

STATUS OF WASTE MANAGEMENT TECHNOLOGY IN PNC, JAPAN

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ABSTRACT

On the basis of the results of research and development activities to date, PNC has constructed and operated facilities demonstrating the technologies to process waste into stable forms which include a Bituminization Facility, Plutonium Contaminated Waste Treatment Facility and Waste Dismantling Facility. TRU waste generated from the Tokai Reprocessing Plant (TRP) and Mox fuel fabrication facilities has been conditioned into bitumen, plastic, ceramic-like block, etc. in these waste treatment demonstration facilities. The technology development on separation of radioactive nuclides from TRU waste is under way aiming volume reduction of the TRU waste markedly. Regarding to the vitrification technology, PNC has been constructing the Tokai Vitrification Facility which will start hot operation in early 1992. The facility is designed to vitrify HLLW from TRP with liquid fed ceramic melter (LFCM) process. R&Ds concerning HLW treatment with advanced technology, separation and fixation of gaseous waste such as Kr, Xe are also being developed. In accordance with the policy of Japan's Atomic Energy Commission, PNC has been conducting research on the concept of geological disposal of vitrified high-level radioactive waste and on methods for enhancing feasibility of the technology. As a result, the first-stage, R&D on identification of a probable geological formation was completed in 1984. PNC is now conducting research to: (a) develop a data bank on the geochemical and geophysical characteristics of Japanese geological formations, together with long-term predictions on changes in those formations; (b) develop engineered barrier system; (c) determine long-term prospects for engineered and natural barriers through the use of natural analog surveys; (d) develop performance assessment technology to improve safety prediction for geologic disposal system; and (e) take the first step in the design of a geological disposal system.

INTRODUCTION

The maturity of the nuclear fuel cycle has cast a spotlight on the serious problem of the disposal of radioactive wastes. Power Reactor and Nuclear Fuel Development Corporation (PNC) is actively engaged in the research and development on all the areas in the nuclear fuel cycle except those of light water reactors and uranium fuel fabrication, and these R&D activities inevitably produce a wide variety of wastes. Most radioactive wastes that are generated as byproducts of nuclear fission reaction are separated in liquid form as high-level radioactive waste during the reprocessing stage. High-level liquid waste is solidified into a glass forming vitrified waste which is physically and chemically stable. The high priority tasks in the area of high-level radioactive waste management at this point in time are development of radioactive waste treatment technologies, such as an efficient and effective vitrification technology, and development of a radioactive waste isolation technology that establishes the proper procedure for disposing vitrified waste.

The Japanese government is working toward establishment of a technology to utilize plutonium as a source of energy, and this type of nuclear power system will inevitably generate plutonium and other TRU wastes as secondary waste material in various stages of the nuclear fuel cycle. Safe control over TRU wastes is essential, but what must be understood in this area is that most TRU wastes are manifested as contaminants on machinery and other facility

hardware and what is needed is an emphasis on development of a set of radioactive waste treatment technologies such as radioactive waste decontamination, volume reduction and stabilization technologies which reduce the amount of radioactive wastes generated.

The basic concept behind geological disposal of radioactive wastes is to ensure that future generations will not be harmed by radiation from high-level radioactive wastes. To meet this requirement, the geological disposal process requires cooling high-level radioactive wastes that have been solidified into a stable form for a certain period of time and then properly burying the solid wastes deep geological formation.

The goal of the geological disposal research and development program is to clearly demonstrate the safety of this procedure. In other words, the feasibility of geological disposal of radioactive wastes must be demonstrated by developing a procedure for safe disposal of radioactive wastes underground and the technologies necessary for putting this procedure into practice. Furthermore, geological disposal of radioactive wastes must be done only after making sure that the people who receive the benefits of nuclear energy believe in the safety of geological waste disposal and concur with the procedures. Thus, the geological disposal research and development program must be implemented with emphasis on demonstrating the

feasibility of the geological disposal process and forming public acceptance for the process.

This report describes the basic concepts and approaches being taken in the research and development activities on radioactive waste treatment and isolation technologies, along with brief summaries of the present status and future issues.

RESEARCH AND DEVELOPMENT ON WASTE TREATMENT TECHNOLOGY

R&D Themes

During the course of PNC's routine activities, a wide variety of wastes are produced, for example, as a result of the processing of about 400 tons of spent fuel in the Tokai Reprocessing Plant (TRP) and the operation of plutonium fuel fabrication facilities at Tokai Works. The amount of the high-level liquid waste produced in the TRP is relatively small (about 1 m³ per one ton of spent fuel), it contains more than 99% of the radioactivity originally contained in the spent fuel. It also contains transuranium elements (TRU radionuclides) which have long half-lives and thus require a long-term isolation, as well as TRU wastes which are produced as much as ten times as the high-level liquid waste. TRU wastes contain as much TRU radionuclides as high-level liquid waste, though it contains less radioactivity.

The principles of the Atomic Energy Commission in Japan with regard to the treatment of radioactive wastes can be summarized as follows;

1. Efforts must be made to reduce the amount of low-level radioactive wastes generated and to process the wastes that are generated to reduce their volumes and to solidify them into a stable form.
2. After they are solidified into a stable form, high-level radioactive wastes must be cooled in storage for 30 to 50 years and then disposed of deeper than several hundred meters from the surface.

PNC, in recognition of the JAEC policies and the fact that PNC itself generates radioactive wastes, is making a great effort to ensure safe storage of radioactive wastes as well as to develop the radioactive waste treatment technologies for waste material volume reduction and solidification. So far, PNC's R&D achievements have enabled construction and, in some cases, operation of facilities such as a bituminization facility, plutonium-contaminated waste treatment facility and vitrification facility that emphasize stabilization processing, which require stability and ease of handling of the processed wastes. In short, PNC has nearly achieved its goals to provide stable operation of fuel cycle facilities such as a reprocessing plant.

PNC considers as its future task for radioactive waste treatment R&D that enables transfer of wastes that have been treated by stabilization processing to the isolation

disposal process and R&D that reduces the load on radioactive waste disposal processing by significantly reducing the amount of radioactive waste generation and other means.

PNC is striving for efficiency in its R&D activities through cooperation with foreign research organizations. At the same time, PNC considers it vital to cooperate with the private sector, as it demonstrated by providing the vitrification technology it developed for use by Japan Nuclear Fuel Services Corporation in its commercial reprocessing plant.

Status of R&D on High-Level Waste Treatment Technology

PNC began R&D on solidification of high-level liquid wastes in 1975 at its Tokai Works, and has performed engineering and mockup tests thus far. In December of 1982, PNC began vitrification tests using actual waste. These tests have involved building a 1/100th scale model of the Tokai Vitrification Facility (TVF) to produce high-level vitrified wastes to demonstrate the vitrification process and to perform characteristics analysis on the vitrified wastes produced.

PNC began construction of the Tokai Vitrification Facility on June 29, 1988. This facility is based on the R&D described above and its purpose is to demonstrate the vitrification process with the ceramic melter (LFCM) method. This facility is scheduled to begin hot operation in 1992.

The LFCM method developed by PNC has been adopted by Japan Nuclear Fuel Services Cooperation in its commercial reprocessing plant, and PNC intends to continue to provide technical support and cooperation to the private sector. Increasing the life and reducing the size of the glass melter, which is the core of the radioactive waste vitrification process, lead to improving the economy and reducing the amount of secondary waste generation. For this reason, PNC is developing refractory materials and a new heating process to produce a high-performance but compact melter.

Status of R&D on Low-Level Waste Treatment Technology

PNC is developing low-level liquid waste treatment technology, bituminization for low-level condensed waste and plastic solidification for spent solvent, through hot operation at waste treatment facilities. As the results of the hot operation, 14,000 drums of bitumen and 500 drums of plastic have been produced by August, 1989.

Characteristics analysis plays an important role to study low-level waste disposal system. PNC intends to implement

tests for the characterization of hot solidified wastes in this year, including the asphalt solidified waste already treated.

In the area of the low level waste, appropriate disposal policies will be established taking into account of the characteristics of wastes, and for this end, PNC is engaged in the development of non-destructive measurement technologies for the identification and the measurement of amounts of radionuclides contained in solidified wastes. This technology is essential for the establishment of the basic database of the source term. Development activities of non-destructive measurement technologies include the development of the passive gamma method which measures and analyzes the gamma ray emitted from the nuclides contained in solidified wastes, and the passive neutron method which measures and analyzes the neutron emitted from the radionuclides. PNC is also planning to develop the active neutron method aiming at achieving the accuracy of about several n Ci/g plutonium.

Low level radioactive wastes, in particular TRU wastes, can be treated as non-radioactive wastes after the extraction of radionuclides contained in the wastes. This leads to a considerable reduction of the amount of TRU wastes, to be disposed of.

PNC is now engaged in the development of the technology for achieving a high decontamination factor of radionuclides contained in low-level condensed liquid waste and spent solvents from reprocessing. We are conducting cold tests for the technology of separation of radionuclides using the co-precipitation and ultrafiltration method and the ion exchange method for the condensed liquid waste, and for waste solvents, tests for the technology of adding a hydrogen peroxide solution and catalysts to TBP and heating the mixture to oxidize and decompose the organic matters followed by the separation of radionuclides in the resultant phosphoric waste liquid using the co-precipitation method. The prospect of waste volume reduction ratio is about one hundredth.

TRU solid wastes from Tokai Reprocessing Plant has been stored at waste storages in PNC Tokai Works. Aiming at stabilization of these waste and conducting hot engineering test for above mentioned nuclides separation technology, Construction of the Low-Level Waste Treatment Facility is planned.

Regarding to radioactive gaseous waste, krypton recovery technology using cryogenic method has been developed. Through the hot test operation of recovery process, 2,000 Ci of krypton gas has been recovered. In order to store recovered krypton gas in a stable form, PNC has conducted

cold tests on immobilization technology using zeolite or ion plantation technique.

Advanced Technology Development

As a long-term basic study, PNC has started research on separating exothermic nuclides and reducing the burden on high-level radioactive waste disposal by forming solid wastes in more highly reduced volumes. The reasons for this research are that each vitrified high-level radioactive waste generates approximately 1.4 kW of heat and must be stored during the cooling process and that each vitrified waste can contain only 20 to 30% by weight of the radioactive components of high-level radioactive liquid waste. As a part of this research, recognizing that high-level liquid waste contains a large amount of rare metal elements, PNC researchers are working on heating calcine of high-level liquid waste to several thousand degrees centigrade to separate the elements with low boiling point including cesium and other exothermic nuclides by sublimation and volatilization and, at the same time, solidify the remaining TRU nuclides into higher reduced volumes of double oxides with rare metal elements.

So far, this study has provided an indication that a higher reduced volume and stable form that is only one tenth the volume of the conventional vitrified form can be obtained from this process.

Along with krypton recovery, xenon, one type of rare gas, is also recovered. In view of the fact that isotopes of xenon have an extremely short half life, the level of its activity being reduced in a short period of time to a negligible level, and that this is a valuable gas used for a powerful lighting source (about 2 million yen per 1 m³), PNC is engaged in the research and development with the view to highly purifying xenon. Each ton of spent fuel from a light-water reactor contains approximately 3 kg of noble metals such as ruthenium and palladium, and an FBR generates approximately 1.7 times more noble metals at the same combustion level. In recognition of this fact, plan using is underway to study the possibility of recovering noble metals from the high-level liquid waste as a part of the OMEGA Project. Currently, a process is under study to recover ruthenium, which comprises 60% of the noble metals in high-level liquid waste, by oxidization using an electrolysis technique. Another process is being studied to deoxidize noble metal oxides into a metal state and separate the

product by heating calcine of high-level liquid waste to high temperature in a reducing atmosphere.

RESEARCH AND DEVELOPMENT ON GEOLOGICAL DISPOSAL

Basic Policy of R&D on Geological Disposal:

The following are various high-level radioactive waste disposal options which have been studied, extensively.

1. Geological disposal
2. Sub-seabed disposal
3. Very deep hole disposal
4. Ice-sheet disposal
5. Space disposal

Except for the underground disposal, most of those options are in the stage of concept assessment and/or basic research and development with regard to the safety assessment and technology.

The geological disposal method is concerned internationally as the most practical and realistic opinion. This scheme is considered as the most attractive target among most of the nations. Major advantages of the geological disposal are summarized below.

1. Radioactive waste can be effectively isolated from human and his environment.
2. To ensure safety, there is no need for human control of the waste after disposal.
3. Each nation can manage the waste within its own territory (under its own responsibility).
4. For the construction of a geologic repository, existing civil engineering and mining technology can be applied to maintain reasonable construction costs.

Japanese policy concerning R&D on geological disposal is as follows:

1. Considering a research and development program on geological disposal is time-consuming and comprehensive, it should be implemented in a consistent and flexible manner.
2. Research and development should be carried out on the variety of multibarrier systems in which accommodate the diverse condition of geological environment in Japan.
3. Safety of geological disposal depends much upon near field performance. On the other hand, for field performance provides an extra margin of safety. From this standpoint, priority should be given to research and

development on the near-field while steadily advancing research on the far-field phenomena.

Power Reactor and Nuclear Fuel Development Corporation (PNC), which is the central institution responsible for research and development, should compile progress reports at appropriate times to provide information to the general public for their understanding on geological disposal. These reports should be evaluated by competent authorities for further implementation of the research and development program.

How to Assure the Safety of the Geological Disposal

The basic philosophy for assuring the safety of the geological disposal of high-level radioactive waste is summarized below.

1. Although radioactivity of high-level waste is high at the time of its generation, the level of radioactivity of most nuclides decays rapidly to a lower level after several hundred years. Complete containment should be guaranteed for such period long enough for short lived nuclides to decay. For this purpose, measures should be provided to reduce the possibility of groundwater coming into contact with the waste.
2. Less radioactive and long lived nuclides will dominate the radioactivity of the waste after several hundred years. For this period, it is important that such radionuclides remain within the near field. For this purpose, measures should be provided to prevent the radionuclides from being dissolved and subsequently transported by groundwater even if groundwater comes in contact with the waste.
3. Radioactivity of nuclides will fall to the level far below the background during such a period and thereby no significant radiological impact on human environment will occur.

Important Aspects in Japan

Natural environment in Japan can be summarized as relatively small-scale, complicated geological structure; and frequent distribution of groundwater. To establish geological disposal system in Japan, it is necessary to prove that extremely long-term safety can be maintained, and to prove the technical feasibility of the multibarrier system under such conditions.

It is impossible, however, that the safety of the geological disposal system can be directly demonstrated in real time scale due to the extension of time considered in this issue. Therefore, research and development has been conducted on geological disposal to prove the important factors by means of Performance Assessment.

The present research and development has been conducted in accordance with the policy that a multibarrier

system can be constructed given the proper geological environmental conditions reflecting the fact that candidate host rock for the repository has not been defined. This will determine the probability of geological disposal in both scientific and technological sense. In other words, the present target is to focus on the development of a multibarrier system widely applicable to a variety of geological environment.

Knowledge to Date

Fundamental understanding on the geological disposal system has been accumulated on the basis of research and development results to date.

1. Engineered barrier effect

Even when the groundwater flow in the vicinity of the engineered barrier is considerable, a static flow is maintained inside the barrier because of the buffer materials.

The candidate overpack materials test showed that an anticorrosive container that will last more than 1,000 years can be designed providing the oxygen content of groundwater is lower than certain level.

Almost all nuclides contained in the waste are highly resistant to groundwater dissolution when vitrified.

Even if groundwater with a higher oxygen concentration may exist in the near field, it may be converted into water with lower oxygen concentration as a result of reaction with the overpack or buffer materials.

It is predicted that most radioactive nuclides can be remained within the near-field for more than 10,000 years.

2. Natural barrier

Groundwater of deep underground generally has a property and condition to be suitable for lower solubility of radioactive nuclides and glass.

The retardation effect of the natural barrier is expected to last for a longer period than in the engineered barriers described above which provides an extra margin of safety. This is based on the results of surveys and research in the uranium deposit and the study of the reaction of rocks to radioactive nuclides. (Natural Analogue)

3. Geological environment in Japan

The geological and mechanical properties of bedrock in Japan have been assessed from various data analysis.

As a result of an aerial photography analysis, the properties of active faults and their distributions have been studied on a nationwide scale.

Major R&D Projects of PNC

The major projects being carried out by PNC are summarized as below.

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1. Performance assessment research

PACE program (integrated near-field performance assessment study) (PERFORMANCE ASSESSMENT CENTER FOR ENGINEERED BARRIERS)

The integration of assessment codes for the record of geochemical conditions in the near field of radioactive waste; corresponding individual phenomena such as the corrosion of containers, the solution and migration of nuclides; and the development of an assessment code for the release of nuclides through an engineered barrier. Thermodynamic-data acquisition is underway regarding the solubility of actinides.

ENTRY program (Engineering Scale Test and Research Facility)

The overall objectives of the ENTRY Program are to conduct relatively large-scale nonradiogenic, laboratory experiments and model development to support performance assessment for a high-level waste repository. The ENTRY Program scope is broad to include the following activities related to the performance assessment of geological disposal.

development, evaluation, and validation of performance assessment models

collection of nonradiogenic laboratory data

development of data base for performance assessment

communication of scientific results to the technical community and public.

2. Geo-scientific research

From the geo-scientific viewpoint, several deep subterranean phenomena, such as underground hydrology, geochemical phenomena, nuclide migration, rock mechanics, and thermal behavior have been studied.

KAMAISHI IN-SITU EXPERIMENT

An integrated In-Situ experiments in granitic rocks at the Kamaishi iron mine.

TONO SHAFT EXCAVATION EFFECT STUDY

A study of the shaft-excitation effect on hydrology and rock mechanics in the vicinity of a shaft excavated in sedimentary rock at the Tono Mine.

3. Geological environmental research

Surface and subsurface geological investigation have been executed. Potential geological formations are classified into areas with common properties of geological environments from the viewpoint of type of rocks, age of rocks, structural features, etc. Geological data base system has been developed for data applications obtained from these activities. The data base will facilitate effective assessment of the characteristics of the geo-

gical environment in Japan, and to provide a model data set for performance assessment.

4. International cooperative research

In addition to the above, PNC is involved in the international and bilateral collaboration as below.

- OECD/NEA International Stripa Project
- OECD/NEA International Alligator Rivers Analogue Project
- AECL Canada
- SCK Belgium
- NAGRA Switzerland
- PNL US

CONCLUSION

PNC is conducted research on management of high-level radioactive wastes by employing the unique capabilities of its numerous divisions and plants. The Tokai Works is the center of PNC's R&D on radioactive waste treatment

techniques, and it has a vitrification technology development facility and plutonium waste treatment technology development facility. The Tokai Works is also central to PNC's research on geological disposal of radioactive wastes because it has the real high-level radioactive wastes and TRU wastes for use in such research. The Chubu Works is working primarily on geo-science research using the knowledge it has gained through uranium resource development and with the cooperation of the Nihon Pass Works. The Oarai Engineering Center is working on developing the decommissioning technology. More than 200 engineers are currently working in this area, and PNC intends to increase professional staffing to further its vitrification technology development and research on disposal technology.

The technology and policy for managing high-level radioactive wastes is extremely important for the safe use of nuclear energy, and PNC is conducting R&D in this area with full understanding of the characteristics of high-level radioactive wastes and with the aim of maximizing safety for people and protecting the environment.