Project Title: Radiochemical Analysis by High Sensitivity Dual-Optic Micro X-ray Fluorescence
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Lead Principal Investigator:
George J. Havrilla
Los Alamos National Laboratory
LANL MS G740
P.O. Box 1663
Los Alamos, NM 87545-1663
505-667-9627
havrilla@lanl.gov

Co-Investigator: (repeatable)
Ning Gao
X-Ray Optical Systems
15 Tech Valley Drive
East Greenbush, NY 12061
518-880-1500
ngao@xos.com

Graduate Students – 0

Research Objective

A novel dual-optic micro X-ray fluorescence instrument will be developed to do radiochemical analysis of high-level radioactive wastes at DOE sites such as Savannah River Site and Hanford. This concept incorporates new X-ray optical elements such as monolithic polycapillaries, which focus X-rays. The polycapillary optic can be used to focus X-rays emitted by the X-ray tube thereby increasing the X-ray flux on the sample over 1000 times. The polycapillary optic will also be used to collect the X-rays from the excitation site. This will effectively screen the radiation background from the radioactive species in the specimen. This dual-optic approach significantly reduces the background and increases the analyte signal thereby increasing the sensitivity of the analysis. This dual-capillary design is essentially a confocal (having the same foci) design, i.e. the detected X-rays are only emitted from the overlap of the two focal spots. This increases spatial resolution and reduces background. The integration of the X-ray optics increases the signal-to-noise and thereby increases the sensitivity of the analysis for low-level analytes. This work will address a key need for radiochemical analysis of high-level waste using a non-destructive, multi-element, and rapid method in a radiation environment. There is significant potential that this instrumentation could be capable of on-line analysis for process waste stream characterization at DOE sites.

Research Progress and Implications

As of May 2003 this report summarizes work from June 2002 through May of 2003, year 2 of a 3-year project. The project is more than 3 months ahead of the original milestone timetable.

After testing in spring of 2002, we discovered that the 100 mm focal length optics, which were specifically designed for this project, lacked sufficient sensitivity. In addition, the 100 mm optics lacked sufficient high energy X-ray throughput. New excitation and detection optics were designed with a compromise between the sensitivity needed (closer to the specimen) and the radiation burden (farther away). The new optics have focal distances at the sample of around 30 mm. The input excitation focal distance is nominally about the same at 55 mm, while the output focal distance for the detector optic was increased from 100 mm to 200 mm. This increase in detector focal distance allows for additional shielding to be placed between the sample and detector, along the optical path to reduce the radiation background. A smaller X-ray spot size of 100 micrometers resulted from the redesign.

The new optics breadboard system was tested with a model analyte system utilizing 1 M sodium chloride as a test matrix for the anticipated sodium hydroxide matrix from the sites. In addition, an iron 55 source was also used to determine the radiation background reduction with the dual capillary (confocal) optic design. The testing on the model analytes showed that the 30 mm focal distance provided parts-per-million sensitivity. The model analyte system consisted of a 100-ppm solution of lead, platinum, palladium, copper, strontium and molybdenum (as a substitute for technetium). All the target elements were detected below the 10-ppm level with a 600-sec acquisition time except for palladium due to the inability to effectively excite this element with the polycapillary optic.

The radiation rejection measurements showed a factor of 20 rejection ratio using an iron 55 source. Further improvements could be achieved with additional shielding. The new optics were incorporated into the solid figure design of the prototype instrument. The parts were ordered and machining on the housing was completed. The prototype is currently being assembled.

Overall the results are quite good. Due to changes in the original funding request and subsequent allocations the doubly curved crystal and on-site testing will not be feasible. The significance of this development work thus far is that this instrument will be useful for radioactive material characterization. The upper limit of an acceptable radiation level needs to be determined, however we expect that the confocal MXRF instrument will be successful in measuring elemental composition of radioactive materials. In addition, this confocal geometry opens new avenues of elemental analysis for general material characterization since it enables the acquisition of three-dimensional elemental information.

Planned Activities
06/01/03 - 10/01/03 – testing and evaluation of prototype
06/01/03 – 08/01/03 – complete prototype fabrication, deliver to LANL
09/01/03 – 12/31/03 – testing and evaluation at LANL, design of controlled hot measurements
01/01/04 – 06/01/04 – controlled hot testing at LANL

Information Access

OPTIONAL ADDITIONAL INFORMATION
OPTIONAL PROPRIETARY INFORMATION