THE EFFORTS TO UTILIZE HIGH-TEMPERATURE MELTING TECHNOLOGIES FOR LILW AND THE DEVELOPMENT OF GUIDELINES FOR THEIR TECHNICAL ASSESSMENT

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ABSTRACT

A couple of domestic institutions have been investigating the application of vitrification technology to treat low- and intermediate-level radioactive wastes in Korea. In the case that such investigations prove to be successful, it is expected that commercial vitrification plants will be constructed. The safety insuring on vitrification plants could not be compatible with criterion on radioactive waste management because the facilities are at high temperature and contain a variety of accommodations for the exhaust gases and residual products. Therefore, it is necessary to suggest a new strategy or modifications of criterion of radioactive waste management on considerations related with the vitrification technology. In order to ensure the safety of vitrification plants, a technical guideline or standard for design and operation of vitrification plants must be established too. A study on the safety assessment of vitrification plants in consideration with general items as an industrial facility, safety and technical requirements as a nuclear facility is needed to be ready before using and permitting them. Also, the stability of vitrified waste forms produced by vitrification plants must be analyzed to ensure their acceptance in final repositories, which includes chemical durability as one of the main considerable items. This paper introduces the status on the utilization of vitrification technology for treating LILW and efforts to develop technical guides with basic study results on chemical durability of forms.

INTRODUCTION

It is very important to develop a more stable technology with high reduction efficiency on volume so that environment regulation is gradually intensified with regard to radioactive waste management. The public aversion towards waste repository places the government in a difficult situation with regard to the selection of waste disposal areas, and then, increases the cost of waste management. At this point, vitrification technology with a high reduction rate of volume and high stability of waste product forms could be a prospective alternative to conventional treatment methods such as incineration and solidification and has been utilized for safe disposal of radioactive wastes even containing low-level activity.

Vitrification technology with its high reduction in volume and high stability of vitrified waste forms has been developed for safe disposal of high-level radioactive wastes and extended to use in low-level wastes in some countries. A couple of domestic institutions in Korea have studied the application of vitrification technology to treat low- and intermediate-level radioactive wastes.

If such studies prove to be successful, it is expected that commercial vitrification plants will be constructed. In order to ensure the safety of vitrification plants, a technical guideline or standard for design and operation of vitrification plants must be established. Therefore, a study on the safety assessment of vitrification plants in consideration with general items for industrial facilities, safety requirements as a nuclear facility, and technical requirements for vitrification plants are needed for the establishment of them.

Also, the stability of vitrified waste forms produced by vitrification plants must be analyzed from the assessment of physical and mechanical properties and chemical durability.

In this study, the technical and regulatory status of radioactive waste vitrification technologies in foreign and domestic operations is investigated, and then factors for consideration are suggested which must be contained in the technical guideline or standard for the safety assessment of vitrification plants. Some related efforts and results of basic studies in the course of developing a guideline are also covered by showing a brief overviewed summary of research.

EFFORTS TO UTILIZE MELTING TECHNOLOGIES FOR LILW

Melting solidification or vitrification is considered to be a prospective technology for stabilizing low-level radioactive waste since it has the advantage of improving the physicochemical properties of waste forms.

High temperature treatment systems enable the following features:

- Complete waste material breakdown and inherent thermal separations (slag, reduced metals, minimal off-gas) and production of homogenous products
- Destruction of hazardous organics
- Large volume reductions of solid waste
- Immobilization of actinides and oxidized metals in nonleachable geologically durable glass/ceramic final waste forms
- Robust processing, with wide ranges of feed acceptance and process variation tolerances

Because of the above-described advantages of vitrification or melting technologies, the following two pilot plants aim to produce better waste forms and have been constructed in Korea. Each is introduced with a brief summary.

A Pilot Plant of Plasma Arc Melter

One pilot plant is the Plasma Arc Melter (PAM) system using graphite electrodes designed and fabricated by MeltTran, Inc., USA and was constructed by "D" company. The PAM was installed near the incineration plant at the Korea Atomic Energy Research Institute site, Korea during 1997-1998.

Trial burn test has the primary objective of demonstrating the melter applicability to treatment of radioactive low-level waste and mixed waste from Korean nuclear power plants. The PAM facility uses a graphite electrode arc melter vitrification system to treat low-level radioactive waste [1, 2]. Figure 1 shows a schematic process diagram of PAM system. The waste feed material is processed through a shredder and fed into the melter.

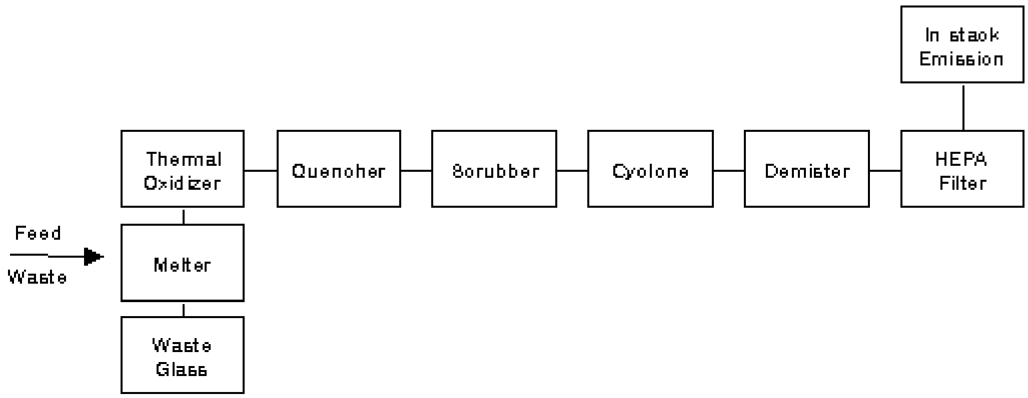

Fig. 1 Flow diagram of pilot-scale vitrification plant PAM

The molten glass inventory in the bottom of the melter provides the heat for dissociation and combustion of organics. Inorganic residues from the combustion process are melted and combine in solution with the glass ingredients. Periodically, the molten glass is tapped into drums where it solidifies forming the leach resistant disposal waste form. The combusted gases pass through an air pollution control system for removal of particulates. The system includes a waste feed system, a 200kg/h melter, a thermal oxidizer, off gas treatment system, monitoring and control systems, and a variety of auxiliary support systems such as cooling water as follows;

- Waste preparation and feed system
  System acceptance of 200liter drums containing combustible or non-combustible waste with no sorting
- Melter system
  The melter system consists of a carbon steel walled chamber containing two graphite-electrodes for arc melting. It is located immediately below the thermal oxidizer chamber. The melter and thermal oxidizer chambers are designed to minimize particulate deposition and fouling. A rammed or a pre-cured refractory brick is provided to simplify refractory replacement and compatibility with different melt compositions
- Off-gas system
  The off-gas quench/scrub system quenches the thermal oxidizer exhaust gas by contacting the gas directly with scrubbing solution in the quench chamber. This action cools the gas rapidly thereby limiting the formation of dioxins.
- Control and data acquisition system
  The major principal purposes of the control and data acquisition system are monitoring and controlling the melter electrodes, the plant and ancillary systems, electronically collecting data and recording the operation of the melter systems, and to provide automatic safety and interlock related functions.
- Auxiliary system

A Combined System of Plasma Torch Melter and Cold Crucible Melter

The pilot plant of combined system constructed and investigated by "N" institute consists of a Plasma Torch Melter (PTM) using plasma torch PT-150C designed by Callidus Technology Inc., USA and a Cold Crucible Melter (CCM) in cooperation with SGN Inc., France. The pilot plant was installed at Taejon, Korea for treating various types of low-level radioactive waste generated from nuclear power plants. The trial burn was undertaken from 1999. In the event that such trial burn tests prove to be successful, it is expected that commercial vitrification plants will be constructed about 2004. Trial burn test have the primary objective of demonstrating the melter applicability to treatment of radioactive low-level waste and ion exchange resin from Korean nuclear power plant. Figure 2 shows a sketch of the system.

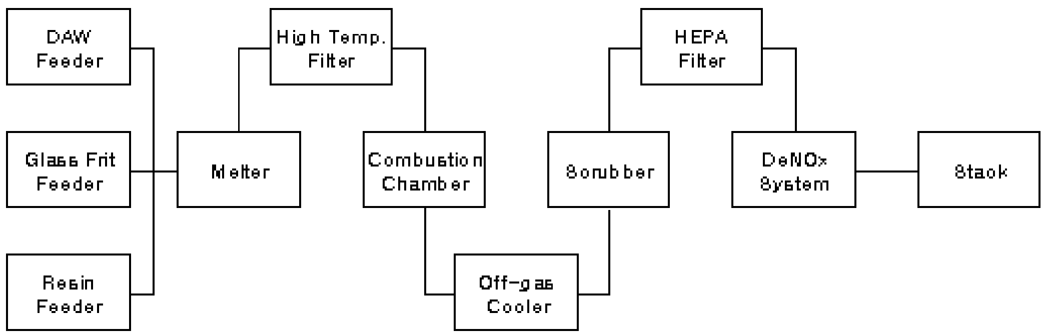

Fig. 2 Flow diagram of pilot-scale vitrification plant CCM

- Plasma Torch Melter (PTM) System

The PTM facility uses a hollow type plasma torch vertical mounted on Clamshell shape furnace to treat low-level radioactive waste from Korean nuclear power plants. The system includes a waste feed system, a 24kg/h melter, power supply, off-gas treatment system [3].

- Cold Crucible Melter (CCM) System

The CCM facility uses a hollow type plasma torch mounted on furnace to treat low-level radioactive waste such as dry active waste and ion exchange resin from Korean nuclear power plants. The system includes glass frit feeder, waste feeder, cold crucible melter, pipe cooler, high temperature filter, combustion chamber, off-gas cooler, scrubber, HEPA filter, reheater 1, extraction fan, reheater 2, deNOx system and stack [4].

REGULATIONS AND GUIDELINES FOR LILW TREATMENT FACILITIES

In order to develop a technical and regulatory guide for the previously introduced vitrification melters, we have studied any related regulations and technical concerns of similar facilities abroad. The related regulatory guidelines for safety possession of a treatment facility for LILW are as follows: IAEA safety guides, NRC, RCRA and state regulation in USA and regulations in Britain [5]. A document for design and operation of radioactive waste incineration facilities published IAEA, the safety series No. 108 advises basic and system contents [6]. Basic safety aspects include system safety analysis, accident condition, fires and explosions, containment of radioactivity, preventative measures, operating conditions, safety control function, criticality, industrial safety, effluent discharges and overpressure protection. System requirements consist of regulatory requirements, process requirements, operational reliability and maintainability, radioactivity containment, construction requirements, materials considerations, quality assurance, systems for alpha bearing waste processing and other considerations. Technical items for a conceptual design contain general requirements and considerations, functional requirements, waste feed, desired ash characteristics, waste volume reduction effectiveness, operational requirements, arrangement and location, system capacity, interface with in-plant systems, economic considerations, system design, combustion technique and off-gas treatment.
Subsystem requirements contain waste feed inspection and pretreatment, waste feeding, ash

removal, ash transfer and immobilization, fly ash removal, spent filter removal and scrub solution treatment. Component requirements include general requirements, redundant and backup components, waste transport containers, waste feed systems, combustion chambers, afterburner chambers, burners, heat exchangers, air injectors, evaporative coolers, cyclone separators, filters, charcoal absorbers, scrubber, off-gas fans and stacks. Section in controls and instrumentation covers system controls and monitoring and radioactive effluent monitoring. There are general requirements, shielding, contamination control, material handling, ventilation, requirements for operation and maintenance, fire protection, radiation monitoring and explosive gas monitoring in building and physical arrangement. In system testing and commissioning there are general requirements, structural integrity and thermal commissioning. System Operation is general requirements, startup, normal operation, abnormal operation, shutdown and organizational and administrative aspects. Documentation section covers control of documents, design criteria and baseline documents.

In the USA, the regulations of safety assessment for incineration facilities are divided into NRC (Nuclear Regulatory Commission), RCRA (Resource Conservation and Recovery Act), and state regulation and construction or operation to treat low-level radioactive waste are needed to satisfy all regulations. The NRC's mission is to regulate the Nation's civilian use of byproduct, source, and special nuclear materials to ensure adequate protection of public health and safety, to promote the common defense and security, and to protect the environment. The regulation of NRC examines safety assessment report and environmental assessment report for low-level radioactive treatment facilities. Requirements of RCRA need successful trial burn test results at constructed facility after approval and state regulation re-examines regulation of NRC in focusing emission controls [7].

In Britain, related regulation of LILW treatment facility contains operational safety assessment, environmental assessment, regulated limit, trial burn test and general requirements.

THE DEVELOPMENT OF GUIDELINES FOR A MELTING PLANT

It is related to general industrial considerations, basic safety aspects, system requirements and documentation for treating low-level radioactive facility considered survey some national regulation and technical guides. With regard to vitrification plants, additions could include melting plant requirements and waste form requirements to representative guidelines for treating low-level radioactive facility. Relevant requirements for vitrification plant are as follows:

- Industrial general requirements

Objective and scope of facility
Building and structure safety
Fires and Explosions
Industrial safety and accident protection
Effluent monitoring
Layout
Utility of electronic, steam and water
Relationships with nearby facilities
Overpressure protection
Operational reliability and maintainability

- Facility safety requirements

Safety assessment report (nuclear safety, structure safety, off-gas treatment and process)
Environmental assessment report
Site consideration
Standard of radioactive effluent
ALARA conceptual application
Workers and residential exposure
Earthquake protection
Radiation protection, source control, ventilation and confinement
Tightness and sealing

- Melting system requirements

System capacity
System design
Waste feed and pretreatment
Melting capacity assessment (melting temperature, residential time, DRE, volume reduction ratio, etc.)
Off-gas treatment
Handling, analysis and treatment of secondary waste (ex: particulates)
Subsystem and components capacity and operation
Trial burn test
Abnormal operation protection
Monitoring (sampling and measurement)
System materials

- Waste form requirements

Pertinent treatment
Safety and compatibility requirements (assessment of chemical durability, physical properties (density, strength), leachability (short and long term leaching test), structural properties, etc)

- Other requirements

Organizational and administrative aspects
Control of documentation
Quality assurance

The results of this study will be utilized to establish a final technical standard for the design, construction and operation of radioactive waste vitrification plants, and then utilized for the safety assessment and permitting of commercial vitrification plants to treat low- and intermediate-level radioactive wastes.

BASIC STUDIES ON CHEMICAL DURABILITY OF VITRIFIED PRODUCTS

In the course of efforts to develop a guideline for vitrification plants, researchers have found that leaching or chemical durability is one of the important items to be tested for insuring the stability of the final product of glassy forms. Investigations include basic leachability tests of simulated vitrified waste forms by various available test methods and observation of leaching behaviors in the connections to surface changes for long-term experiment. Also some prediction models of leached components from glassy forms are also suggested with comparison of results.

In this presentation are some representative results from basic studies for leaching of components from glassy products.

To determine an applicable pertinent leaching test, two different types of glassy waste forms - simulated radioactive, hazardous waste forms - were fabricated, and short- and long-term leaching tests were conducted using various types of leaching test methods such as ISO, MCC-4S, MCC-1P, MCC-5S, and PCT. By comparison of different test methods in terms of test apparatus, easiness, data analysis, ISO method for long-term test and PCT method for short-term test were suggested.

The cumulative fractions leached of calcium decreased in the order of ISO test, MCC-1P test, and PCT, while those of sodium decreased in the order of PCT, MCC-1P test, and ISO test [8]. This means that the leaching of calcium is greatly affected by leachant replacement and that the surface area of glassy waste form and temperature mostly affects the leaching of sodium.

The leaching mechanism of the components from glassy waste forms can be classified into wash-off, diffusion, and dissolution at the interface. Semi-empirical model of Eq. (1) was used to analyze the dominant leaching mechanism of waste glass.

The following time-dependent terms describing important rate-limiting leaching mechanisms were used to simulate the cumulative fraction leached and then to figure out the leaching behavior of materials as a function of time.

$k_1(1-\exp(-k_2t))$: Wash-off (chemical reaction between the glass surface and the aqueous solution).

$k_3t^{0.5}$: Transport by diffusion in a porous matrix.

k_4t : Leaching as a result of matrix dissolution.

These terms were combined into one general expression to describe the overall leaching behavior [9].

$$F(t) = k_1(1-\exp(-k_2t)) + k_3t^{0.5} + k_4t \qquad \text{(Eq. 1)}$$

As shown in Table I, the leaching mechanisms of silicon and calcium could be interpreted by dissolution or dissolution associated with diffusion because the difference between the order of magnitude of k_3 and k_4 was not very large. The leaching mechanisms of sodium and lithium were diffusion controlled.

Table I. Leaching mechanism of major glass components as radioactive incineration ash content

		Radioactive incineration ash content, wt%			
		0	30	50	70
Si	k_1	1.36E-04	2.54E-01	-1.45E+01	2.95E-04
	k_2	3.31E-01	2.87E-07	3.13E-10	2.48E-01
	k_3	1.19E-14	1.49E-04	5.82E-13	6.69E-05
	k_4	1.83E-05	4.16E-05	5.58E-05	6.71E-05
		0	30	50	70
Ca	k_1	-1.63E+00	2.56E+00	1.41E+00	-5.07E+03
	k_2	4.56E-10	1.55E-07	8.31E-07	1.31E-09
	k_3	3.11E-04	5.21E-04	6.74E-04	5.99E-04
	k_4	2.39E-05	2.39E-05	2.52E-05	2.11E-05
		0	30	50	70
Na	k_1	-3.62E+01	-7.24E+01	-5.68E+01	-5.62E+01
	k_2	2.44E-07	4.88E-07	3.82E-07	3.79E-07
	k_3	8.14E-04	2.28E-03	1.65E-03	1.72E-03
	k_4	8.30E-17	5.26E-16	3.58E-16	2.51E-16
		0	30	50	70
Li	k_1	-3.07E-04	-4.56E+01	-1.94E-01	-7.29+01
	k_2	6.53E+03	3.05E-07	1.25E-05	4.90E-07
	k_3	7.67E-04	1.93E-03	8.09E-04	2.70E-03
	k_4	6.79E-14	1.47E-16	2.57E-16	8.85E-16

Estimates of the amount of nuclides and glass matrix released from a vitrified waste form, which enters into the environment over long periods of time, are needed. The Prediction model estimates the amount of nuclides and glass matrix released from a vitrified waste forms over a time period considerably longer than that of the recorded data. The data are analyzed with a variety of functions, including the diffusion coefficient and dissolution rate, using Eq. (2) in order to predict the results [10].

$$\left(\frac{\sum a_n}{A_0}\right)\left(\frac{V}{S}\right) = (Dk)^{\frac{1}{2}}\left[\left(t + \frac{1}{2k}\right)erf(kt)^{\frac{1}{2}} + \left(\frac{t}{\pi k}\right)^{\frac{1}{2}} e^{-kt}\right] \qquad \text{(Eq. 2)}$$

For the prediction model, experimental data were applied with various initial leaching periods to find out the proper period with acceptable diffusion coefficients to avoid wash-off effect to the model. Until 7 days, the phenomenon of wash-off was the main leaching behavior in most experimented components. Therefore, diffusion coefficients D were calculated by leaching data between 7 days and 14 days in both prediction models.

The long-term prediction of cumulative fraction leached with increasing leaching period was found by a searching or successive iteration using two important factors such as diffusion coefficient and dissolution rate constant. A comparison of the experimental and predicted values for silicon using Eq. (1) is shown in Fig. 3.

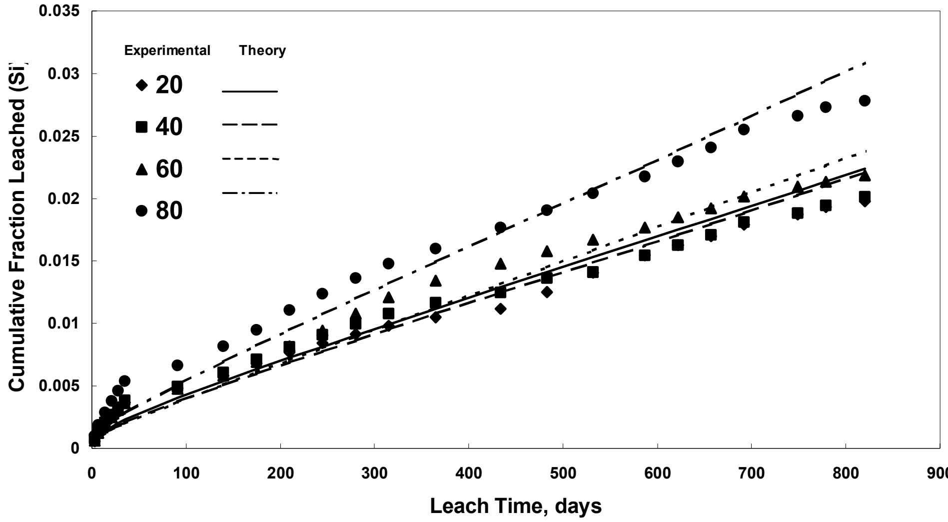

Fig. 3. Comparison of model calculation and experimental results
of silicon by long-term prediction model.

The accuracy of cumulative fraction leached between experimental and predicted values for silicon approached over 0.99 as the content of simulated incineration ash in the prediction model.

The SEM picture has been observed to investigate the surface of glass after the leaching test. As shown in Figure 4, the surface changes observed for the tested sample for 28 day. For the 2000 times magnification picture there is a stepped layer between 'A' part and 'B' part in Figure 4.

Additionally from the measurement of EPMA for reading the concentrations of components, sodium could have leached out more from surface than inside of glass. Such results support the leaching mechanism of sodium, which shows higher leaching in the initial period of leaching.

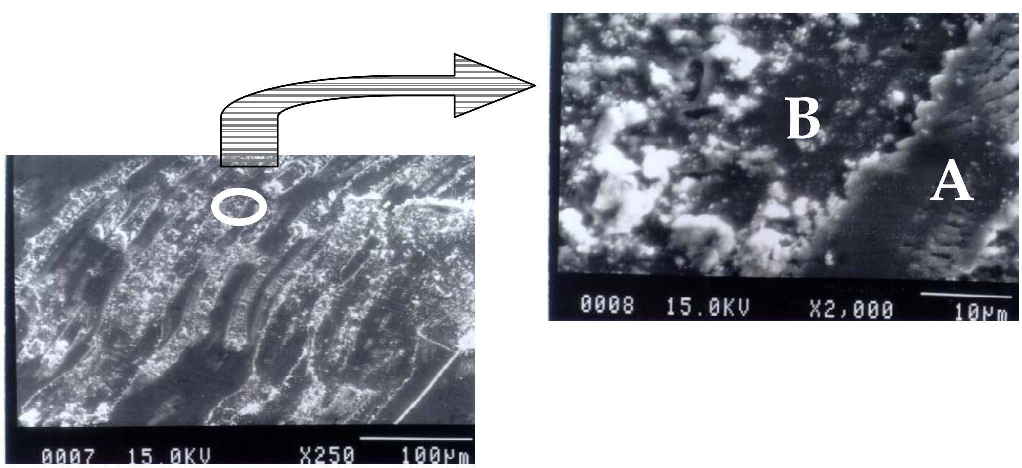

Fig. 4 SEM results on altered surface of glassy after 28days leaching test

CONCLUSIONS

1. Two pilot plants have been undergoing testing for the utilization of vitrification technologies to treat LILW by Korean companies and are expecting to commercialize and apply to get a permit soon.
2. A technical and regulatory guideline is also under development and will be established to permit and regulate such future plants of vitrifying wastes.
3. Some basic researches related to the utilizing technologies and regulatory guidelines have been carried out for better understanding on leaching of radionuclides from vitrified forms by testing different test methods, morphological analysis and model development.

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REFERENCES

[1] J. H. Kim, et. al., "High temperature melting technology using electronic arc", '99 Symposium on dioxin and ash treatment new technology, (1999).
[2] B. H. Yoon, et. al., "Characterization of glassy waste form using electronic arc melter", Proceedings of the 2001 annual conference of the Korean Solid Wastes Engineering Society Autumn, (2001).
[3] Y. P. Moon, et al., "Melting experiment on concrete waste using a hollow type plasma torch mounted on furnace", Proceedings of the 2000 annual conference of the Korean Nuclear Society Spring, (2000).
[4] S. C. Park, et. al, "Evaluation of off-gas characteristics in vitrification process of ion exchange resin", Proceedings of the 2000 annual conference of the Korean Nuclear Society Spring, (2000).
[5] Y. C. Seo, et. al., "A study on the safety of radioactive waste incineration facilities", KINS/HR-113, Korea Institute Nuclear Safety, (1994).
[6] IAEA, "Design and operation of radioactive waste incineration facilities", IAEA Safety Series No. 108, (1992).
[7] EPA, "Radiation and Mixed Waste Incineration", EPA Research Series, (1994).
[8] Y. C. Seo, et. al., "The comparison of leaching characteristics of glassy waste forms tested with various leaching procedures", '01 International thermal treatment technology conference, (1999).
[9] K. S. Lee, et. al., "A study on long-term leaching characteristics of vitrified waste forms containing hazardous and simulated radioactive incineration ashes", Journal of Korean Solid Wastes Engineering Society, (1999).
[10] Y. C. Seo, et. al., "The application of two prediction models on long-term leaching behavior of glass components and heavy metals in waste glasses contained incineration ash", International conference on incineration, pyrolysis, emission and combustion, (2000).
[11] I. T. Kim, et. al., "Leaching characteristics of waste glass forms containing two different two incineration ashes", 99 International thermal treatment technology conference, (1999).