

ADVANCED LLW CLASSIFICATION SYSTEM

T. Yagi, H. Yamazaki, A. Yokota, H. Kuribayashi
JGC Corporation

ABSTRACT

In the land disposal of low-level radioactive waste (LLW) in Japan, it is legally required that the concentration of each nuclide in the waste package be confirmed at each existing power station. The scaling factor method has been investigated as the optimum method of estimating the concentration of each nuclide. It has become necessary to develop a simple analytical method to determine the scaling factor because radiochemical analysis has been proved to be both difficult and expensive, requiring much time and manpower.

JGC has developed an advanced LLW classification system to confirm the concentration of each nuclide. The system consists of a new radionuclide-analysis system (IC-ICPMS), a scaling factor estimation system (computer code) and a direct assay system (gross gamma method).

The system can accurately evaluate the radioactivity of the waste, improve the detectable limit and reduce the work volume required for measurement, compared with previous manual radiochemical analysis.

INTRODUCTION

A Radioactive Waste Storage Center will be constructed in Rokkasho Village, Aomori Prefecture for low-level radioactive waste disposal in Japan. Operation of the Center will commence in 1992.

In considering applicable laws and regulations governing the land disposal of radioactive waste packages at the Center, most important are the upper limits and the total radioactivity of wastes to be received, and the total adopted for determining the radioactivity in the package.

Waste packages to be sent to the disposal site will be subject to the following requirement concerning the radioactivity concentration in each package:

The radioactivity concentrations of the waste packages shall not exceed the maximum concentrations specified in the licensing procedure for the Center.

The following 11 nuclides are specified in this procedure: H-3, C-14, Co-60, Ni-59, Ni-63, Sr-90, Nb-94, Tc-99, I-129, Cs-137, and total alpha nuclides. (See Table 1.)

Each waste package shall be identified by its radioactivity concentration expressed to two significant figures.

Considering such a background, the concept of the method for determining radioactive concentrations, the composition of the new classification system and test results are discussed herein. In addition, the application of the system to power stations is proposed in this report.

BASIC CONCEPT OF DETERMINING RADIOACTIVITY CONCENTRATIONS

Available Methods for Determining Radioactivity Concentrations

The methods listed below are considered to be available for determining the radioactivity concentrations and

the radioactivity of waste packages at nuclear power stations:

- a. Direct sampling from waste packages and radiochemical analysis of the sample.
- b. Direct sampling of process wastes and radiochemical analysis of the sample.
- c. Scaling factor method based on actually measured data and nondestructive measurement from the outside of waste packages.
- d. Estimate by using a radioactivity balance code.

Method (a) is a method whereby a sample is taken from each waste package and radiochemically analyzed, and method (b) is a method by which waste package source liquids such as evaporator concentrates are sampled and radiochemically analyzed.

In method (c), however, the radioactivity concentration of each waste package is estimated by using the results of nondestructive gamma nuclide measurements and the correlation factors between radioactive materials which are determined from a typical sample in advance. In method (d), the radioactivity concentration of each waste package is estimated by using a radioactivity balance code which contains plant operation control data, design data, and measured data on gamma nuclides contained in reactor coolants, such as Co-60 and Cs-137.

Each of the radioactivity concentration determination methods mentioned above has the merits and demerits shown following:

Method	Merit	Demerit
(a)	Measured data means package data.	Impossible to measure all samples. Impossible to extract typical samples in a strict sense. Increase in the possibility of personnel exposure and the spread of contamination.
(b)	Easy to relate measured data with packages.	Radiochemical analysis requires a large amount of manpower.
(c)	Easy method. NRC recommended.	Determining the scaling factor requires extensive radiochemical analysis in order to obtain significant statistical data.
(d)	Easy to calculate the radioactivity concentration in each waste stream.	Certifying the balance code and base data.

Based on the result of evaluation the merits and demerits of each of the radioactivity concentration determination methods, JGC developed an optimum method made up of a combination of these methods.

CONCEPT OF NEW CLASSIFICATION METHOD

Figure 1 shows the concept of the proposed new classification system based on the scaling factor method. The scaling factor must be developed from sufficiently persuasive data through the statistical analysis of actually measured data obtained from various wastes. However, analytical data which form the basis of scaling factors are obtained by the accumulation of the data on each sample. To obtain accurate data, a considerable amount of manpower is required for the analysis of each sample.

To ensure that data are statistically significant and to enable the data to be efficiently collected, it is important to enhance the system performance by utilizing the following three improvements:

Especially for the identification or quantitative analysis of radioactive material, it is a general practice to use methods based on radiation measurement. However, for long-lived radionuclides such as alpha-ray or beta-ray emitting nuclides represented by TRU nuclides and low-energy gamma-ray emitting nuclides, such methods require chemical separation of the nuclides (complete removal of obstructive radionuclides). Skilled analysis and a great deal of manpower are required for accurate analysis.

Labor Savings by Using IC-ICPMS Analyzing System

On the other hand, in mass spectrometric analysis, quantitative analysis is performed for the mass of each material. Therefore, the removal of obstructive material having the same mass as the material to be analyzed enables some other elements to be easily quantitatively analyzed at the same time.

Labor-saving radiochemical analysis is achieved by adopting an IC-ICPMS analyzing system for the analysis of reactor coolants.

Improvement of Balance Code

Improvement of Balance Code

The conventional computer code consists of a code for calculating the radionuclide concentrations of reactor coolants from reactor coolants analysis data (iodine isotopes) and core structural material data, and a code for estimating radioactivity concentrations for each waste stream from the calculated radionuclide concentrations and process data. In the new system, the computer code was improved so as to

TABLE I

Total Radioactivity and Maximum Radioactivity Concentration for the Radioactive Waste Storage Center

	Total Radioactivity	Maximum Radioactivity Concentration
	Ci	Ci/Ton
H-3	3.3×10^3	8.3
C-14	9.1×10^1	2.3×10^{-1}
Ni-59	9.4×10^1	2.4×10^{-1}
Co-60	3.0×10^4	7.5×10^1
Ni-63	1.2×10^4	3.0×10^1
Sr-90	1.8×10^2	4.5×10^{-1}
Nb-94	9.0×10^{-1}	2.3×10^{-3}
Tc-99	2.0×10^{-1}	5.0×10^{-4}
I-129	3.0×10^{-3}	7.5×10^{-6}
Cs-137	1.1×10^3	2.8
Total α	6.3	1.5×10^{-2}

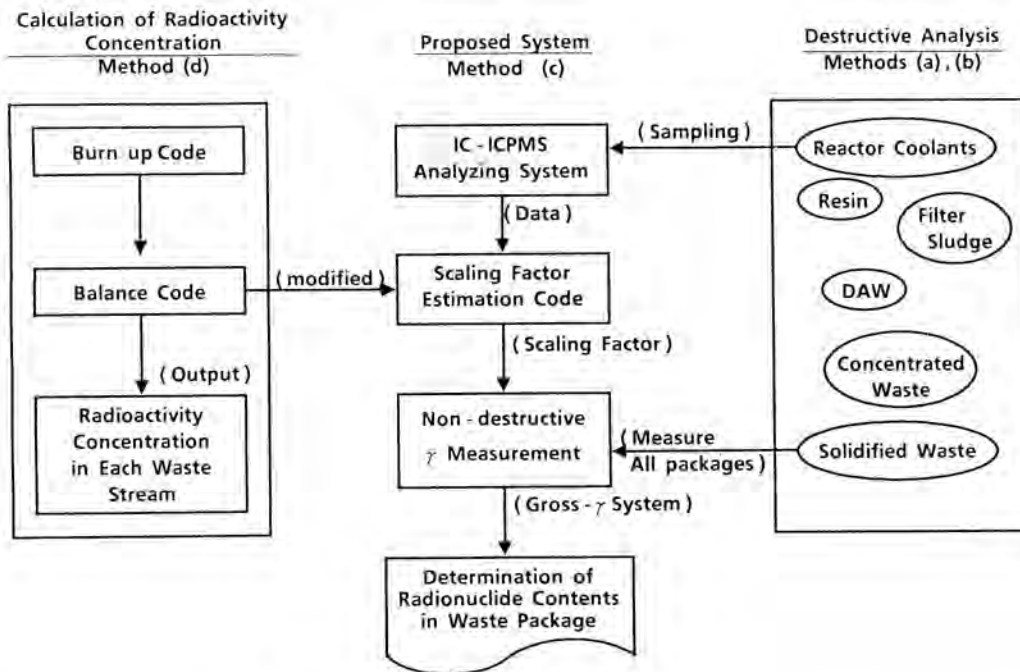


Fig. 1. Concept of New Classification System.

enable the scaling factor balance to be calculated for each waste stream.

As a result, a scaling factor for each waste stream is calculated by inputting into the improved code the data obtained by radiochemical analysis.

The scaling factor of each stream is calculated by inputting into the new code the data that is collected by using IC-ICPMS analyzing system.

Nondestructive Measurement

Gross gamma-ray coefficients which will not be easily affected by the radioactivity and density distributions in a waste package, are determined by measuring the gamma-ray including scattered rays from the whole surface of the package, without collimating them, using a plastic scintillator.

Each radionuclide is identified and its radioactivity is quantitatively determined from the results of gamma-ray spectrometry using a Ge semiconductor detector and the gross gamma-ray coefficients mentioned above, considering the detention efficiency for each radionuclide.

In this report, a basic study on the IC-ICPMS analyzing system is dealt with as a topic and, in particular, the possi-

bility of labor savings in the analysis of long-lived nuclides is studied.

BASIC TESTS ON IC-ICPMS ANALYZING SYSTEM

IC-ICPMS Experimental System

The system consists of ion chromatography (IC) for removing isobars and ICPMS for analyzing effluents. (See Fig. 2.)

The abbreviation ICPMS stands for "Inductively Coupled Plasma Source Mass Spectrometry" in which inductively coupled plasma is used as an ion source.

The IC and ICPMS are coupled by connecting the IC effluent port and the ICPMS nebulizer, using a 0.35 mm \varnothing PETEF tube. The separation performance and quantitative analysis performance of the system were verified by analyzing the specimen for each mass number with the elapse of time.

Test Example

The important matters in this test are the verification of the isobar separation performance of the IC and the verification of the qualitative and quantitative analysis performance of the system.

In order to verify that the latter performance can be judged by the ICPMS, a UV detector and a scintillation counter are inserted in series between the IC and ICPMS.

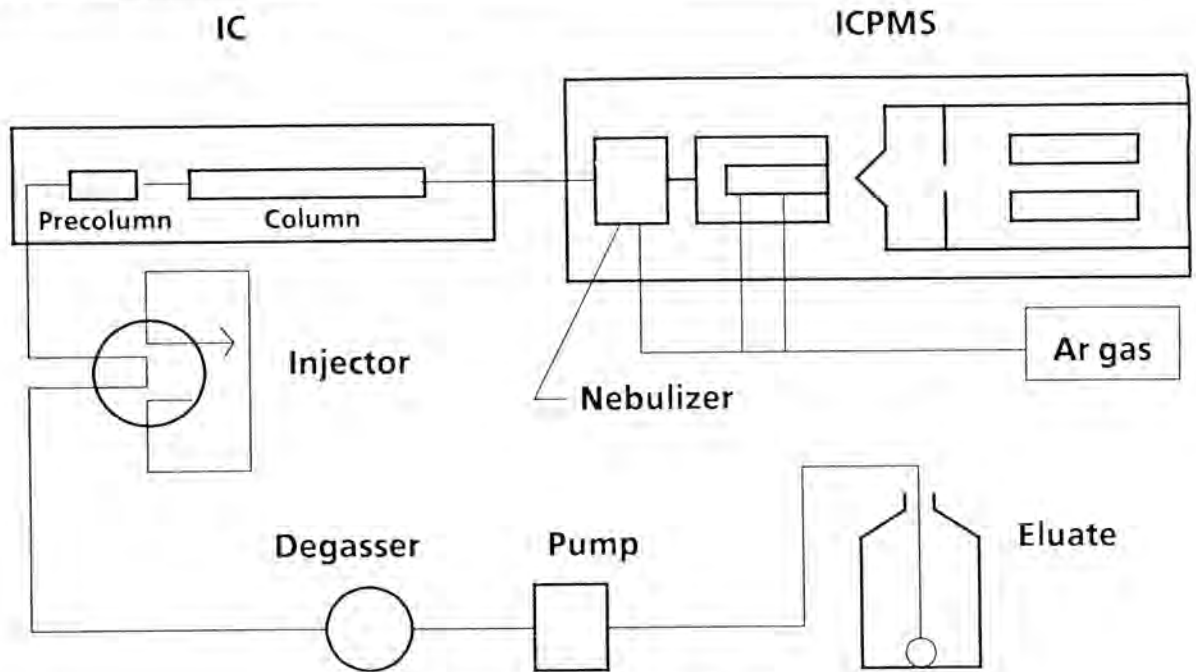


Fig. 2. Schematic Flow of IC-ICPMS.

The results of measurement by these detectors are studied together.

Fig. 3 shows the results of the IC effluent analyses by the UV detector and the ICPMS with respect to the Ni-63 and Cu-63 system.

The result of the analysis by the UV detector shows that Cu, Ni and Co ions are efficiently separated by the IC in this order.

In the ICPMS, the isobars are separated in the order of Cu and Ni and their peaks can be quantitatively determined when attention is paid to the measured value of mass number 63.

When attention is paid to mass numbers 58 and 59, it is judged from the time-serial analysis by the ICPMS that mass number 58 corresponds to the peak of the Ni isotope and mass number 59 corresponds to the Co peak.

When attention is paid to the Tc-99 and Ru-99 system, the Tc-99 peak appears at about 200 seconds in the scintillation counter as shown in Fig. 4, although it is not detected by the UV detector.

On the other hand, from the result of the detection by the ICPMS, it is seen that Ru-102 which is the stable isotope of Ru-99 appears at about 100 seconds and is clearly separated from Tc-99 appearing at about 200 seconds. In addition, this result shows that results can be obtained by one analysis also for the stable isotopes, Ru-99 and Ru-102.

Application to Power Station

When this system is applied to the analysis of diluted samples such as reactor coolants, the concentration of sam-

ples is required prior to analysis.

Fig. 5 shows a system flow of the IC-ICPMS analyzing system.

A sample is introduced into the concentration columns connected in series.

After the concentration is completed, the columns are connected to the separation and measurement system and the concentrated liquid is introduced into the system. The eluted nuclides are separated into the respective kinds of ions in the cation and anion separation columns by ion chromatography, and then introduced into the ICPMS.

The required analysis time is about two hours when the nuclides to be measured are Ni-59, Ni-63, Co-60, Sr-90, Nb-94, Tc-99, I-129, Cs-137, U-235 and U-239.

In the case of manual analysis by skilled analysts, 100 to 200 hours are required to obtain equivalent results. Figure 6 shows the detectable concentration range in manual analysis and IC-ICPMS analysis for liquid samples. This figure shows that the detection limits of manual analysis and ICPMS analysis are almost the same when the half-life period of the nuclides is shorter than several hundred years and that it is advantageous to use the ICPMS when the half-life period of the nuclides is longer than that.

CONCLUSION

When waste packages are classified based on an evaluation of the concentrations of nuclides important to disposal, it is practical to measure the concentrations of gamma-ray emitting nuclides for each waste package using a nondestructive measuring technique, and then determine

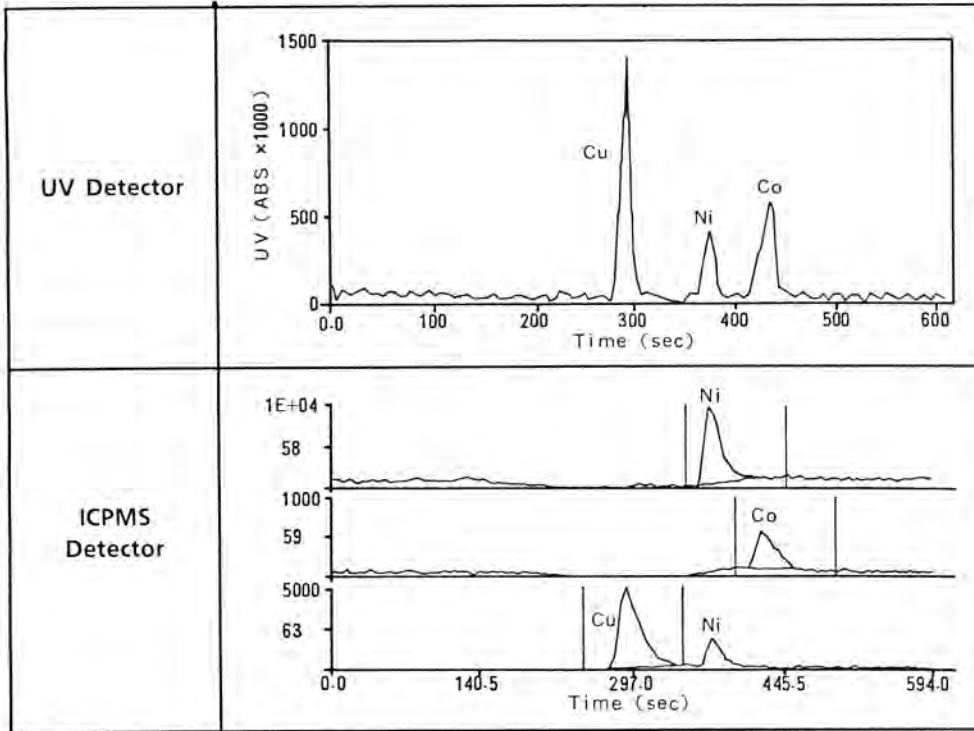


Fig. 3. NI-63/Cu-63 Chromatograph.

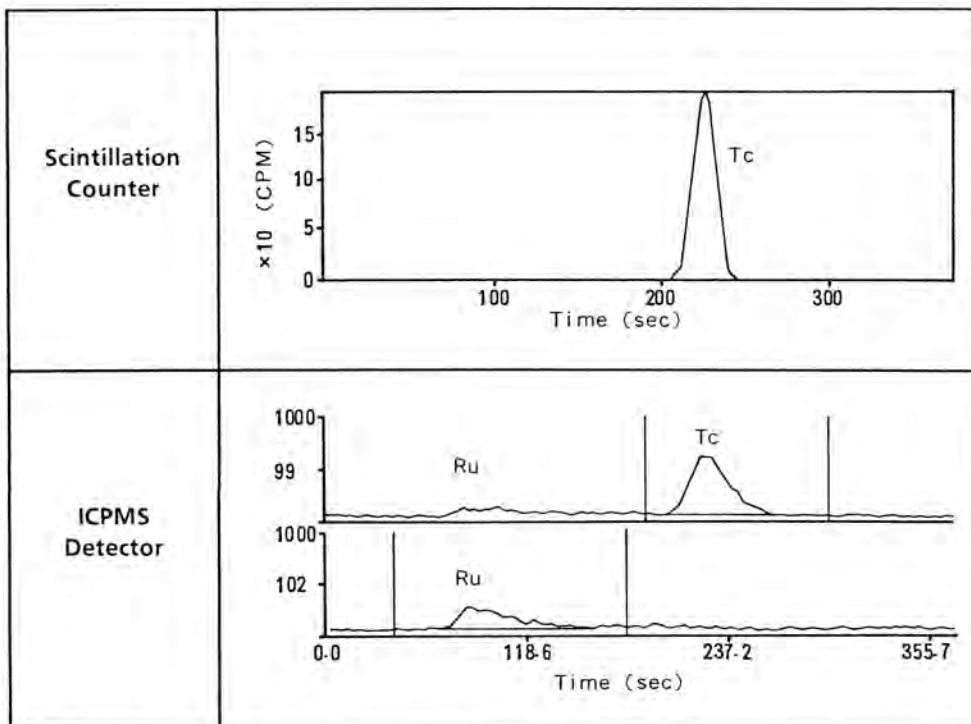


Fig. 4. Tc-99/Ru-99 Chromatograph.

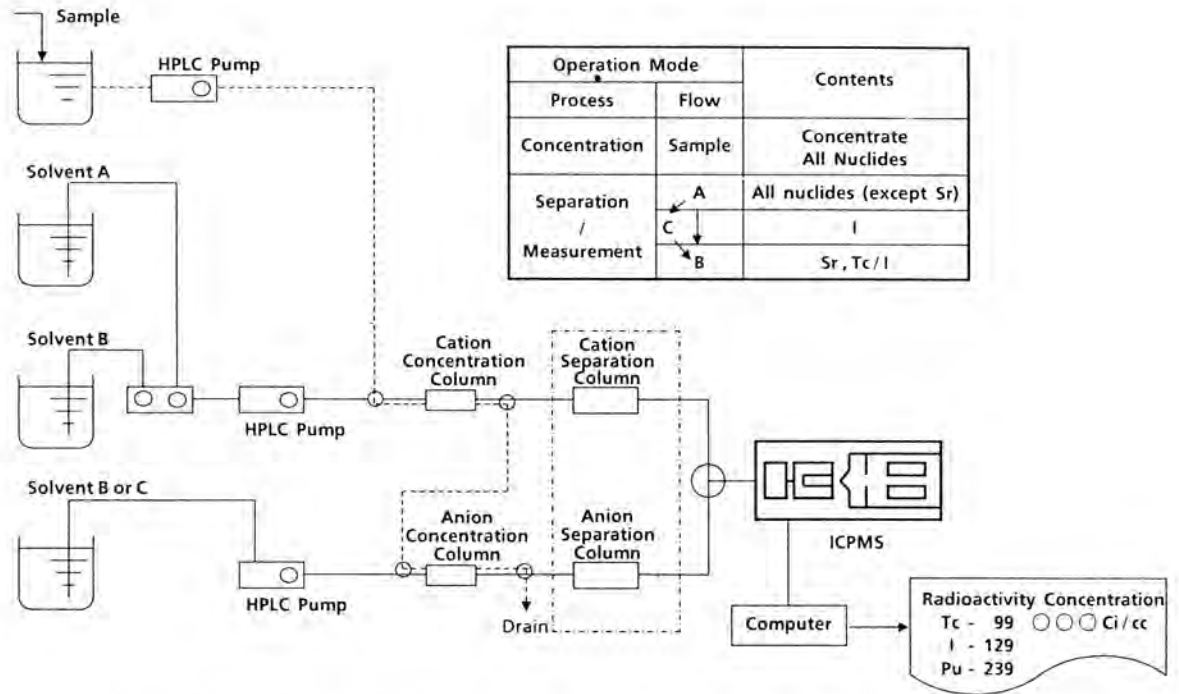


Fig. 5. System Flow of IC-ICPMS Analyzing System (Applied for Reactor Coolants).

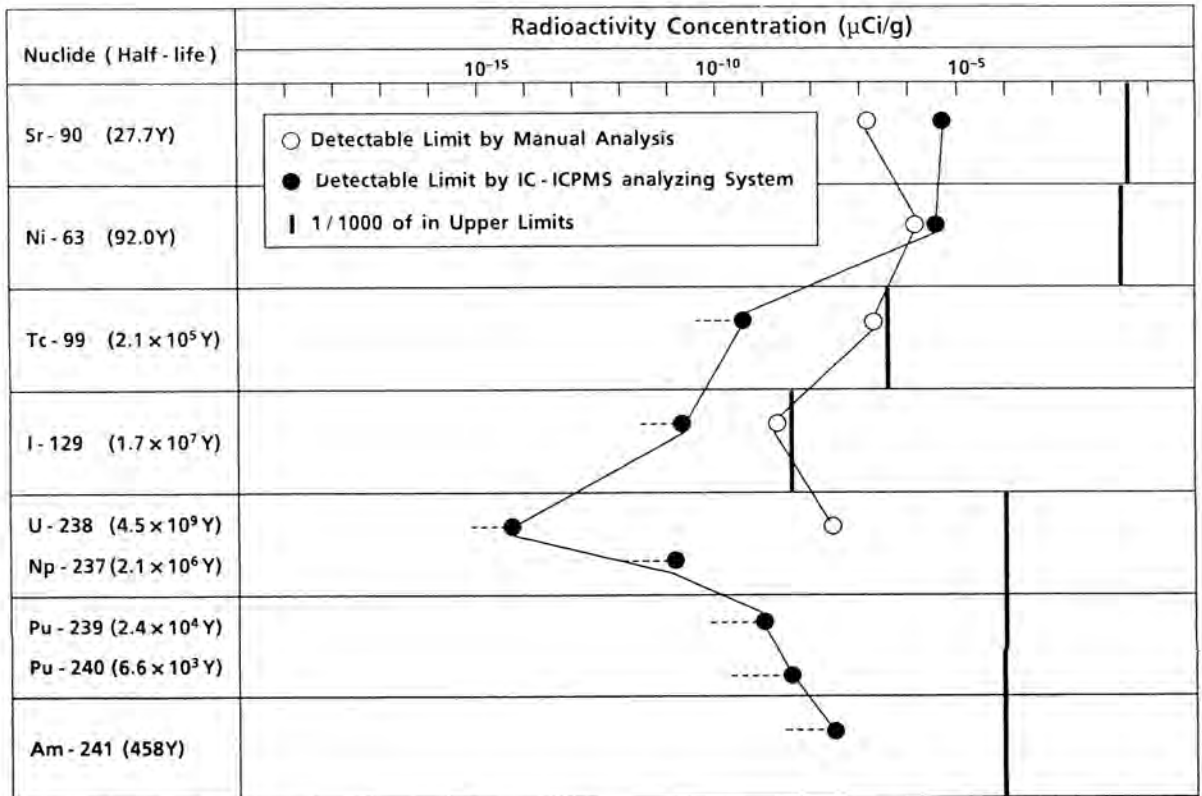


Fig. 6. Detectable Limits of IC-ICPMS Analyzing System and Manual Radiochemical Analysis.

the concentrations of difficult-to-measure nuclides by using the scaling factor method. Most important in such a method are highly reliable scaling factors and a high-accuracy non-destructive measuring device. Moreover, a large amount of manpower is required for data collection.

This report proposed a new classification system which is composed of the IC-ICPMS analyzing system for reactor coolant analysis, a computer code for estimating the scaling factors of each waste stream from the result of the IC-

ICPMS analysis, and the gross-gamma method for nondestructive measurement of the gamma nuclide concentrations in the package.

The new classification system is superior to existing methods for determining the radioactivity concentrations from the viewpoints of analysis accuracy, detection limits and labor-savings.