THE MINIMIZATION OF RADIOACTIVE RELEASES TO THE SEA FROM THE TOKAI REPROCESSING PLANT

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Abstract

The Tokai Reprocessing Plant(TRP) started hot operation in September 1977. The total amount of about 790tU of spent fuel, generated in Japan, has been successfully reprocessed as of December 1994. Low-level liquid wastes have been treated safely with the low-level waste treatment process.

The design of TRP was based on foreign technology. In the early stage of designing, the radioactivity released to the sea was estimated at approximately 2.6 TBq/day (70 Ci/day) for beta activity (except for tritium). Later, PNC added an evaporator to the process to reduce the level down to 1/100, i.e. 9.6 TBq/year (260 Ci/year) or 2.6×10^{-2} TBq/day (0.7Ci/day). In addition, under the supervision of the government, PNC started R&D to further decrease the radioactivity released to the sea in terms of ALARA. Aiming at reducing the activity from 9.6 TBq/year (260 Ci/year) to 1/10 of that value (i.e. 26 Ci/year), the release reduction technology development facility was added. This facility was incorporated into the low-level waste treatment process in 1980, before starting the regular operation of TRP.

Since the fuel reprocessing commenced, total radioactivity discharged to the sea has been 1.9×10^{-2} TBq (0.51 Ci) for beta activity, as of December 1994. Before incorporating the release reduction technology development facility, the yearly level was $3.7\times10^{-3}\sim7.4\times10^{-3}$ TBq (0.1～0.2 Ci). After incorporation of the facility, radioactivity released to the sea was greatly decreased to non-detection levels in recent years, in spite of increasing annual reprocessing amounts. Although serious equipment failures have occurred such as the acid recovery evaporator and the dissolvers, there was no influence on radioactivity released to the sea.

1. INTRODUCTION

1.1. Overview of Tokai Reprocessing Plant.

PNC started on the reprocessing project in 1956 when Atomic Energy Commission (AEC) of Japan decided that reprocessing of spent fuel and treatment of radioactive waste should mainly be done by Atomic Fuel Corporation (AFC, the predecessor of PNC). In 1959, AEC formed Advisory Committee for Reprocessing within AEC to formulate a guideline for

development of reprocessing technology. Along with AEC's and Advisory Committee for Reprocessing decision, AFC began the preparation for construction of reprocessing plant in Tokai-mura, about 100km northeast of Tokyo.

After studying the situation of foreign reprocessing plants, Advisory Committee for Reprocessing recommended that it was desirable to construct a practical size plant of 0.7-1 t/day based on a foreign technology, in 1962. In 1963, the AFC entered contracts of preliminary design of plant with foreign companies, and detailed design was carried out by SGN from 1966. In 1967, PNC succeeds to reprocessing project from AFC. Since 1968 and in parallel with the ongoing detailed design, the governmental licensing procedure had been followed and permission for plant construction was gained in 1970.

As shown in Table Ⅰ, plant construction was started on 1971 and was completed on 1974. In this construction period, blank test was made in parallel. Chemical test and uranium test (using unirradiated uranium) were proceeded step by step. The active test was started at September 1977, and the prior inspection for operation was conducted. PNC got the license for operation in 1980, and operation started on 1981. Recently, the scheduled long period of shut-down of plant operation was set to replace and improve the plant equipments, to prevent sudden equipment failure and to increase reprocessed amounts.

Up to the end of 1994, the total amount of reprocessed fuel from LWRs and ATR (Advanced Thermal Reactor using heavy water as the moderator) "Fugen" was about 790 tons, with a maximum burn-up of 35000MWD/tU. Total amounts of recovered plutonium nitrate as final product reached about 5.4 tons up to December 1994. Most of recovered Pu has already been sent to Plutonium Conversion Development Facility for use at the ATR "Fugen", the experimental FBR "Joyo", and proto-type FBR "Monju"[1]. PNC came up with some difficulties through this reprocessing project. This paper deals with the experience of the low-level liquid waste treatment.

1.2. Difficulty in the Low-Level Liquid Waste Treatment.

Difficulty was occurred because of social and environment change[2]. This difficulty was beyond what was considered at initial designing of plant. The management of radioactive waste in Japan is important problems, because not only very densely populated but also people depend much upon fish and shells for their important resources of protein. So, Japanese interests in the sea are immeasurable. Construction permit authorization was given by government in 1970 and construction work was started on 1971, but demand for improvement for environment safety continued even after construction started.

2. Description of the Low-Level Liquid Waste Treatment Process

2.1. Origin and Classification of Liquid Wastes.

The liquid wastes generated from the reprocessing plant are classified into high-, intermedium-, and low-level (HLLW, ILLW, LLLW). The origin of liquid waste are shown in Table Ⅱ.

HLLWs and ILLWS are treated in the separations and purification plant which usually called MP (Main Plant), whereas LLLWs are transferred to low-level waste treatment facility, which is usually called AAF (Auxiliary Active Facility), through underground pipelines after routinely checking up activity and chemical properties.

2.2. Initial Process.

At the initial design, TRP was provided with LLLW treatment facility with AAF only.

AAF is provided with one unit evaporator, one unit flocculater, and one unit sand filter. This evaporator is self-thermocompressing natural-circulation type with capacity of 50m^3/day, and provided with a cyclone and a wire mesh demister to remove mist, which is called 1st.-evaporator now. The flocculater has capacity of 120m^3/day, which called chemical treatment process. Coprecipitation was made by adding of calcium carbonate and ferric hydroxide.

LLLWs were treated with some of above equipments depend on their radioactive concentrations and chemical properties. This initial process is simple, as shown in Fig.1. Higher-LLLW, whose activity concentration is 370kBq/cm^3 to 37Bq/cm^3, is subjected to one-stage evaporation. Whereas lower-LLLW, whose activity concentration is less then 37Bq/cm^3, is coprecipitated and filtrated. The distillate from acid recovery process, however, was discharged without any treatment. The resultant concentrate and chemical sludge are stored in the exclusive storage vessels.

2.3. Modification of the Process.

2.3.1. Oil removal from effluent.

In 1976, oil contamination limit of sea discharge effluent was tighten because of revised chemical pollution regulation. PNC finds that the oil concentrations in distillate from the acid recovery process in MP and 1st.-evaporator were over alternate limit sometimes. To solve this problem, PNC entered design for oil removal facility and chemical treatment process was modified temporary by adding the active carbon to remove entrained TBP, as shown in Fig.2. The addition of active carbon was very effective in reducing the oil discharge.

2.3.2. Addition of Evaporator.

Although the initial regulation value on the sea-discharge was just 37GBq/day (except for tritium), demand for improvement in environment safety enhanced during the construction period. Fisherman's associations and related organizations entered suit against construction work. To avoid this problem, PNC modified the low-level liquid (LLLW) waste treatment process by constructing another facility. This facility called Second LLLW evaporation facility (usually called E-facility).

E-facility provides one unit of natural-vertical-circulation evaporator with capacity of 90m^3/day. This evaporator called 2nd-evaporator. 2nd-evaporator treated the distillate from 1st.-evaporator. The evaporation technique is very effective to produce the sufficient low-level distillate and reduces the radioactivity in the sea-discharge effluent. E-facility was designed and constructed by domestic companies, and construction was completed early 1975, before beginning of active test. This evaporation facility was incorporated with LLLW treatment process to modify the process as shown in Fig.3.

2.3.3. Further Addition of Evaporator.

In addition to the above, PNC has been made effort to reduce the radioactivity discharged to environment from Tokai Reprocessing Plant (TRP) along with "ALARA" philosophy. PNC decided to construct 3rd-evaporation facility (usually called Z-facility in connection with Zero discharge).

Z-facility contained one unit evaporator. This 3rd-evaporator is same type as 2nd-evaporator but it capacity is 210m^3/day. The construction of this new facility start on 1977 and completed on 1979. This facility was designed and constructed also by domestic companies.

In the early time of the active test, PNC came up with difficulty of high alpha activity in the distillate from acid recovery process which exceed the regulation value for sea discharge. As a result of provisional measurement, PNC was obliged to modify the process again to treat a part of the distillate from the acid recovery process and a part of lower-LLLW by 2nd-evaporator, but it had no capacity to treat all liquids. So, PNC intended to use 3rd-evaporator for operation routinely. PNC had made the tests using actual liquid waste during the active test and the results showed sufficient decontamination factors. The modification of this process is carried out as shown in Fig.4. 3rd-evaporator has enough capacity to treat the produced waste. So, the 2nd-evaporator retired from the service and became supplemental evaporator now.

2.3.4. Effluent Clean-up by Charcoal Adsorption Column.

In order to protect the surrounding seawater from chemical pollutants due to entrained TBP, the charcoal adsorption columns installed before sea-discharge process, instead of the modified chemical treatment. This new facility has charcoal adsorption columns, and which called the Oil Removal facility (usually called C-facility). C-facility was incorporated into the process in 1979. The final process for LLLWs is shown in Fig.5. Higher-LLLWs are subjected to two-stage evaporation, whereas low-LLLW is coprecipitated or evaporated in one stage, and the distillate from the acid recovery process (which was not treated at the initial process) is also subjected to one-stage. To reduce radioactivity discharge, part of lower LLLW is treated by evaporator. Finally, decontaminated liquid wastes are de-oiled in charcoal adsorption columns. LLLWs treated as above are discharged through sea-discharge pipeline after confirming the radioactivities and the chemical pollutants in the effluent.

3. Results.

LLLWs are decontaminated by evaporators or flocculater sufficiently below the discharge limits (2.96×10^{-2}Bq/cm^3 of alpha activity and 12.2Bq/cm^3 of beta activity) and are further de-oiled below 5.0ppm before being discharge into the Pacific Ocean, about 3.7km apart from seaside. As shown in Fig.6 and Fig.7, discharged radioactivity to the sea is remarkably decrease since 3rd-evaporator has been incorporated in LLLW treatment process since 1980. Up to December 1994, total discharged radioactivity is only 4.6×10^{-4}TBq for alpha, 1.9×10^{-2}TBq for beta (except for Tritium). On the other hand, concentrate have been fixed in bitumen since October 1982.

4. Another Improvements.

4.1. Reduction of iodine discharge.

At the beginning of the active operation, we had the problem of that how to control the iodine released into the environment. In order to solve this problem, the measurements of iodine in the gaseous and liquid streams have been made through the active operation.

From the result of measurements, it was estimated that more than 99% of iodine in the spent fuel was released into off-gas circuit during the dissolution. Most of the iodine released to off-gas circuit was resorbed at scrubber to liquid streams (higher-LLLW) which was transferred to AAF and then subjected to two-stage evaporation[3]. Methods for decreasing the volatility of iodine include adjusting of the pH of solution adding NaOH to produce new conditions under which the species is non-volatile.

It is desirable to have as high volume reduction as possible to minimize the size and of concentrate vessel and reduce the burden on the bituminization process. PNC has made the

test for reducing iodine discharge by the alkaline process. Test results indicated the technical feasibility of reducing iodine discharge sufficiently by adaptation the second stage only. Fig.8 shows that radioactive iodine discharged into the sea has been considerably reduced by adaptation of the alkaline process. The amount of radioactive iodine released into the sea is currently about 1% or less than of estimated inventory in the spent fuel.

4.2. Prevent the Foaming.

Foaming is a major problem associated with evaporation. A small amount of entrainment can reduce the decontamination factor to an unsatisfactorily low. Once foaming occurs, the decontamination factor decreases markedly due to the rise in liquid level, which causes an increase of entrainment and, in the worst case, carry-over of solution into the distillate. The ways of prevent foaming employed are improvement of evaporation control and ITV-monitoring.

5. CONCLUSION

The design of Tokai Reprocessing Plant was based on a foreign technology. However, social environment is different in each country and it has a great influence upon waste management. Accordingly, we modified the LLLW treatment process by our own technology, in order to fit and fix the Tokai Reprocessing Plant operation to the state of affairs in Japan.

This modification reduced radioactivity being discharge to the sea over many years, thereby satisfied the demands of environment safety. These results have been effective in obtaining the public acceptance for fuel reprocessing in Japan.

Reference

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2. Y.Nojima, et al. : "Operational Experience in the Low Level Liquid Waste Treatment at Tokai Plant", Proc. of Fuel Reprocessing and Waste Management, Vol.1, p.505-515, Jackson, 1984

3. G.Fukuda, K.Matsumoto and K.Miyahara : "Experience and Projects Concerning Treatment, Conditioning and Storage of All Radioactive Waste from Tokai Reprocessing Plant", IAEA-CN-43/131, Vol.2, p.279-292, Seattle, 1983

Table I Operation Schedule on Tokai Reprocessing Plant

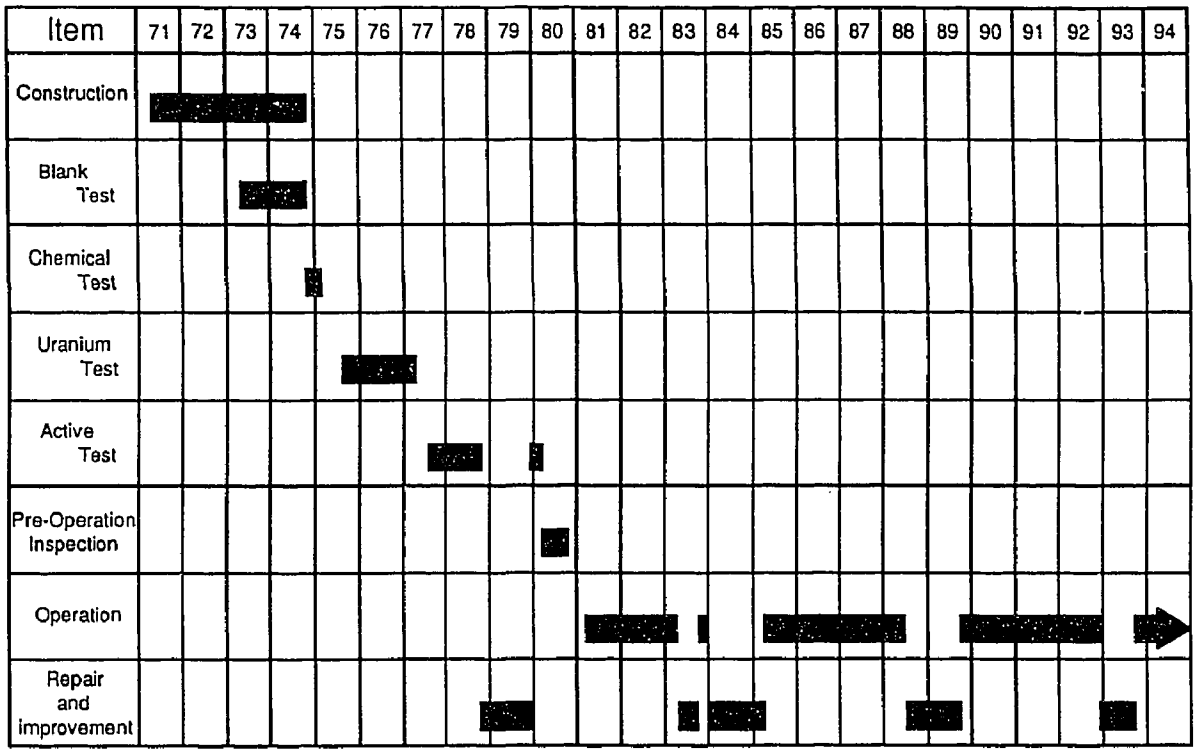

Table II Classification of Liquid Waste

High-level liquid waste	1st cycle effluent 1st cycle solvent wash effluent Concentrate from acid recovery process
Intermedium-level liquid waste	2nd and 3rd Uranium and Plutonium cycle Aqueous effluents DOG and HLLW vent condensates Plutonium evaporator condensate
Low-level liquid waste	Off-gas scrubbing effluents Storage pool cleanup system backwash Solvent wash effluent Floor drain Laundry effluent Distillate from acid recovery process

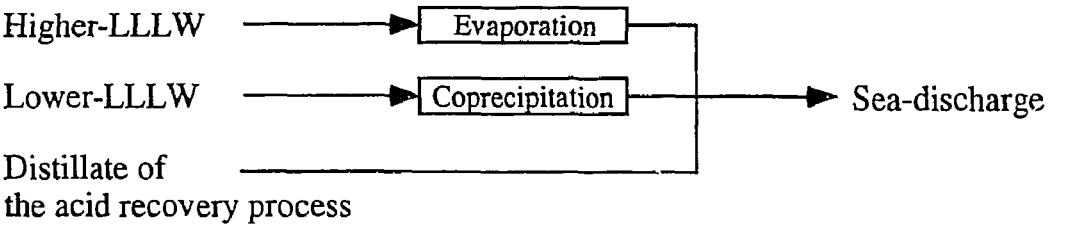

Fig.1 Initial LLLW Treatment Process

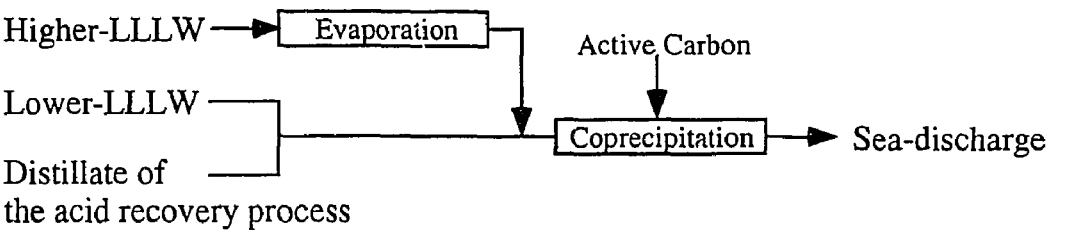

Fig.2 Modified Process

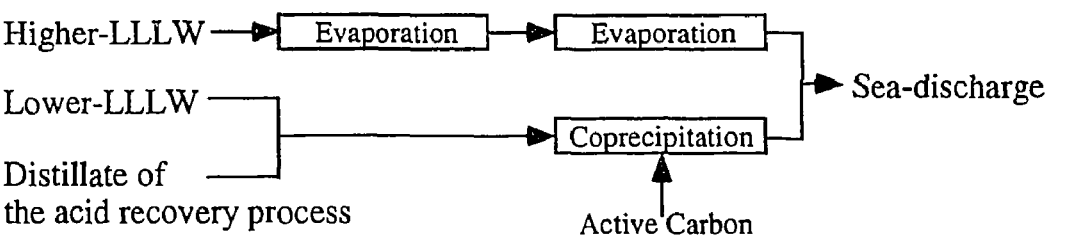

Fig.3 2nd-modified Process

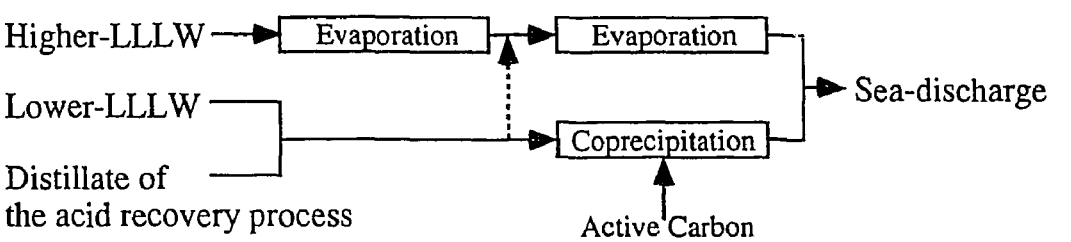

Fig.4 3rd-modified Process

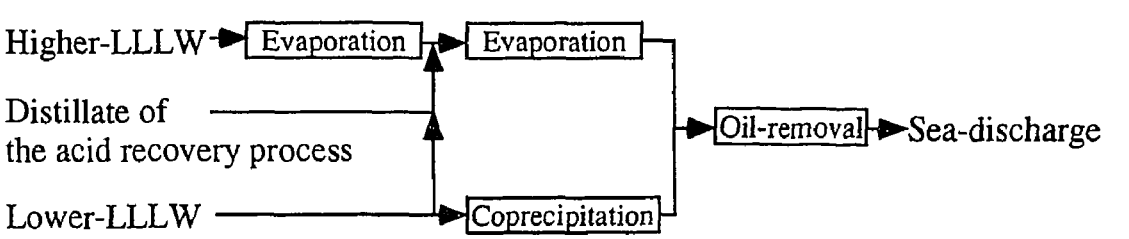

Fig.5 Final modified Process

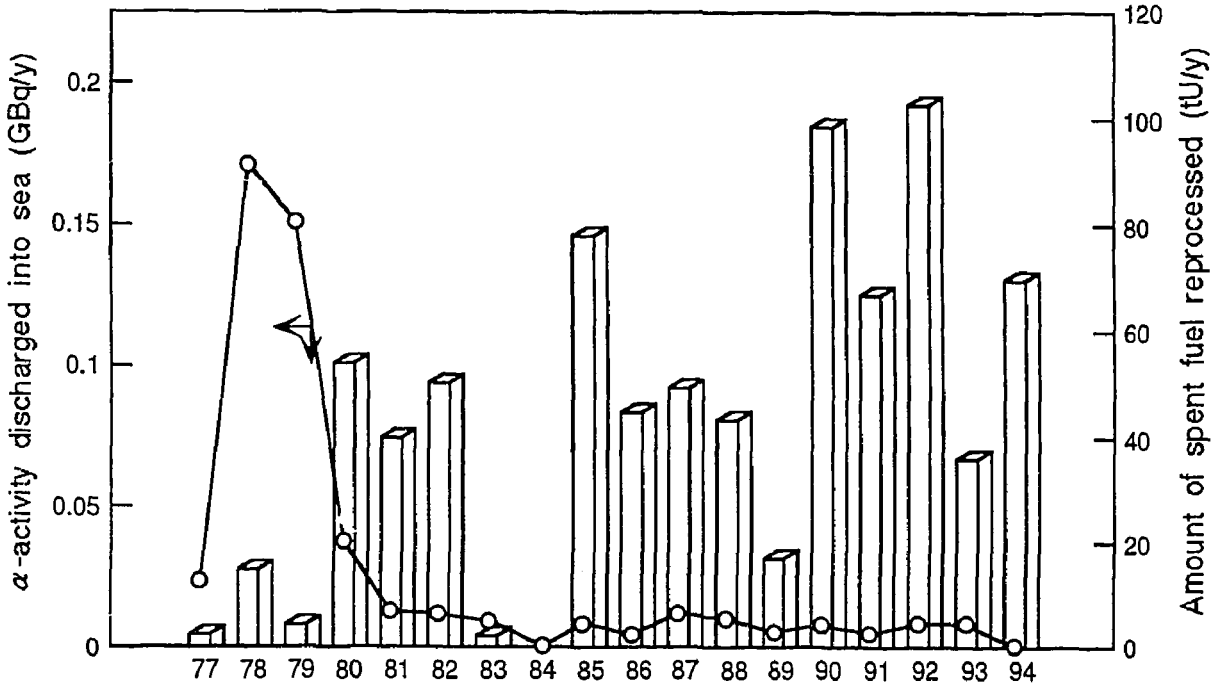

Fig.6 Discharged α-activity to Sea

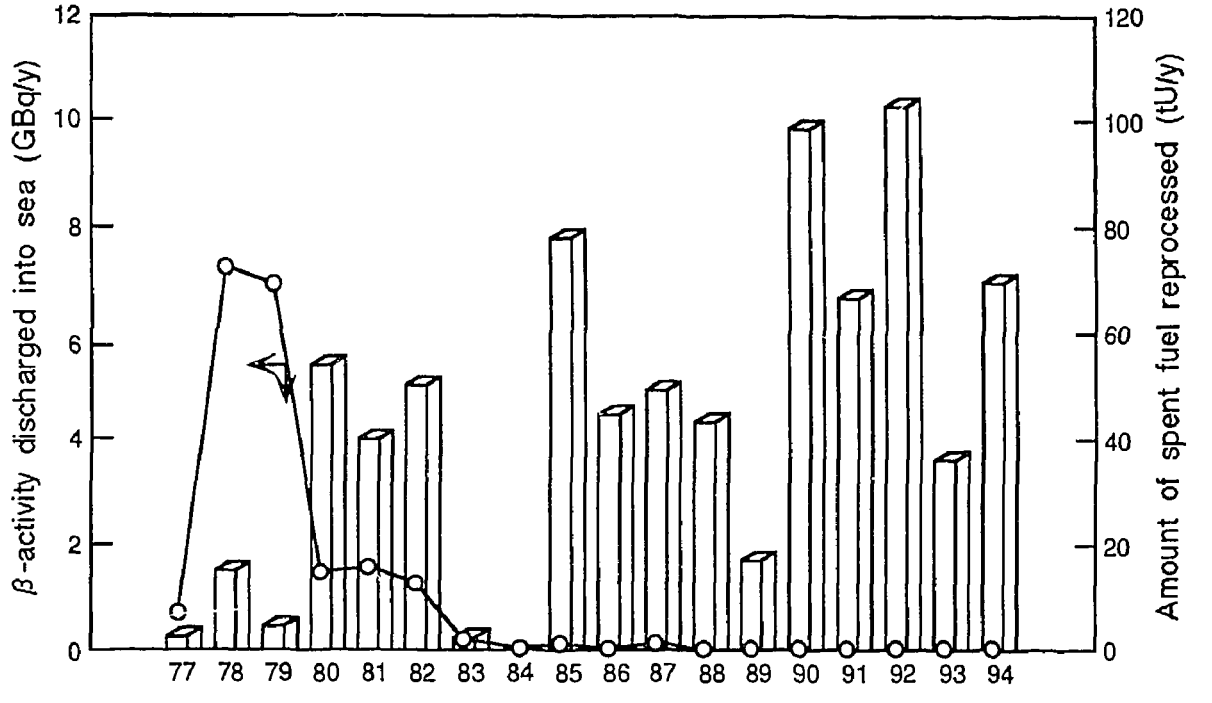

Fig.7 Discharged β-activity to Sea

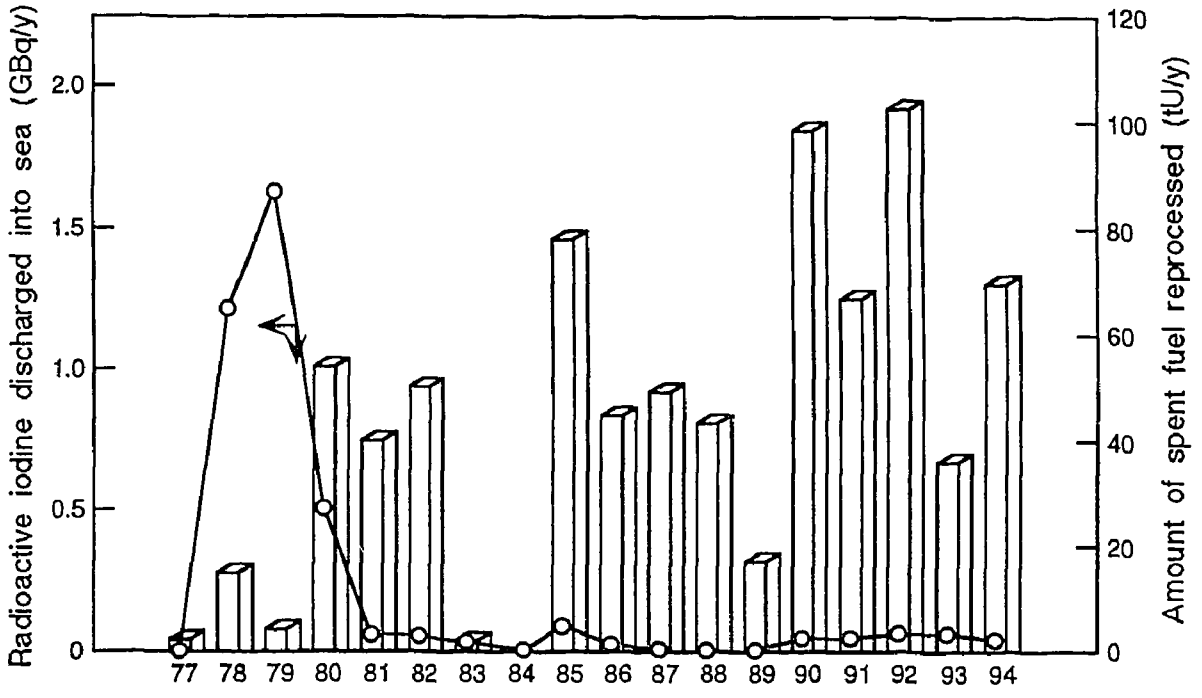

Fig.8 Discharged Radioactive Iodine to Sea