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A Study on the Application of Ion Crystallization Technology to the APR 1400 Liquid Waste Management System

핵종 이온 광물화 처리기술의 APR 1400 발전소 액체방사성폐기물관리계통 적용 위치에 대한 고찰

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The application of ion crystallization technology was considered as a way to increase the operating efficiency and improve the operating performance of a liquid waste management system (LWMS) in the Advanced Power Reactor 1400 (APR 1400). Although ion crystallization technology has not been practically applied to Nuclear Power Plants (NPPs) until now, a previous experimental study demonstrated that it is possible to selectively remove at least 95% of various nuclide ions present in the liquid radioactive waste of NPPs. We reviewed the possibility of applying ion crystallization technology to the existing LWMS by applying the nuclide removal rate of ion crystallization technology and prepared a way to improve the existing LWMS in the APR 1400. Furthermore, we determined the optimized application location of ion crystallization technology in the existing LWMS by considering decontamination characteristics of the ion crystallization technology and the existing LWMS design features and operating experiences. The application of ion crystallization technology to the liquid waste collection tank, where liquid radioactive materials are collected, will have the least impact on the existing design while providing the greatest improvement. It is expected that the application of ion crystallization technology to the current APR 1400 or new NPPs would increase the operating efficiency of the LWMS and result in an improvement of system performance.

Keywords: LWMS, Reverse Osmosis, Demineralizer Module (Cation Bed, Mixed Bed)

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APR 1400 액체방사성폐기물관리계통 효율성 증가와 계통의 성능 개선을 위한 방안으로 핵종 이온 광물화 처리기술을 적용하는 것을 고려하였다. 핵종 이온 광물화 처리기술은 현재까지 발전소에 실제적으로 적용되진 않았지만 원자력발전소의 액체방사성폐기물에 존재하는 다양한 핵종 이온을 최소 95% 이상 선택적으로 제거 가능 하다는 것을 실험적으로 증명한 바 있다. 본 논문은 핵종 이온 광물화 처리기술의 제염율을 반영하여 기존 설계에 적용 가능성을 확인하였으며, 기존 설계를 개선할 수 있는 방안을 마련하였다. 핵종 이온 광물화 처리기술의 제염 특성과 기존의 액체방사성폐기물관리계통 설계 및 운전 경험을 고려하여 최적의 적용 위치를 결정하였다. 원자력발전 운영에 따라 발생하는 액체방사성물질이 수집되는 수집탱크에 핵종 이온 광물화 처리기술을 적용하는 것이 기존 설계의 영향이 가장 적을 것이며, 개선 효과도 가장 큰 것으로 해석되었다. 핵종 이온 광물화 처리기술이 현재의 APR 1400 발전소 또는 신규 원전에 적용될 경우 액체방사성폐기물관리계통의 운전 효율성 증가와 계통의 성능 개선이 기대된다.

중심단어: 액체방사성폐기물관리계통, 역삼투압설비, 정화설비 (양이온교환기, 혼상이온교환기)

1. Introduction

The liquid radioactive materials of nuclear power plants can be classified into radioactive suspended particles floating in the liquid of the system, radioactive materials floating in the liquid without dissolving, and radioactive substances dissolved in the liquid. Among the radioactive materials generated during operation of NPPs, the radioactive materials that are dissolved in the liquid or radioactive suspended particles floating in the liquid of the system are collected and concentrated by means of filtering, ion exchanger, evaporation, and concentration and become radioactive waste through a solidification process. Domestic NPPs have applied various liquid radioactive waste treatment technologies such as a single evaporator, dual evaporators, centrifuge and reverse osmosis to handle liquid radioactive materials generated during operation. Liquid radioactive waste treatment technology has been continuously improved due to the increase of social and political interests in the environment and also the strengthening of regulatory requirements [1]. However, efforts and research to reduce public anxiety about nuclear energy and to minimize the generation of radioactive waste should be continued.

Shin-Kori Units 3&4, which were built with the first model APR1400, are in commercial operation, and have an improved LWMS using reverse osmosis packages and ion exchange modules to properly treat liquid radioactive materials generated during operation. The LWMS of Shin-Kori Units 3&4 is a system that can collect, separate, store, process, sample collection and monitor liquid radioactive wastes, and is installed to protect the plant workers, the publics and the environment. The LWMS of Shin-Kori Units 3&4 has sufficient redundancy to continuously collect, store and processes the liquid effluent most efficiently by classifying the liquid effluent in compliance with its radiological characteristics [2]. This can reduce the load on the process facility by monitoring and discharging a large amount of potential pollutant waste, which is almost or completely uncontaminated, after a minimum treatment process. The main process of the LWMS effectively reduces the emission of radioactive effluents and minimizes the amount of radiation emitted to the environment by using a reverse osmosis (R/O) package which is the major component [2].

To maintain the continuous operation and efficient management of NPPs, waste generation during NPP operation should be reduced. The domestic NPPs have applied a steadily improved liquid radioactive waste treatment technology. As a result, the total amount of liquid radioactive effluents released to the environment from the operation of the LWMS did not increase compared to the increase

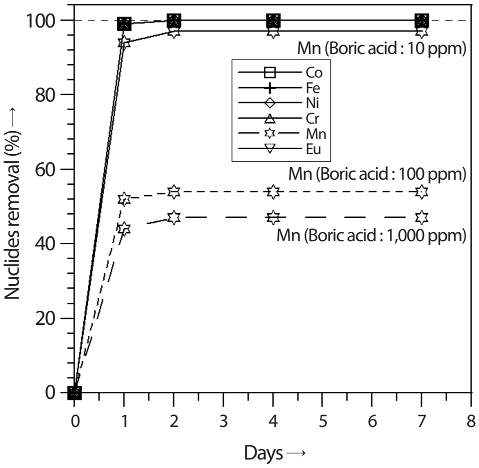

Fig. 1. The removal ratio of major nuclides by ion crystallization technology [4] .

in the number of NPPs [3]. As the liquid radioactive waste system is improved, the annual amount of solid radioactive waste generation from the operation of the LWMS was also decreased [1]. However, efforts to reduce the generation of liquid radioactive waste are still required because of the public's sensitive response to NPP operations and the tightening restrictions on NPPs by governments and regulators. Various researches have been conducted to develop decontamination methods that can reduce spent resin generation. As part of these efforts, ion crystallization technology, which converts the major nuclide in radioactive liquid waste into inorganic crystal minerals, was developed [4]. In an early study, the technology used a chemical method to remove the major nuclide present in radioactive effluent by converting it into inorganic crystal minerals. It can remove the major nuclides up to 99% as shown in Fig. 1. Through the application of ion mineralization technology, wastewater treatment performance can be improved and the amount of spent resin wastewater generated can be reduced. Also, the application of the technology to the APR 1400 LWMS design is expected to improve the operating efficiency of the reverse osmosis which is the main component in the LWMS. In this paper, we analyzed the variation of the radiation concentration in the application of the ion crystallization technology to the LWMS in the APR1400 using the design basis liquid effluent concentration values in one of the liquid waste collection tanks in the LWMS and compared the advantages and disadvantages according to the candidate application location.

2. Review of the Liquid Waste Management System in the APR 1400

The LWMS installed in all NPPs in Korea has major components and design characteristics that differ from plant to plant, and has been applying improved technologies to increase operational efficiency and to compensate for disadvantages [2, 5].

2.1 Design Features and Classification of the LWMS in Domestic NPPs

The liquid waste management system in domestic NPPs can be classified into four categories according to its design features as shown in Table 1. Category Ⅰ is a design that was initially applied to domestic NPPs. A single evaporator was provided as a major component and the capacity of the system was decided based on the amount of liquid radioactive waste generated during in normal operation. Category Ⅱ increased the processing capacity compared to category Ⅰ, and applied the concept of redundancy to improve the processing performance. As a result, the LWMS had a capacity to demand not only normal operation but also in the anticipated operational occurrences (AOOs) condition. However, there were some problems such as the deterioration of the function of the evaporators due to oil and floating solids accompanied by the inflow of effluent. Category Ⅲ reduced the amount of solid radioactive wastes generation by applying centrifuge and ion exchange component to improve the LWMS design. However, the emission of radioactive materials was increased compared to category Ⅱ [1]. Category Ⅳ applied a

Table 1. LWMS categorization according to major component and design features [1, 2, 5]

Categorization	Applied NPPs	Major Components	Design Features
Category Ⅰ	Kori 1&2, Hanul 1&2	- Single evaporator	- Single component - Low capacity
Category Ⅱ	Kori 3&4, Hanbit 1,2,3&4, Hanul 3&4	- Dual evaporators	- Redundancy - Upgrade capacity
Category Ⅲ	Hanbit 5&6, Hanul 5&6	- Centrifuge - Ion exchanger	- Centrifuge - Selective ion exchanger
Category Ⅳ	Shin-Kori 1&2, Shin-Wolsung 1&2, Shin-Kori 3,4,5&6, Shin-Hanul 1&2	- Ion exchanger - Reverse osmosis	- Pre-treatment module - Reverse osmosis module - Demineralizer module (1 Cation Bed and 2 Mixed Beds) - Increased reuse rate of the processed liquid waste

reverse osmosis (R/O) component instead of the centrifuge to increase processing efficiency and allow reuse of processed liquid waste. Compared with previous designs, the amount of solid radioactive wastes generated was significantly reduced and the processing efficiency was increased [1]. However, the design of NPPs applying a reverse osmosis component has experienced common difficulties due to scaling and fouling, which reduces permeate flux [6]. According to the operating experience of domestic NPPs, the flow rate of permeated water decreased due to scale formation and the number of chemical cleaning increased during the operation of reverse osmosis component [7]. As a result, fine control techniques are required to suppress scale formation, and efforts are required to improve processing performance. Any processing technique has its advantages and disadvantages, and may not be perfect but can overcome difficulties by using additional techniques or equipment that complement each other or mitigate disadvantages. According to the operating experiences of domestic NPPs, the environmental emissions of liquid effluents were mainly caused by the design characteristics of the LWMS rather than the difference of NPP power load [8]. In other words, it is possible to reduce the amount of waste generated according to the design characteristics of the LWMS and the operation characteristics of the main equipment. Accordingly, it is necessary to develop newly improved technology more for the safety of the general publics even though the LWMS has reduced radiation emissions through continuous design improvements. In addition, the LWMS in the APR 1400 generates a large amount of spent ion exchange resins, which are not easy to process and dispose. Therefore, it is necessary to develop and introduce new technologies that can solve present and future difficulties in NPPs operation.

2.2 Operation of the LWMS in APR 1400

The LWMS in the APR 1400 is equipped with liquid waste collection tanks, which include a floor drain tank (FDT), an equipment waste tank (EWT), and a chemical waste tank (CWT), process filters, reverse osmosis package, process pumps, monitoring tanks, and a remote operation control facility. The main subsystems of the LWMS are composed of the floor drain subsystem, the equipment wastewater subsystem, the chemical wastewater subsystem, and the radioactive laundry subsystem.

Floor drainage of reactor buildings and auxiliary buildings is sent to the floor drainage subsystem, auxiliary building equipment drainage and the equipment wastewater are sent to the equipment waste subsystem, and chemical waste water generated during operation is sent to the chemical wastewater subsystem and processed by a reverse osmosis package. The main processing equipment of the APR 1400 LWMS is a reverse osmosis package, which consists of a pre-treatment module, a reverse osmosis module, a demineralizer module (one cation bed and two mixed bed), and a concentrate treatment facility. The pre-treatment module is equipped with oil removal filters and microfilters to remove particles and organic materials in collected waste water to maintain the performance and safety of the reverse osmosis membrane, and finally transfer the processed waste water to the reverse osmosis module. The reverse osmosis module, which is the main processing equipment of the reverse osmosis package, separates the permeated water and concentrate from the pre-treated waste liquid through several stages of reverse osmosis to remove soluble salts and radioactive ions which have not been removed from the pre-treatment module. The permeated liquid is discharged to monitoring tank, and concentrate is discharged to the concentrate treatment facility. The permeated liquid from the reverse osmosis system is discharged to the monitoring tank after additional radioactive ions (Co, Sb, Cs) are removed from the demineralizer module consisting of three ion exchangers (one cation bed and two mixed beds). According to the result of sampling, the treated liquid in the monitoring tank is transferred to the circulating water discharge conduit or discharged to the plant to reuse. If the liquid in the monitoring tank does not meet effluent concentration limit and water quality, it is discharged to R/O package for additional re-processing [2].

3. Optimization of Application of Ion Crystallization Technology

3.1 Effluent Concentration Changes in the LWMS Using Ion Crystallization Technology

Domestic NPPs have continuously improved the design of the LWMS to improve performance and operating efficiency. As a result, the annual solid radioactive waste generation rate is decreased as the performance of the system is improved [1]. However, as a result of the operation of the LWMS, a large amount of waste water will be generated continuously, and the development of new technology is necessary considering the limited storage capacity of domestic disposal facilities. Therefore, this paper analyzes the changes in the radionuclide concentrations in each LWMS process, assuming the application of ion crystallization technology, and proposes an optimal application location considering the configuration and design characteristics of the existing system. The considerations and assumptions for this are as follows:

- The nuclides considered in this analysis were Cs, Co, Mn and Fe as studied in the early study [4], and the design basis liquid effluent concentration values of EWT provided by FSAR were used.
- The application location was chosen on the premise that the existing design is changed as little as possible. As a result, the application location was selected as EWT itself, after the pre-treatment module, and after the R/O module.
- The experimental results of the early study [4] are applicable to the LWMS process.
- Based on the results of the early study [4], the radioactivity removal rate of the nuclear species ion crystallization technology was at least 95%. However, 90% of radioactivity removal rate was conservatively applied regardless of the nuclide.
- The equations used in the radioactivity removal rate are as follows.

$$\text{Radionuclide removal rate} = \frac{\begin{array}{c}(\text{Radionuclide concentration before process}\\ - \text{Radionuclide concentration after process})\end{array}}{\text{Radionuclide concentration before treatment}}$$

Table 2 shows the results of calculating the radionuclide

Table 2. Estimation of effluent concentration in each process by applying ion crystallization technology to the LWMS in the APR 1400 [2, 4]

Tank	Nuclide	Concentration	R/O Module Inlet Stream			R/O Module Outlet Stream			Demineralizer Outlet Stream		
			ⓐ	ⓑ	ⓒ	ⓐ	ⓑ	ⓒ	ⓐ	ⓑ	ⓒ
Equipment Waste Tank	^{51}Cr	7.89×10^{1}	7.89×10^{0}	7.89×10^{0}	7.89×10^{1}	7.89×10^{-1}	7.89×10^{-1}	7.89×10^{0}	7.89×10^{-5}	7.89×10^{-5}	7.89×10^{-5}
	^{54}Mn	9.13×10^{0}	9.13×10^{-1}	9.13×10^{-1}	9.13×10^{0}	9.13×10^{-2}	9.13×10^{-2}	9.13×10^{-1}	9.13×10^{-6}	9.13×10^{-6}	9.13×10^{-6}
	^{59}Fe	1.71×10^{0}	1.71×10^{-1}	1.71×10^{-1}	1.71×10^{0}	1.71×10^{-2}	1.71×10^{-2}	1.71×10^{-1}	1.71×10^{-6}	1.71×10^{-6}	1.71×10^{-6}
	^{58}Co	2.62×10^{1}	2.62×10^{0}	2.62×10^{0}	2.62×10^{1}	2.62×10^{-1}	2.62×10^{-1}	2.62×10^{0}	2.62×10^{-5}	2.62×10^{-5}	2.62×10^{-5}
	^{60}Co	3.02×10^{0}	3.02×10^{-1}	3.02×10^{-1}	3.02×10^{0}	3.02×10^{-2}	3.02×10^{-2}	3.02×10^{-1}	3.02×10^{-6}	3.02×10^{-6}	3.02×10^{-6}
	^{134}Cs	2.03×10^{3}	2.03×10^{2}	2.03×10^{2}	2.03×10^{3}	2.03×10^{1}	2.03×10^{1}	2.03×10^{2}	2.03×10^{-3}	2.03×10^{-3}	2.03×10^{-3}

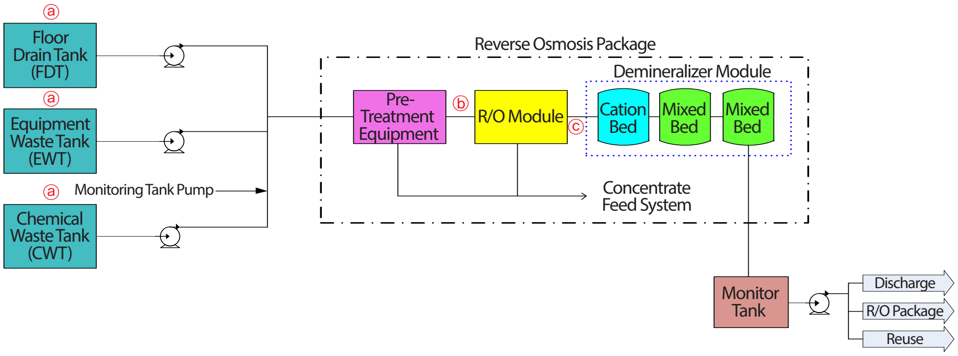

Fig. 2. Schematic diagram of the LWMS in the APR 1400.

concentration of each nuclide in each process when ion crystallization technology is applied to the existing LWMS in the APR1400. The removal rate of each component is based on the FSAR [2] and the result of the early study [4]. The Fig. 2 is the schematic diagram of the LWMS in the APR 1400. As shown in Fig. 2 and Table 2, the application locations ⓐ and ⓑ were found to have a lower concentration of liquid effluent flowing into the R/O than the application position ⓒ. In the final stage of the LWMS process, the radionuclide concentrations of all application locations were found to be the same. This is because the removal rate of nuclides in each process was applied in the same manner regardless of the application location of the ion crystallization technology. In other words, the application location of ion crystallization technology should be applied in consideration of the advantages and disadvantages of the existing design status and characteristics, not the degree of the radionuclide concentration changes at this time.

3.2 Optimization of Application Location of Ion Crystallization Technology

The LWMS processes equipment wastes, chemical wastes and floor drain wastes from reactor containment building, auxiliary building, and compound building. In addition, during the period when leakage occurs from the primary side to the secondary side in steam generator tube, the LWMS processes liquid wastes generated from the turbine building. The influx source of the LWMS is classified into three subsystems that are equipment waste including relatively high purity waste, floor drain waste including low purity waste, and chemical waste including various chemicals. The purpose of classification into three subsystems is to enable more efficient processes by waste type and to reduce solid waste [2]. In other words, a large amount of potential waste with little or no contamination can be monitored and discharged after a minimum process, resulting in a reduction in the operating load on the system. Another design feature of the APR 1400 LWMS is to minimize and effectively reduce effluent releases using reverse osmosis membranes. We believe that it is desirable to maintain these design features as much as possible because the design of NPPs is the result of many years of operating and design experience. The followings are the considerations for applying ion crystallization technology to the existing LWMS in the APR 1400:

- Application location
- Removal efficiency
- Minimize design changes (maintaining existing facilities and processes as much as possible)

Among the three mentioned considerations, the application location is considered to be the most important factor. This is because the application location of ion crystallization technology has a great influence on the application effect of new technology and existing design. In other words, while the process is improved, the effect on the existing LWMS should be minimized, and the installation space of the site should be considered. Considering the current configuration of the LWMS in the APR 1400, there are three locations where ion crystallization technology can be applied. The application locations are three liquid waste collection tanks (FDT, EWT and CWT), after the pre-treatment facility, and after the R/O module.

3.2.1 At the collection tanks (ⓐ in Fig. 2)

The first candidate location for the application of ion crystallization technology is the collection tank(s) itself. Liquid radioactive materials generated during NPP operation are collected in liquid waste collection tanks and then processed. The liquid radioactive materials collected in the tanks are not processed at all, and the radioactivity level is relatively higher. Therefore, the application of the ion crystallization technology to the liquid waste collection tank is expected to have the highest nuclide removal rate and the highest effect compared to other locations. Another advantage of this location is that it can be selectively applicable as needed among the three tanks. It can result in performance improvement while maintaining the characteristics of the existing design.

Only a part of the existing collection tank design might be changed and the installation space is unlikely to be affected. Thus, the impact on existing designs is the least compared to other candidate locations. In addition, further analysis and research may also be used to select tanks to which the ion crystallization technology is applied, as needed. When the ion crystallization technology is applied to the LWMS, the particles of the inorganic solids generated by selective removal of the nuclides in the wastewater are larger than 1μm. These particles will be adsorbed in the pre-treatment module which is equipped with microfilter installed after the collection tanks. The operating load of microfilter, which is a part of pre-treatment module, would be slightly increased, but the effect of load increase is not expected to be significant because microfilter is applied to the backwashing method. In addition, the removal of various nuclides included in wastewater in front of the R/O will help reduce scaling and fouling, which are typical problems in the reverse osmosis equipment. In other words, the operating efficiency of the R/O will be increased and the lifetime of ion exchange resin will be increased.

3.2.2 After Pre-treatment module (ⓑ in Fig. 2)

The pre-treatment module of the APR 1400 LWMS is the component that prevents contamination, scaling and damage of the reverse osmosis module. In terms of the location ⓑ, the radioactivity change of the liquid waste according to the application of the ion crystallization technology is analyzed as the same as the first candidate location.

Table 3. Comparison of application effects for three selected locations

Application Location	Advantages	Disadvantages
At the collection tanks (ⓐ in Fig. 2)	- Highest removal rate - Least impact of existing design	- Required more cost when applied to three tanks. But it can be selectively applicable among three tanks.
After Pre-treatment module (ⓑ in Fig. 2)	- Higher removal rate - Improve R/O operating efficiency	- Less application effect - Increase of additional device
In front of Demineralizer Module (ⓒ in Fig. 2)	- Reduce spent resin generation	- Least application effect - Increase of additional device

Removing the nuclides in waste water by ion crystallization technology results in the formation of inorganic solids. When the ion crystallization components are located after the pre-treatment module, the number of additional devices needed such as tanks and filters will be increased, and installation may be difficult floor space limitations in the plant. The impact on the existing design will be somewhat increased, and it is inevitable to change the general arrangement design.

3.2.3 In front of Demineralizer Module (ⓒ in Fig. 2)

The last candidate location for the application of ion crystallization technology is after the reverse osmosis module, which is the main treatment component of the LWMS. Since the process is performed at the reverse osmosis module in front of ion exchangers, the application effect of the new technology is not expected to be greater than other candidate locations. Also, it is not helpful in solving the typical R/O module problem such as scaling. From an installation space point of view, new components such as tanks and filters are added, so design changes will be increased. The applicability at this location is the lowest compared with other candidate locations mentioned above.

3.3 Results

The study on the application of ion crystallization technology to the APR 1400 LWMS was performed. It was found that the removal rate of nuclides and the range of design change depend on the application location. When the ion crystallization technology is applied to the waste collection tank, the amount of the required components such as a tank and a filter would be less. It is also expected that the application effect and removal rate of nuclides would be higher than other locations. Table 3 shows the comparison of application effects for three selected locations.

4. Conclusion

In order to reduce liquid radioactive effluents discharged to the environment and solid waste, the design and components of the LWMS of domestic NPPs has been improved. Nevertheless, further efforts to improve system design should continue for operational considerations and protect general public and the environment. In particular, in an earlier study, the amount of liquid effluent discharged to the environment was mainly caused by the treatment process of the LWMS [8]. Accordingly, it is necessary to apply the improved technology or new processing method.

In this paper, the possibility of applying ion crystallization technology with a highly selective decontamination rate to the APR 1400 was investigated. To confirm this, the radioactive concentration data of radioactive liquid wastewater in the FDT of Shin-Kori units 3 and 4 were used and the decontamination rate was also estimated. In addition, since the ion crystallization technology has high decontamination rate, it is applicable to the existing LWMS or future NPPs and this study showed the possibility of optimizing existing facilities. In addition, optimization of the application location of the ion crystallization technology was derived. As a result, it was interpreted that applying the technology to the collection tank itself, which has a relatively high radioactivity level among the process, has the most advantageous in terms of efficiency and applicability.

Since the result of this study is based on design requirement and operating experiences, it will be more meaningful if the decontamination rate test is carried out using the actual liquid waste in NPPs. We hope that the actual test in the field will be performed soon.

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Abbreviation

AOOs	Anticipated Operational Occurrences
APR 1400	Advanced Power Reactor 1400
CWT	Chemical Waste Tank
EWT	Equipment Waste Tank
FDT	Floor Drain Tank
FSAR	Final Safety Analysis Report
LWMS	Liquid Waste Management System
NPPs	Nuclear Power Plants

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