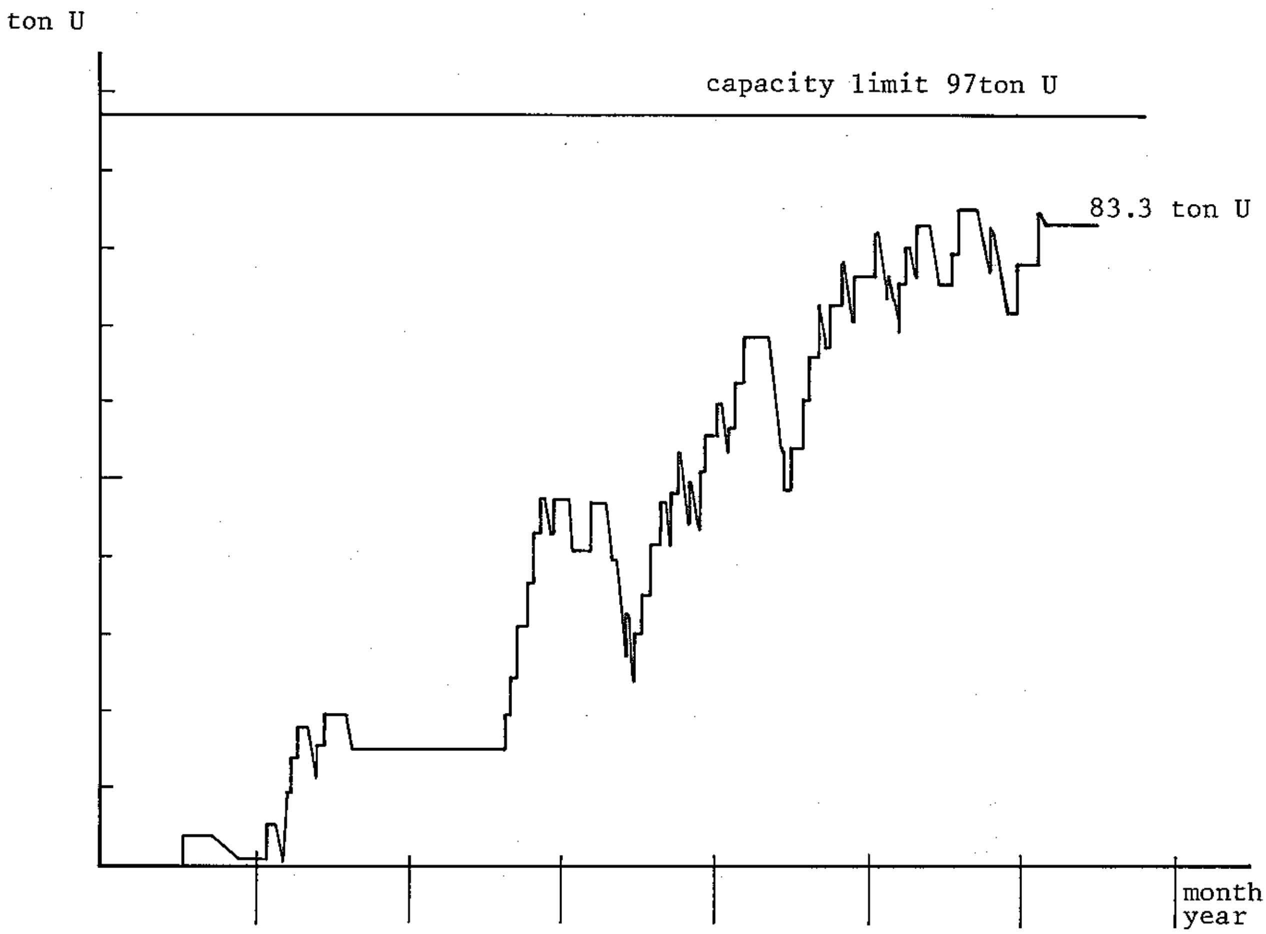


Fig.6 The transition of the amount of fuels stored

5. Measurement of micro elements in fuel storage pool water

1) Measurement procedure

Because of the very small quantities of microelements in the water, it was necessary to investigate the most suitable analytical method for application. Then, the preliminary measurement was performed by the standard samples prior to the measurement of the actual pool water. Fe, Cr, Ni, the principal elements of stainless steel (SUS 304L), were designated as standard samples. The concentrations of Fe, Cr, Ni were adjusted to 2.0 μg/ℓ, 0.4 μg/ℓ, 0.4 μg/ℓ, respectively. Because the micro elements may exist both as ion and colloid in the water, evapolating concentration was applied to concentrate the samples. The samples were concentrated by factor of 50. As a instrument to measure the micro elements, the flameless atomic absorption spectrophotometer was adopted because of its excellent detection sensitivity. As the result of repeated preliminary measurements with the standard samples, the recovery rate was confirmed to be stable quantitatively for measuring micro elements. One of the results of the preliminary measurements is shown in Table 5.

Table 5 The results of the preliminary measurements

	standard sample (μg/ℓ)	measured value (μg/ℓ)	recovery rate (%)
Fe	2.0	1.23	62
Cr	0.4	0.31	78
Ni	0.4	0.30	76

The detection limit of Fe, Cr, Ni resulted in 0.5 μg/ℓ, 0.1 μg/ℓ, 0.1 μg/ℓ, respectively.

The samples of fuel storage water were taken from the fuel storage pool. The measurement of the samples for concentration of micro elements was done in parallel with standard samples and blanc samples at every measurement in order to compensate errors which might enter at operation (caused by instrument, reagnet etc.).

2) Measurement results

The results of measurements (for four months) is shown in Table 6. The value of dissolved oxygen and chlorine were also measured and are given 8 ppm and below 10 ppm, respectively. Several conditions of storage pool water are summarised in Fig.7.

Table 6 The results of measurements of micro-elements concentration (μg/ℓ)

	May	June	July	August
Fe	3.2	1.8	3.0	2.5
Cr	0.3	0.1	0.5	0.5
Ni	0.3	0.1	0.2	0.4

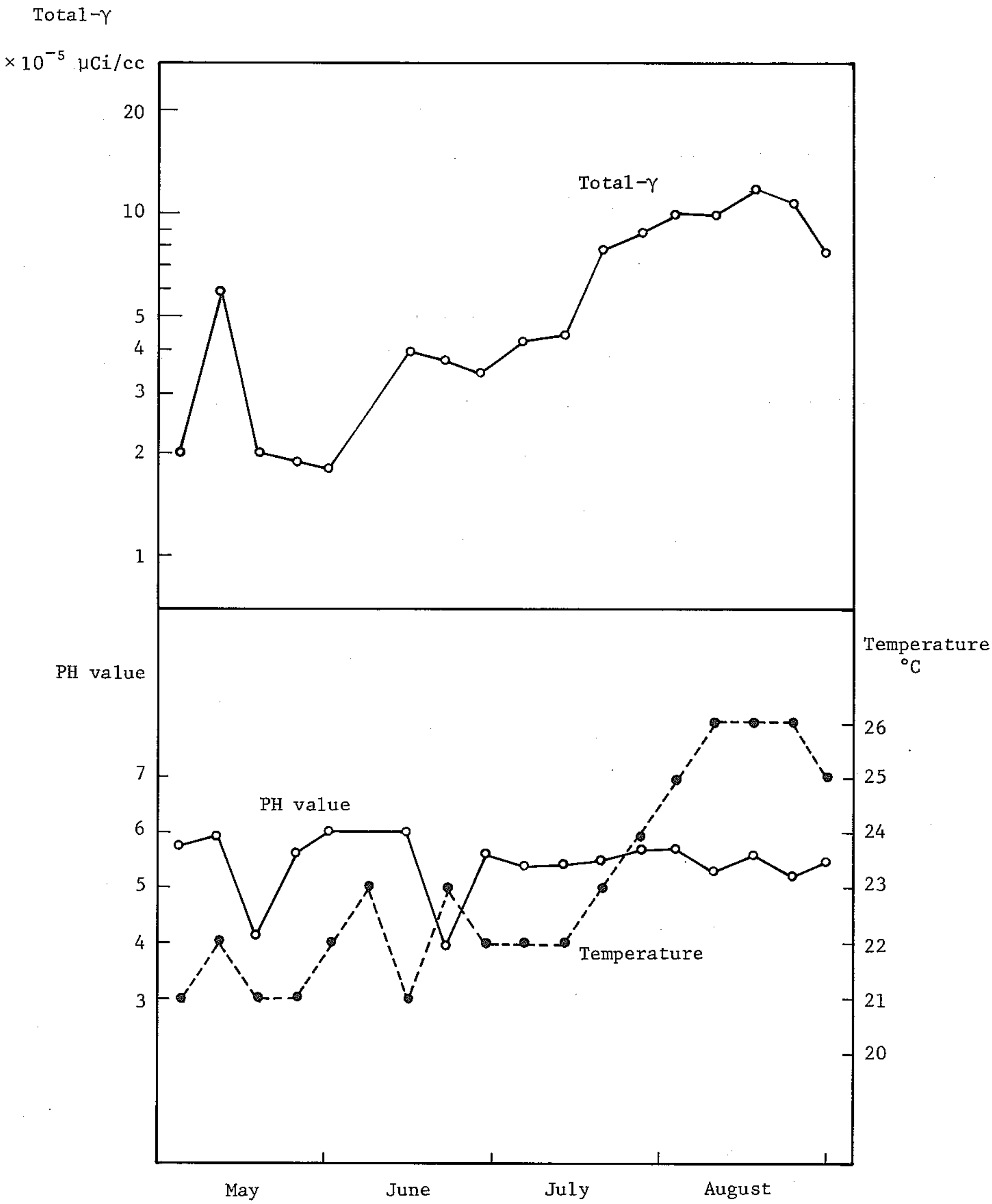


Fig.7 The results of measurements of water condition

6. Conclusion

1) Concentration of micro elements

Concentration obtained by the measurements were Fe 1.8 ~ 3.2 (μg/ℓ), Cr 0.1 ~ 0.5 (μg/ℓ), Ni 0.1 ~ 0.3 (μg/ℓ). According to the experimental data of the report(1), metal composition of released corrosion products from stainless steel is described as Fe 62.7 (%), Cr 15.6 (%), Ni 19.6 (%), Co 2.20 (%), at the dissolved oxygen of 8 ppm. In considering from this composition ratio, the concentration of Fe is appeared to be higher. This is supposed to be the influence of entry from the outside. The detectability of Ni element is rather well as compared with the composition of alloy. Cr seems adequate to an indicator for corrosion control.

Because the measured values were close to the detection limit, dispersion was found in considerable range. To get the lower detection limit, there are the way to improve the concentration factor or to adopt a higher sensitive instrument like spark source mass spectrometer. However, it is very difficult to obtain the instrument because of expensiveness.

To obtain the information on the corrosion of pool components by the estimation of micro elements variation in lapse of time, it is necessary to analyze statistically the measured data. For this purpose, the monthly measurements are continued to accumulate much data. The measurement of dissolved oxygen and chlorine concentration are also performed to know the environmental factors relating to the corrosion.

2) Corrosion rate

As one method to evaluate the result of measured value, the calculation for corrosion rate of the storage pool was tried. In simplifying the pool water treatment circuit for the fuel storage pool as a model like Fig.8, the relation between the corrosion rate of pool components x (mg/hr) and the concentration of micro elements in fuel storage water y (mg/m^3) can be stated by the equation as follow.

$$L\,dy = x\,dt - A\eta_1\,y\,dt - B\eta_2\,y\,dt \qquad \text{———— (1)}$$

where A : Flow rate of sand filter 140 (m^3/hr)

B : Flow rate of ion exchanging line 30 (m^3/hr) × 0.6
The ion exchange line is used only in case of necessity.
The ratio of time on using is about 60 %

L : Water capacity of storage pool system 2700 (m^3)

η_1 : Efficiency of sand filter 0.5 (assumed value)

η_2 : Efficiency of ion exchange column 0.5 (assumed value)

t : Time 6 years ≃ 53000 (hr) has passed since the operation started

The demineralized water of pool is supplied from plant utility, but this quantity is small (20 m^3/month), so it is not considered in this equation.

From equation (1), the differential equation is given as follows

$$\frac{dy}{dt} + \frac{A\eta_1 + B\eta_2}{L} \cdot y = \frac{x}{L} \qquad \text{———— (2)}$$

and its solution is

$$y = \frac{x}{A\eta_1 + B\eta_2}\left\{1 - \exp\left(-\frac{A\eta_1 + B\eta_2}{L} \cdot t\right)\right\} \qquad \text{———— (3)}$$

Because t is very large, y is expressed as follows.

$$y = \frac{x}{A\eta_1 + B\eta_2} \qquad \text{———— (4)}$$

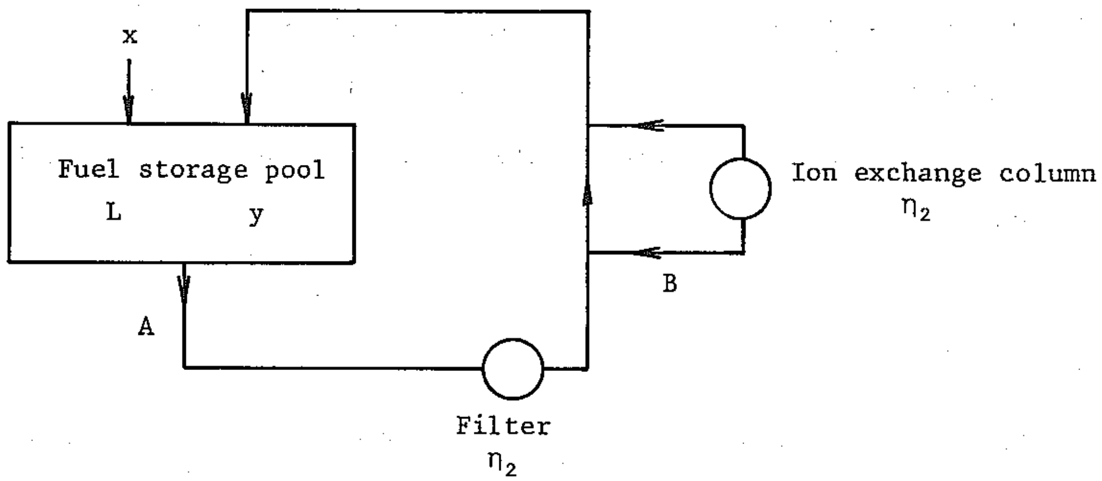


Fig.8 Model circuit

In applying the measured value of Cr 0.3 (μg/ℓ), for example, Fe and Ni correspond to 1.2 (μg/ℓ), 0.1 (μg/ℓ), respectively, from the composition ratio of alloy. Therefore, the concentration of stainless steel corresponds to 1.6 (μg/ℓ). Here, subsituting $y = 1.6$ (μg/ℓ), $A = 140$ (m^3/hr), $B = 18$ (m^3/hr), $\eta_1 = 0.5$, $\eta_2 = 0.5$ to the equation (4), the corrosion rate becomes $x = 126$ (mg/hr). This corrosion rate is much greater than the data of report. For the definite evaluation, more data accumulation is required.

By taking the assumption that the whole surface corrosion is occurring, the reduction of thickness R (mm/year) is written as follows.

$$R = \frac{x}{S} \times \frac{1}{\rho} \times 8.76 \times 10^{-3} \qquad (5)$$

where S :	Surface area of pool components	5380 (m^2)
	pool lining	1250 (m^2)
	fuel storage basket	3680 (m^2)
	fuel storage racks	360 (m^2)
	piping	60 (m^2)
	others (transfer cart, crane grappler, etc.)	30 (m^2)
ρ :	Specific weight	7.8

Substituting $x = 126$ (mg/hr), $S = 5380$ (m^2), $\rho = 7.8$ to the formula (5), the reduction $R = 2.6 \times 10^{-5}$ (mm/year) is obtained. This is only a little as compared with the pool lining in thickness of 3 mm (the side) or 4 mm (the base).

7. Acknowledgment

We would like to thank MM.K. Miyauchi and H. Hirayama (Analysis Section) for the cooperation of measurement and fruitful discussions.

8. Reference

(1) T. Iwahori et al., "Influence of dissolved oxygen and hydrogen and surface film on the corrosion products release from stainless steel in high purity water at room temperature" Boushokugigyutsu, 32, 202-207 (1983) (in Japanese)