CURRENT STATUS OF JAERI PROGRAM ON DEVELOPMENT OF ULTRA-TRACE-ANALYTICAL TECHNOLOGY FOR SAFEGUARDS ENVIRONMENTAL SAMPLES

Japan Atomic Energy Research Institute (JAERI)
Tokai-mura, Ibaraki-ken 319-1195 Japan

Abstract

In order to contribute to the strengthened safeguards system based on the Program 93+2 of the IAEA, Japan Atomic Energy Research Institute (JAERI) has been developing analytical technology for ultra-trace amounts of nuclear materials in environmental samples, and constructed the CLEAR facility (Clean Laboratory for Environmental Analysis and Research). The construction of CLEAR was completed in December 2000, followed by the facility performance tests and the installation of analytical equipment, and full operation started in June 2001. This paper describes current status of research and development on the analytical technology, as well as the outline of CLEAR.

1. Introduction

In the JAERI, a program to establish the environmental sample analysis techniques for the strengthened safeguards system based on the Program 93+2 of the IAEA has been carried out since the middle of the Japanese fiscal year (JFY) 1996 [1-15] under the auspices of the Science and Technology Agency of Japan (STA), which was reorganized, in January 2001, into the Ministry of Education, Culture, Sports, Science and Technology (MEXT).

Table 1 shows the schedule of the program. At the early stage, technical issues necessary for construction and operation of a clean laboratory was investigated through literature survey and

### Table 1. Schedule of the JAERI Program (in the Japanese fiscal year)

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<td>1. Survey and investigation</td>
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<td>2. Preparation of the CLEAR facility</td>
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<td>3. Development of the techniques</td>
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<td>Particle analysis</td>
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Before the completion of CLEAR, the preliminary examination of the analytical techniques for ultra-trace amounts of nuclear materials in environmental samples had been carried out at existing laboratories in the following fields: screening, bulk analysis and particle analysis. From June 2001, the place of the R&D works was moved to CLEAR. The first phase of the program continues until March 2003. During this period, essential technology for ultra-trace analysis for uranium and plutonium will be established with sufficient sensitivity and accuracy for the environmental sample analysis. The JAERI will contribute to the strengthened safeguards system of IAEA by joining the community of Network Analytical Laboratories as the first member from Asian area, as well as (contribute) to the domestic environmental sample analysis.

2. DEVELOPMENT OF THE ANALYTICAL TECHNIQUES

A flow diagram of safeguards environmental sample analysis is shown in FIG. 2.

2.1. Screening technique

Screening aims at estimating, by non-destructive analysis, the amounts of nuclear materials in environmental samples to be introduced into the clean rooms, and is the first step to avoid cross-contamination among the samples and contamination of the clean rooms themselves. For this purpose various kind of radiation spectrometry has been examined.

HPGe detectors are often used but they are not efficient for screening because photo-peaks of actinides’ γ-rays (mostly below 200 keV) are severely interfered by Compton continuum of
2.2. Bulk analysis

As for the bulk analysis, efforts are temporally made on uranium in swipe samples. Preliminary examination for optimization of sample pre-treatment conditions is in progress. At present, four methods: 1) leaching with nitric acid, 2) dry ashing, 3) low-temperature plasma ashing, and 4) acid digestion have been examined from the view points of uranium blank, cross-contamination,
chemical yield and manipulation condition in a clean laboratory.

For the isotopic ratio measurement, performance of inductively-coupled plasma mass spectrometry (ICP-MS) is mainly examined because sample preparation for ICP-MS is simpler than that for thermal ionization mass spectrometry (TIMS). Interference of polyatomic ion, PtAr⁺, on the uranium ions and mass bias caused by ICP-MS operating conditions are being investigated for precise measurement of uranium isotope ratio.

It was found by our preliminary measurements (see FIG. 4) that the swipe material (TexWipe TX-304, usually used by the IAEA) contains non-negligible uranium blank with large deviation (2-6 ng/sheet). This would introduce significant uncertainty in the analysis, therefore, study on selective recovery of uranium particles from the swipe matrix is sought. Otherwise, alternative swipe materials with less uranium blank would be preferable.

![Uranium blank in Tex Wipe TX-304](image)

**FIG. 4. Uranium blank in Tex Wipe TX-304**

### 2.3. Particle analysis

The analytical technology for individual particles in the environmental samples is an important issue to develop. Works are continued with total reflection X-ray fluorescence spectrometry (TXRF) for screening, electron-probe microanalysis (EPMA) for elemental composition and morphology of each particle, and secondary ion mass spectrometry (SIMS) for isotopic ratio measurement.

![Glassy carbon mount compatible among TXRF, EPMA and SIMS](image)

**FIG. 5. Glassy carbon mount compatible among TXRF, EPMA and SIMS**
A special mount (see FIG. 5) made of glassy carbon was designed in order that the mount could be commonly used among the three apparatuses. The detection limit of uranium in particle screening by TXRF was achieved to 0.4 ng (see FIG. 6).

At present, isotope ratio of uranium particle with the diameter of several µm was measured (see FIG. 7). By combination of TXRF, EPMA and SIMS, the throughput for analysis on uranium particle of 1 µm was one swipe per day, which is to be increased by improvement of the technique for particle mapping.

3. OUTLINE OF THE CLEAR FACILITY
The overview of CLEAR is shown in FIG. 1. The facility consists of the analytical building and the administration building. The former is divided into the following three areas: clean laboratory area, general laboratory area and support area. The general laboratories are used for screening, pre-treatment of environmental samples and so on. The support area has machine rooms (hot and cold) and an electricity room. The clean laboratory area is farther divided into the chemical treatment area (class 100, defined by Fed. Std. 209 E), the instrumental analysis area (class 1,000 and 10,000) and the service area (see FIG. 8).

Chemical treatments, such as sample pre-treatment, separation, purification and reagent preparation, are conducted in the chemical treatment area (215 m²). Analytical equipment for isotope ratio and radioactivity measurement is installed in the instrumental analysis area (480 m²). Utility machinery for the equipment is allocated in the service area. Most of the clean rooms were equipped with clean fume-exhausted hoods and clean work benches. The hoods were specially designed ones that prevented the room-air blowing onto the working surface of the hood. Prior to the installation of analytical equipment, the facility performance tests were carried out and satisfactory results, e.g., class 10 on working surfaces of the clean hoods and benches were achieved.

The clean laboratory area and the hot machine room were zoned for the radiation controlled area, because very minute amounts of nuclear materials, e.g., about 10 ng per sample for uranium and about 1 ng per sample for plutonium, are to be used as spike, mass calibration and reference. For confinement of radioactive materials within the area, the pressure in the service area is kept lower than the outer environment.

The materials used for clean room structure were carefully selected in order that they might not corrode with strong acids, such as hydrochloric acid, perchloric acid and hydrofluoric acid, and if necessary, metallic surfaces were coated with corrosion-resistant resin.
4. COLLABORATION WITH IAEA, DOE AND EURATOM

Under the framework of Japan Support Program for Agency Safeguards (JASPAS), a task concerning the Qualification of IAEA’s Environmental Network Laboratories has been undertaken since October 1999. Some test samples offered by the IAEA were analyzed at the JAERI with EPMA and SIMS.

Under a specific memorandum of agreement between the JAERI and the DOE concerning research and development of safeguards, a part of this program has been carried out since 1997. Protocols for the general clean laboratory operation and system performance tests were jointly established in 2000. The task has recently concentrated on quality assurance and quality control measures. In addition, on-site reviews on CLEAR from the viewpoint of QA/QC by DOE experts are scheduled for 2001 and 2002.

Under an agreement between the JAERI and the EURATOM in the field of nuclear materials research and development, both researchers have exchanged the information on research and development of trace analysis techniques. The JAERI participated in the IRMM Nuclear Signature Interlaboratory Measurement Evaluation Programme (NUSIMEP-2: uranium) and demonstrated its ability to measure isotopic composition of trace-amount uranium in solution samples. For particle analysis, the JAERI will take part in SIMS round-robin exercise organized by the ITU.

5. SUMMARY

The JAERI has been developing analytical technology for ultra-trace amounts of nuclear materials in environmental samples at existing laboratories since 1998. Construction of CLEAR was completed in December 2000 and its operation started in June 2001. The place of the R&D was, thereafter, moved to CLEAR and works is in progress with a target to attain the qualification of IAEA’s Environmental Network Laboratories by March 2003. International collaboration, especially with the IAEA, the DOE and the EURATOM, is indispensable for promoting this program. CLEAR and the techniques to be developed will be also used for the CTBT verification and for basic research and development of environmental sciences.

REFERENCES


