REINTRODUCTION OF HYBRID K-EDGE DENSITOMETRY TO UNITED STATES FUEL REPROCESSING FACILITIES

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ABSTRACT

Hybrid K-edge densitometry (HKED), a combination of the K-edge and X-ray fluorescence densitometry techniques, is an existing technology that is ideally suited for measurements involving reprocessing plant solutions. It has become the workhorse method in reprocessing plant laboratories around the world. For input solutions, the accuracy associated with HKED measurements approaches that of the more traditional methods, such as isotope dilution mass spectrometry (IDMS). The system has also been used for plutonium product solution measurements in place of IDMS or coulometry when quick turnaround and minimal sample preparation are desired.

The most recent application of HKED involves providing quantitative measurements of other minor actinides. This is a major consideration in the U.S. Department of Energy's Global Nuclear Energy Partnership (GNEP) processing schemes and in the application of other nondestructive assay measurements relying on neutron measurement. Although HKED is not a new technology, currently there are no deployments of HKED systems domestically, with the exception of a single development system at Los Alamos National Laboratory.

Oak Ridge National Laboratory (ORNL) has been working to procure and install an HKED system at the ORNL Radiochemical Engineering Development Center, the site for the GNEP Coupled End-to-End (CETE) Demonstration. Because actual spent fuel dissolver solutions will be available, the potential for multiple applications and opportunities involving an HKED system exists. The reintroduction of HKED technology in the production environment of the CETE Demonstration will not only offer an unprecedented opportunity for U.S. users to gain experience domestically, but will also provide a qualification testing center for upgrades and changes to HKED systems worldwide, a need expressed by the International Atomic Energy Agency and the U.S. Support Program.

1.0 BACKGROUND

The U.S. Department of Energy's (DOE's) Global Nuclear Energy Partnership (GNEP) has the goal of expanding the reprocessing of spent nuclear fuel in a way that encourages domestic and international use of nuclear energy, helps to resolve nuclear waste management issues, and develops technologies to significantly reduce nuclear proliferation risks in reprocessing technologies. GNEP is a plan to form an international partnership devoted to developing proliferation resistant methods of reprocessing spent nuclear fuel. The specific goals and approach of the GNEP program have been recently defined in DOE's GNEP strategic plan and the GNEP program management plan.

Group actinide separation and recycle of actinides back to the reactor have never been performed on any significant scale. The Oak Ridge National Laboratory (ORNL) GNEP Coupled End-to-End (CETE) Demonstration addresses technical uncertainties related to the integration and scale-up of the activities required to deploy group actinide separation and reactor transmutation.[1]

Hybrid K-edge densitometry (HKED) has become the workhorse for analytical determination of uranium (U) and plutonium (Pu) concentrations in process solutions in reprocessing plants. While it has not yet displaced isotope dilution mass spectroscopy (IDMS) for accountancy determinations in dissolver solutions, it has gained wide acceptance for process control application by operators and verification measurements by inspectorates. The technique requires only a passive measurement on solution sample bottles, offering quick and accurate measurements without any chemical preparation or the ensuing consideration of generated analytical wastes. The International Atomic Energy Agency (IAEA) now typically analyzes 100% of the input dissolver solution samples and plutonium nitrate product samples in reprocessing plants by HKED, with only 10% of the samples analyzed by IDMS for quality control.

Accuracies for U and Pu concentrations in dissolver solution measurements by HKED are approaching IDMS standards (better than a few tenths of a percent for U, and better than half a percent for Pu). The procedure for measurement is much more timely and less intensive for laboratory operations (i.e., count times of 20 minutes with no chemical preparation). The most recent application of the HKED system at the Rokkasho Reprocessing Plant in Japan has extended the technique to measure the minor actinides, using HKED to determine the plutonium-to-curium ratio. Determination of other actinides will be essential as GNEP processes move forward.

While HKED technology is widely deployed in reprocessing facilities and by regulatory agencies around the world, unfortunately it has not been deployed in U.S. facilities because the fuel cycle processes most suitable for the measurement did not exist in the United States. GNEP is dynamically reshaping fuel cycle processes in the United States, and it is time to deploy the technology and provide U.S. personnel with the requisite experience to use it. This would also allow the United States to actively participate in the emerging HKED International Users Group.

Herbert Ottmar developed HKED technology at the Karlsruhe Institute of the European Atomic Energy Community (Euratom).[2] Ottmar and Karlsruhe have been the principals in making the technology available to IAEA and Euratom. Mike Collins at Los Alamos National Laboratory (LANL) is the principal following development in the United States and recently contributed to upgrades in the evaluation codes for actinide determinations.

HKED couples K-edge densitometry and X-ray fluorescence (XRF) densitometry into one system that can report U and Pu concentrations in process solutions. K-edge densitometry is a technique that measures the transmission photon spectrum of a photon beam passing through a process solution sample. The K-edge typically refers to the dramatic drop in transmission at an energy that is characteristic of the K-shell absorption energy for a given element. By measuring the transmission of the material and knowing the mass attenuation coefficient for the material, the concentration can be determined.[3]

XRF densitometry is a technique that takes advantage of the characteristic X-rays emitted during de-excitation of U and Pu. As an X-ray beam passes through the solution sample, the X-rays excite the electrons to a higher energy level. When these excited electrons de-excite, a characteristic X-ray with an intensity that is proportional to the elemental concentration is emitted.[4]

HKED technology is licensed for commercial availability through Canberra Industries, Inc., and ORTEC/AMETEK. These suppliers are currently interested in upgrading existing systems to include the latest evaluation software and upgrading the platform to a PC-based system. Current users of HKED systems are also interested in these upgrades; however, any upgrades will require testing before deployment.

There is only one HKED measurement unit in the United States, at LANL, but it is an older development unit requiring upgrade. The ORNL activities sponsored by DOE include installing a modern unit in a location where real process solutions will be measured as part of the CETE Demonstration. This system will be used to train personnel throughout the DOE complex, from universities, and from industry, who will join the GNEP program as it evolves from design to the construction of new facilities. In addition, the system could support international collaborations in testing and activities of the HKED International Users Group and IAEA. The installation would be part of the CETE Demonstration and use the structure and capabilities of the ORNL Safeguards Laboratory as the platform for training and international collaborations.[5]

2.0 GOALS OF HKED INTEGRATION

HKED will be integrated into the GNEP CETE Demonstration. The purpose of this integration will be to verify the U and Pu concentrations in dissolver and other process solutions. HKED will also become part of the ORNL Safeguards Laboratory's activities to facilitate access, training, and international collaboration.

The goals of the current effort to reintroduce HKED to the United States go beyond simply installing a unit and include helping to develop a base of expertise in the United States. The main tasks of the HKED integration include the following.

2.1 Assembly of a knowledge base on HKED technology

The first part of the project will involve developing a comprehensive report on the current status of HKED around the world, including the current users and applications of the HKED technology and the steps involved in the purchase, installation, and integration of a new HKED system. The report will draw upon research documented in pertinent papers and reference materials and information provided through existing collaborations, visits to facilities using HKED systems, and discussions with experienced users.

This report will cover several areas of significance: the extent of current applications; types of solutions measured; locations and numbers of units deployed; estimates of typical costs of equipment, installation, and training; assessment of possible applications related to GNEP processing facilities; discussion of system benefits to facility operations, users, and inspection agencies; and finally, current international cooperation opportunities.

2.2 Purchase of an HKED system

This task will include purchase of an HKED system and documentation of all the activities associated with the purchase. Purchase documentation will include an advanced procurement plan, purchase specifications, and statement of work.

The purchase specifications will provide all details required by the vendor to produce an HKED system that is compatible with the installation facility. In turn, the vendor will provide all specifications for the manufacture and production of the HKED system components and guidance on assembly and installation requirements.

2.3 Assembly and installation of an HKED system

The HKED system purchased for the HKED integration project will be installed in the Radiochemical Engineering Development Center (REDC) at ORNL (see Figure. 1). This part of the project will document activities associated with assembly and installation of the system. One significant challenge encountered with the installation of HKEDs is the

design of the interface between the hot cell and the sample transfer tube. In the case of the REDC installation at ORNL, the sample transfer tube will be designed by the vendor as specified by the facility. The facility staff will then devise a method for delivering the sample to the sample transfer tube.

A number of facility-specific requirements will have to be included for this particular installation. For example, the location designated for the installation of the HKED is a transfer cubicle used to bag out waste from hot cells. One of the facility-specific requirements is that the transfer cubicle must also be accessible for normal operations. This implies that the HKED system cannot by fixed in its location. The system, when not in use, must be “rolled” out of the way and stored in a location that does not interfere with regular transfer cubicle operations. This requirement introduces a number of other concerns regarding contamination, space, and transfer tube design which will have to be resolved before installation.

Radiation shielding calculations must be performed to protect staff and other personnel from radiation from the dissolver solution while it is outside the hot cell in the sample changer of the HKED. This issue will be addressed when radiation levels and isotopics of current dissolver solutions are determined. Other safety requirements will be assessed as the installation progresses.

2.4 Integration of HKED into the CETE Demonstration and future GNEP activities

HKED will be used to verify the U and Pu concentrations of process solutions from CETE spent fuel processing campaigns, including dissolver solutions typical of the input accountability tanks of reprocessing plants. All solutions will be measured using HKED, and current analyses by traditional mass spectroscopy will be available for comparison. The HKED technology will also be used to characterize samples from the processes for conversion of spent fuel, including the mixed actinide products. Experience gained with the HKED installation during the CETE Demonstration, in particular with respect to sampling methods and handling and evaluation of laboratory loads, can contribute to the design of future GNEP facilities. This measurement technique can then be qualified for use in ongoing and future GNEP work.

2.5 Provision of training and collaboration opportunities

Involving the HKED system with the ORNL Safeguards Laboratory, a DOE National User Facility, will facilitate training and access for DOE and contractor personnel, students, and foreign nationals. New and experienced users alike would benefit from coordinated training courses and workshops using actual process solutions. New users might include GNEP facility designers and/or operators, new professionals in the safeguards and reprocessing fields, new inspectors from various international inspection agencies, and students from universities around the world. Experienced users would benefit from training workshops that incorporate the new software and upgraded systems.

3.0 HKED APPLICATION DESCRIPTION

The proposed HKED system will be installed at REDC, an existing, fully operational nonreactor nuclear facility at ORNL (Fig. 1). The majority of the CETE testing and demonstrations (e.g., separations, conversion, and fuel fabrication studies) will be performed in the REDC facility during several CETE spent fuel processing campaigns. REDC is a Hazard Category II nuclear facility with

a highly trained staff that has performed similar nuclear operations and separation processing campaigns for decades.

Fig. 1. Radiochemical Engineering Development Center.

The availability of an HKED system at REDC will create an unprecedented opportunity for the training of students and international users. This installation will enable timely measurement of direct process solution samples, with essentially no sample preparation or subsequent waste issues, using technology that has been demonstrated in various facilities throughout the reprocessing community for the past 20 years.

4.0 HKED INTEGRATION PROJECT OUTCOMES

The following are among the specific outcomes anticipated from the HKED integration.

- A status report on existing HKED applications.
- Documentation of typical HKED purchase activities and requirements.
- Experience assembling and installing a typical HKED system.
- Documentation on installation issues and concerns.
- Measurement data quantifying U and Pu concentrations of CETE Demonstration spent fuel processing campaigns.
- Training materials for training courses.
- Training courses for domestic and international process monitoring teams, universities, facility operators, and safeguards agencies.
- Information for future GNEP facility design and sample handling.
- Working relationships and experience partnering with laboratory, industry, academic, and international organizations.

The following are the deliverables and broad schedule for the program.

FY08

- 2008 HKED implementation and applications report
- Purchase specifications and statement of work
- Purchase order
- Sample transfer tube design
- Delivery of components
- Draft training materials
- Draft installation paperwork

FY09

- Delivery of components
- Complete assembly of HKED system
- Complete installation of HKED at REDC
- Initial training course for ORNL users
- Collection of data during CETE spent fuel processing campaigns
- Second training course for ORNL users
- Training workshop for international collaborators and students
- Component upgrades
- Acceptance testing of upgrades
- Training workshop for international inspection agencies

5.0 SCOPE

With different short- and long-term goals involved with the HKED integration, the technical scope of work is expected to change periodically. The current focus of the HKED project is the physical installation. HKED installation is not facile and in fact will present the biggest challenge of the integration. Once the system is installed, however, the scope of work will shift to data collection for CETE Demonstration spent fuel processing campaigns and training initiatives. Based on data analysis, the technical scope related to the HKED will then shift to providing useful facility and sample handling designs for GNEP-related work. Lastly, the scope of work will focus on testing upgrades for existing systems.

6.0 CONCLUSIONS

The reintroduction of HKED to the United States is an exciting and promising endeavor. Not only does it imply that the United States is finally ready to take a step forward in the area of spent fuel reprocessing, but it will allow the United States to finally join the worldwide group of HKED users. As the purchase, assembly, and installation of the HKED progress, training materials will be drafted. This will allow ORNL to become a training facility for students and collaborators worldwide.

7.0 REFERENCES

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