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FOREIGN TRIP REPORT

ORNL/FTR-3292

DATE: June 19, 1989

SUBJECT: Report of Foreign Travel of J. R. Stokely, Section Head, Analytical Chemistry Division

TO: A. W. Trivelpiece

FROM: J. R. Stokely

Purpose: To attend and participate in an international conference on analytical chemistry at Karlsruhe, West Germany, June 5-9, 1989

SITES VISITED:

6/5-8/1989	Conference	Karlsruhe, FRG	H. S. Ache R. Berg

ABSTRACT: The traveler attended the Second International Conference on Analytical Chemistry in Nuclear Technology and presented recent ORNL work on chemical and radiochemical analysis to characterize mixed radioactive waste. The conference consisted of morning sessions where 3-5 invited papers were given. Invited papers dealt, for the most part, with analytical chemistry in nuclear fuels reprocessing and safeguards and with new analytical techniques of general interest. Poster sessions were held in the afternoons, and a wide diversity of subjects were covered in these sessions. Subjects of particular interest were in-line and on-line process analysis using fiber optic spectrophotometry, x-ray fluorescence, and gamma-ray spectrometry, chemical characterization of radioactive waste, new neutron activation analysis methods, and use of computers and robotics in analytical chemistry laboratories.

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REPORT OF FOREIGN TRAVEL

The Second International Conference on Analytical Chemistry in Nuclear Technology was held at the Nuclear Research Center at Karlsruhe. The conference brought together approximately 300 analytical and radiochemists with a broad representation from a large number of countries. The conference consisted of morning sessions where invited talks were given, afternoon poster sessions, and three evening receptions. In the afternoons and at the receptions, there were ample opportunities for informal technical discussions. In general, the conference was well run, and there were a number of papers and posters which gave information that will be beneficial to our work at ORNL. Highlights from some papers and posters that are of particular interest for our work are described in this report.

Many of the talks and posters dealt with nuclear fuel reprocessing and safeguard type activities. The western Europeans, in particularly, are very active in these areas. Since we in the United States are doing little new work in the fuel reprocessing now, we are falling behind in this important technical area. European activities in fuel reprocessing analytical chemistry are oriented to on-line and in-line, automated type analyses using spectrophotometry, x-ray fluorescence, and gamma-ray spectrometry. The basic idea for this work is to provide process control data on a near real time basis for mass and flow balancing, troubleshooting, and accountability.

Colin Allan of UKAEA at Dounreay gave an interesting talk on the role of in-line analytical instrumentation for control of nuclear fuel reprocessing. Dr. Allan reported that reprocessing plant control is still very much dependent on classical off-line, laboratory analysis because it is often very difficult to back fit a processing system for on-line analysis. Also, laboratory analysis costs are small relative to overall costs and there is relatively little cost incentive for utilization of on-line analysis. Dr. Allen recommends that laboratory deployment of in-line sensor technology be encouraged so that the in-line sensor and system development and acceptance process can go forward even though on-line, process usage is not anticipated for several years.

E. A. Hakkita and Patrick Day from Los Alamos National Laboratory gave two separate papers related to in-line, near-real-time analyses for process control. Hakkita's paper discussed primarily safeguard

type applications, and Day's paper described new sensor development for in-situ measurements. As discussed by Hakkita, a new word -optrodes- is being used to describe spectrometric techniques for in-line measurements that utilizes lasers and fiber optic systems. X-ray absoption/fluorescence techniques, as developed at Karlsruhe, are working out well for on-line analysis for plutonium. Active and passive neutron measurements can also be effectively utilized for measurement of both uranium and plutonium in process solution. Hakkita also talked briefly about developments in the use of robotics system for chemical separations and mass spectrometric analysis of plutonium and uranium. Day talked about new work at Los Alamos on development of chemical sensors for actinides solution that could have application for in-line process analysis as well as laboratory analysis. The Los Alomos group is working toward incorporation of a specific chemical reagent into a porous polymer coating attached to a solid substrate. Normally, the reagent forms a colored compound with the species of interest and the spectral response is measured. So far the group has developed a sensor (using chromazoral) for strong acid (1-10 M) measurements. They are also investigating the use of quartz microbalance (piezoelectric detector) with specific coatings for use as sensors. The group is interested in sensor development for total acid, fluoride, oxalate, peroxide, low-level actinides, and plutonium valence states.

Patrick O'Rourke from Savannah River Laboratory gave a very good presentation on the use of chemometrics in nuclear fuel reprocessing. O'Rourke showed examples of the use of chemometrics in the simultaneous spectrophotometric determination of uranium, plutonium, nitrate, and acid in process solution. He also described the residual analysis technique for multicomponent analysis of PCBs by gas chromatography. Since ORNL performs many PCB analysis as part of the environmental monitoring program, this could be a useful technique for us to investigate.

A paper by R. Schott of CEA described some application of liquid chromatography (HPLC) in hot cells that may have use in our work at ORNL. HPLC has a real advantage for analysis of radioactive materials because small volumes of waste are generated (besides volatile solvents). Schott described a system for HPLC analysis of uranium in a hot cell. A shielded sampling value (outside of hot cell) is used to take a small volume of sample. The sample is passed through a HPLC column (also shielded) and then through

a UV detector. Very little non-volatile waste is generated and very reliable and precise (RSD: 0.7%) results were obtained. The CEA group is also investigating use of HPLC for plutonium and TBP analysis. We should follow this work closely and consider setting up and evaluating of a HPLC system in one of our hot cells for general analysis purposes (could be useful for radioactive waste analysis).

Because of work at ORNL and other DOE laboratories in radioactive mixed waste analysis, I was somewhat disappointed to see so few papers at the conference dealing with this topic. Besides my poster, two other posters were given on mixed waste analysis, and both of these were reports on U.S. work. John McCown from PNL gave a poster on their work with mixed hazardous waste. Tony Toste of Southwest Missouri State University (formerly at PNL) gave a poster on organic compound diagenesis in nuclear waste. Tony and I talked for some time about his work which is investigating the radiolytic reactions of organic compounds in nuclear waste. He has so far successfully identified approximately 75% of the soluble organics in some samples of Handford waste by performing derivitization gas chromatography and is beginning to understand the radiolytic mechanisims going on within the waste.

Christopher Lierse from the University of Munich gave an invited talk on the redox behavior of actinides that has implications for long term disposal of the radioactive TRU elements. (Lierse was presented the Fritz Strassmann Award at the conference for this work). I talked with Lierse about this work because of our former work with actinide redox reactions and the implications of the work for underground storage of TRU waste. Lierse has studied the radiolytic reactions of actinides in high chloride media (brine). He found that some interesting free radical reactions take place that produce strongly oxidizing chlorine-containing species. The higher oxidation states of americium and plutonium are formed and these are highly soluble and are more mobile than the lower oxidation states. Although this work is basically a fundamental study, it does generate information that could be important in the future for high level TRU waste disposal. Lierse is continuing work in the area investigating other actinides (Cf, Np) and may get into other media.

There were a small number of posters on neutron activation analysis and environmental radiochemistry, but little new material was presented that could help in our work. Dick Lindstrom of NBS

gave a poster on iodine-129/iodine-127 ratio measurements that was of very high quality. Dick uses optimized fluxes of both thermal and fast neutrons to minimize interferring nuclear reactions. He finds that the reaction ^{127}I $(3n,\gamma)^{130}I$ at high flux sets the limit for sensitivity for neutron activation analysis of ^{129}I. Other papers on neutron activation analysis were concerned with analysis of toothpaste, and thorium analysis using fast neutrons together with delayed neutron counting. Several posters were given on low-level radiochemical analysis of contaminated foods, water, and environmental materials coming from the Chernobyl fallout.

There were a large number of papers and posters (approximately 200) presented at the conference. Some general observations about the work presented are as follows:

(1) Inductively coupled plasma spectrometry is being widely used for analysis in the nuclear field. Several laboratories had papers describing applications for both radioactive and non-radioactive materials. A few applications of atomic absorption analysis were reported.

(2) X-ray fluorescence and absorption techniques are being developed at a number of laboratories for both laboratory and in-line process analysis of uranium, plutonium, and other elements.

(3) There were a few attendees from the People's Republic of China at the conference, and they gave a few posters (of little interest).

(4) Analytical chemistry development work continues in the nuclear fuel reprocessing area with the main emphases on sampling methodology and in-line analysis.

(5) Several posters were presented on neptunium analysis in nuclear fuel solution. Most posters were modifications to methodology developed in United States in the 50s and 60s.

(6) Conventional mass spectrometry continues to be widely used particularily in the accountability /safeguards area (plutonium and uranium analysis).

(7) Only a few papers or posters were presented about radioactive waste characterization. Except for work in United States, there is little interest in hazardous chemicals in radioactive waste at this time.

(8) One particularily interesting application of a laboratory robotics system was described. This application dealt with preparation of samples for thermal emission mass spectrometric analysis of U and Pu.

Conclusion

Overall, I think the Second Karlsruhe Conference was a very worthwhile experience for me. I came away from the conference with a number of technical ideas that will hopefully improve our operation and analysis techniques at ORNL, particularily in our work in chemical analysis of radioactive materials associated with radioactive waste and the transuranium element production program. Also, I have a much better feel for the work going on in Europe in the nuclear analytical chemistry area as a result of my attendance at the conference and opportunity to become acquainted with chemists working in the area.

APPENDIX

A. Itinerary

6/5-8/1989	Conference	Karlsruhe, FRG

B. Persons Contacted (Primary)

H. J. Ache	Nuclear Research Center	Karlsruhe, FRG
R. Berg	Nuclear Research Center	Karlsruhe, FRG
M. K. Holland	Savannah River Plant	Aiken, SC
B. F. Myasoedov	Vernadsky Institute	Moscow, USSR
C. Lierse	Universitat Munchen	Munchen, FRG
D. W. Adaway	USAEA	Dounreay, UK
J. K. Li	Los Alamos National Lab	Los Alamos, NM
N. N. Papadopoulos	Nuclear Research Center	Athens, Greece
A. P. Toste	PNL	Richland, WA
E. Tachikawa	JAERL	Tokai, Japan
J. W. McMillan	Harwell	Kidcot, UK
C. G. Allan	USAEA	Dounreay, UK
L. Salonen	Center for Nuclear Safety	Helsinki, Finland
F. Marsh	Los Alamos National Lab	Los Alamos, NM

C. Bibliography

Abstract Booklet for Second International Conference on Analytical Chemistry in Nuclear Technology, Karlsruhe, West Germany, June 5-9, 1989.

Distribution

1. Assistant Secretary for International Affairs, DOE, Washington
2. D. K. Stevens, Associate Director for Basic Energy Sciences, Office of Energy Research, DOE, Washington
3. Richard L. Egli, DOE/ORO
4. W. G. Phelps, Acting Director, Safeguards and Security Division, DOE/ORO
5-6. Office of Scientific and Technical Information, P. O. Box 62, Oak Ridge, TN 37831
7. J. R. Stokely
8. A. W. Trivelpiece
9. W. D. Shults
10. J. A. Carter
11. M. R. Guerin
12. W. R. Laing
13-14. Laboratory Records Department
15. Laboratory Records Department - RC
16. Laboratory Protection Division
17. ORNL Patent Section
18. ORNL Public Relations Office