LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

LLNL-TR-400056

Confocal μ-XRF for 3D analysis of elements distribution in hot environmental particles

M. Bielewski

M. Eriksson

J. Himbert

R. Simon

M. Betti

T.F. Hamilton

Paper submitted to the 2007 Annual Report of the Synchrotron Facility ANKA at the Forschungszentrum Karlsruhe, Germany

December 2007

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Lawrence Livermore National Laboratory is operated by Lawrence Livermore National Security, LLC, for the U.S. Department of Energy, National Nuclear Security Administration under Contract DE-AC52-07NA27344.

LLNL-TR-400056

Confocal μ-XRF for 3D analysis of elements distribution in hot environmental particles

M. Bielewski[1], M. Eriksson[2], J. Himbert[1], R. Simon[3], M. Betti[1], and T.F. Hamilton[4]

[1] European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany

[2] International Atomic Energy Agency, IAEA-MEL Laboratories, MC 98000 Monaco, Monaco

[3]ANKA – Institute for Synchrotron Radiation, FZK, D 76021 Karlsruhe, Germany

[4] Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94551-0808, USA

December 2007

Confocal μ-XRF for 3D analysis of elements distribution in hot environmental particles

M. Bielewski[1], M. Eriksson[2], J. Himbert[1], R. Simon[3], T. F. Hamilton[4], M. Betti[1]

[1] European Commission, Joint Research Centre, Institute for Transuranium Elements, P.O. Box 2340, 76125 Karlsruhe, Germany
[2] International Atomic Energy Agency, IAEA-MEL Laboratories, MC 98000 Monaco, Monaco
[3]ANKA – Institute for Synchrotron Radiation, FZK, D 76021 Karlsruhe, Germany
[4] Lawrence Livermore National Lab., P.O. Box 808, Livermore, CA 94551-0808 USA

Studies on the fate and transport of radioactive contaminates in the environment are often constrained by a lack of knowledge on the elemental distribution and general behaviour of particulate bound radionuclides contained in hot particles. A number of hot particles were previously isolated from soil samples collected at former U.S. nuclear test sites in the Marshall Islands and characterized using non-destructive techniques [1]. The present investigation at HASYLAB is a part of larger research program at ITU regarding the characterization of environmental radioactive particles different locations and source-terms. Radioactive particles in the environment are formed under a number of different release scenarios and, as such, their physicochemical properties may provide a basis for identifying source-term specific contamination regimes. Consequently, studies on hot particles are not only important in terms of studying the elemental composition and geochemical behaviour of hot particles but may also lead to advances in assessing the long-term impacts of radioactive contamination on the environment.

Six particles isolated from soil samples collected at the Marshall Islands were studied. The element distribution in the particles was determined by confocal μ-XRF analysis using the ANKA FLUO beam line. The CRL (compound refractive lens) was used to focus the exciting beam and the polycapillary half lens to collimate the detector. The dimensions of confocal spot were measured by "*knife edge scanning*" method with thin gold structure placed at Si wafer. The values of 3.1 x 1.4 x 18.4 μm were achieved if defined as FWHMs of measured L_α intensity profiles and when the 19.1 keV exciting radiation was used. The collected XRF spectra were analyzed offline with AXIL [2] software to obtain net intensities of element characteristic lines. Further data processing and reconstruction of element distribution was done with the software "R" [3] dedicated for statistical calculations. In figure 1 the distributions of Pu, Fe and Ti obtained for one of the studied hot particles are presented. The strongest signal was recorded for plutonium; the signals from iron and titanium are respectively 14 and 38 times less. It means that Pu is the most abundant of the observed elements. However, since the light elements are not detectable with the applied measurement conditions, it cannot be definitely stated if plutonium is the main element present in the sample. The isosurfaces are calculated at 20 % of maximum intensity for each element. Please note that the isosurfaces on the drawing are transparent. Changes in

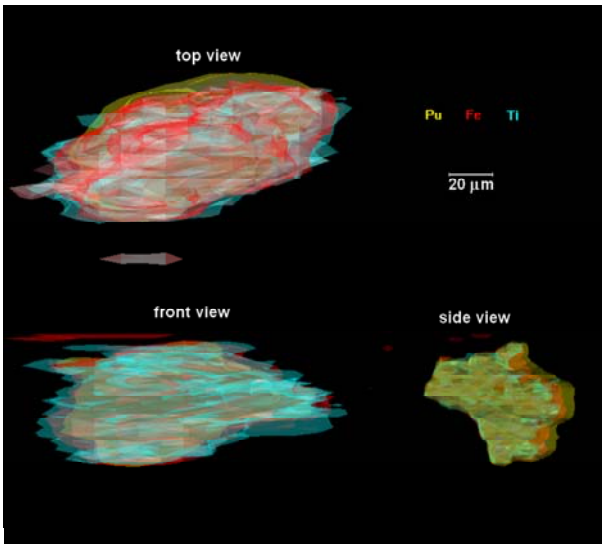

Fig. 1. Spatial distribution of selected elements reconstructed from the results of the confocal μ-XRF experiment for one of the particles.

the spatial distribution of Pu, Fe, and Ti within the particle are shown in Fig. 2a, 2b, and 2c. Distinct elemental patterns are clearly visible at the higher concentration levels. The distributions of Cr, Cu, and Pb were also reconstructed but the results are not presented here.

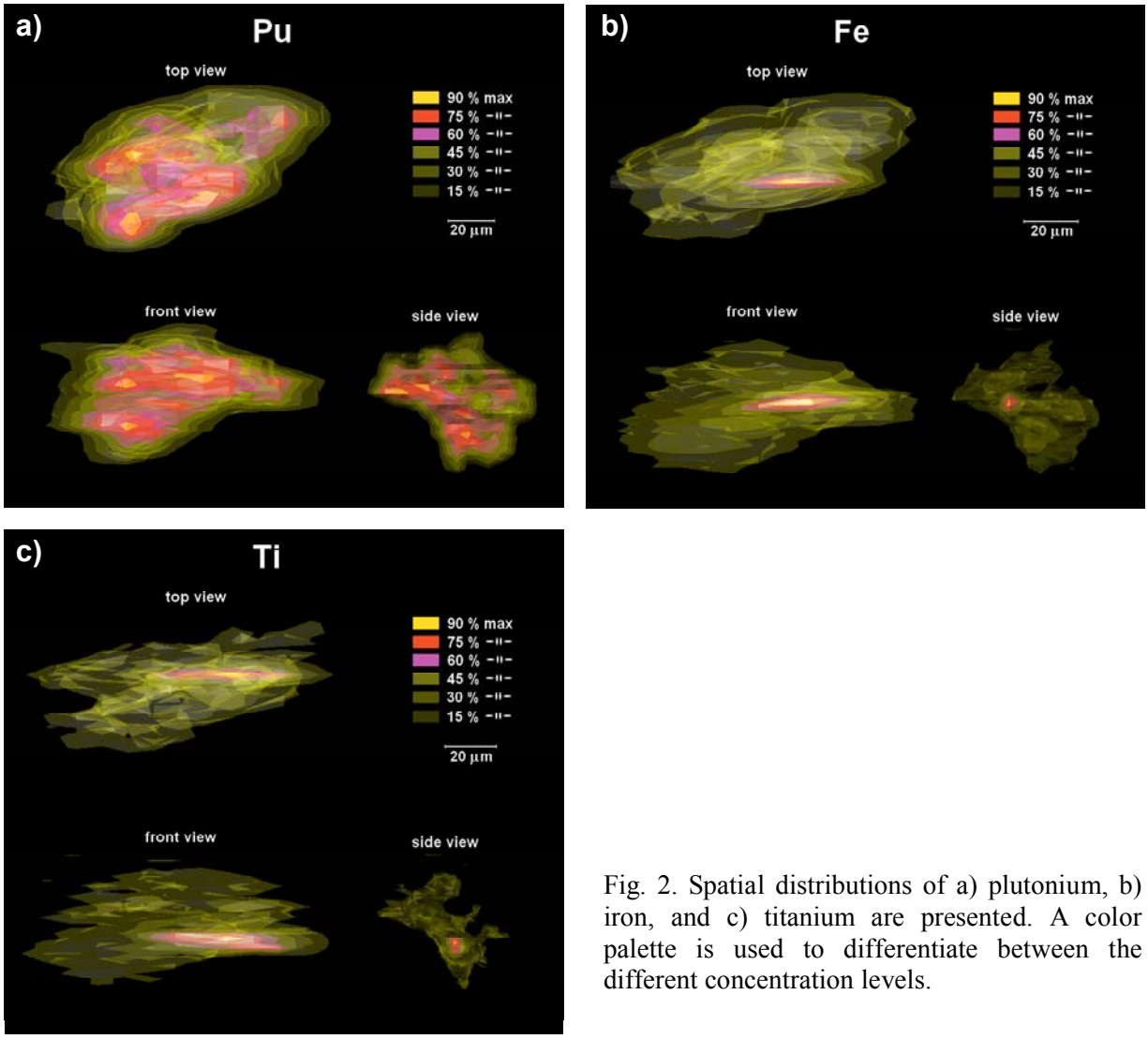

Fig. 2. Spatial distributions of a) plutonium, b) iron, and c) titanium are presented. A color palette is used to differentiate between the different concentration levels.

As it is shown in Fig. 1, the correlation between elements is good at low concentrations but the maxima of concentrations are not strongly correlated (see Fig. 2.). In general, the particle is inhomogeneous in terms of its elemental composition. Similar inhomogeneities were found for other particles with Pu identified as a major element in three of the six particles examined.

[1] Jernström J, Eriksson M, Simon R, Tamborini G, Bildstein O, Carlos Marquez R, Kehl S R, Hamilton T F, Ranebo Y, Betti M, *Spectrochim Acta B* 61 (2006) 971.

[2] XRF group, IAEA Laboratories Seibersdorf, QXAS Quantitative X-ray Analysis System, version 2.0, IAEA, Vienna, Austria, 2005. http://www.iaea.org/OurWork/ST/NA/NAAL/pci/ins/xrf/downloads/QXAS_Manual.pdf

[3] R Development Core Team, R Foundation for Statistical Computing, Vienna, Austria, 2006, ISBN 3-900051-07-0, http://www.R-project.org.

University of California
Lawrence Livermore National Laboratory
Technical Information Department
Livermore, CA 94551