IAEA NUCLEAR FORENSICS RESIDENTIAL ASSIGNMENTS

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Abstract

The IAEA Residential Assignment program places a technically qualified nuclear scientist at a leading nuclear forensic laboratory. The goal of this IAEA program is to enhance the skills and confidence of the resident scientist performing key tasks (such as analytical measurements) in support of a comprehensive nuclear forensic examination. These skills seek to improve th e knowledge and expertise available for nuclear forensic examinations at the respective home organization. To date, the IAEA Residential Assignment has been hosted by the Hungarian Academy of Sciences Centre for Energy Research (MTA EK), the European Commission's Joint Research Centre (JRC) and Lawrence Livermore National Laboratory (LLNL) through support of the U.S. Department of Energy's Office of Nuclear Smuggling Detection and Deterrence (DOE/NSDD). The paper seeks to elaborate on these three institutions models for hosting of scientists participating in the IAEA Residen tial Assignment program. Each model is developed alongside the partner country with specific project goals, deliverables, and desired outcomes. Ultimately, each participating host country and resident scientist will develop a strong collaborative relations hip that will continue far beyond the Residential Assignment.

1. INTRODUCTION

The IAEA Residential Assignment program provides an opportunity for scientists from around the world to collaborate with subject matter experts from leading nuclear forensics laboratories. The stated objective of the IAEA Residential Assignment program is for the resident scientist to learn nuclear forensics by researching relevant topics under the leadership of the hosting laboratory scientists who may also give an orientation to nuclear forensics within the context of that laboratories research program [1]. This knowledge exchange leads to scientists from both institutions collaborating on nuclear forensics engagements and co-authoring peer-reviewed scientific manuscripts. The topics explored in the Residential Assignments to-date have been wide ranging, exploring many nuclear forensics topics including, but not limited to, non-destructive source characterization, crime-scene management, and radio chronometry. The application of the Residential Assignment model has been successful in three different nuclear forensics laboratories thus far and has provided unique experiences to over a dozen scientists from around the world. Future Residential Assignment models will continue to be flexible and designed with the host institution and visiting scientists' interests and goals in mind in order to make this experience the most beneficial to all parties. It is this flexibility that makes the Residential Assignment program so successful to date.

2. 2. MODELS OF RESIDENTIAL ASSIGNMENTS

Each host organization developed a different model, or approach to the implementation of the IAEA Residential Assignment. This flexibility allows the host institution and the resident scientist to develop a mutually beneficial plan that can be executed successfully with potential implementation in the resident scientists' home institution. There is no one-size-fits-all approach to a Residential Assignment. Each host institution utilizes their capabilities, available instrumentation, and subject matter expertise in various nuclear forensics fields to design each project. The approaches used by three different nuclear forensics laboratories who have participated in the IAEA Residential Assignment program, Hungarian Academy of Sciences Centre for Energy Research (MTA EK), European Commission's Joint Research Centre (JRC), and Lawrence Livermore National Laboratory (LLNL) are described in detail below.

2.1. Hungarian Academy of Sciences – Centre for Energy Research

MTA EK in Hungary centred its Residential Assignment Program around a team exercise, involving real samples and fictious scenarios, and covered all aspects of a nuclear forensics examination, starting from radiological crime scene management and in-field categorization, planning, laboratory analysis, interpretation, use of a national nuclear forensics library (NNFL) and reporting, as required by law enforcement. Therefore, it is a "from-crime-scene-to-court" type model program. Hungary's approach to the IAEA Residential Assignment is designed for multiple resident scientists to participate together as a team. The Residential Assignment Program of the IAEA was piloted in Hungary in 2015. During the past 4 years nuclear scientists and police officers from Bulgaria, Croatia, Czech Republic, Ghana, Kazakhstan, Kenya, Lebanon, Malaysia, Romania, Slovakia, South-Africa and Thailand (altogether 13 persons) have participated in the program so far. During 2019 two representatives from the same Member State were involved into the program in the same period. The goal of this new direction is the integration of nuclear forensics in a country in a wider scale, as well as to build stronger cooperation within a state among stakeholders.

The main aim of the Residential Assignment Program in Hungary is to follow a nuclear forensics examination in a real time-frame (2 months) considering the international guidelines and standards of the IAEA and the Nuclear Forensics International Technical Working Group (ITWG) and using all the analytical techniques which are offered by the guidelines and available at MTA EK. In the program a model case exercise is used as part of the laboratory study to emphasize the key learning objectives. The goal is to guide participants through the entire examination to gain a better understanding of each element and the context of nuclear forensics. This procedure helps participants to become a nuclear forensics examiner by learning key elements of nuclear forensics and the specialized knowledge necessary to be successful.

The main structure of the Program implemented at MTA EK is based on Collaborative Material Exercises organized by ITWG and the Nuclear Forensics Methodology Training Course from the IAEA, but in a modified and extended version. It is a comprehensive Residential Assignment in nuclear forensics including theoretical education, practical exercises (non-destructive and destructive techniques with laboratory work) together with an overview on the Hungarian regulation and law enforcement including evidence handling techniques. Different authorities help the program like the Hungarian Atomic Energy Authority, Hungarian Police Criminal Technical Department (Crime

Scene Investigators) and the Traditional Forensics Institute. Participants have access to these organizations and have the opportunity to get to know their procedures, as well as the Hungarian national response activities. Besides, they also have the possibility to visit other facilities, like the Paks Nuclear Power Plant, other Hungarian nuclear reactors and some auxiliary analytical techniques such as the Transmission Electron Microscope, Neutron Tomography and X-Ray scanning systems.

FIG. 1. Use of different analytical techniques during Residential Assignment Program in Hungary (left SEM/EDS analysis; right: physical characterization in the glovebox)

FIG. 2. Radiological Crime Scene Management exercise and gamma-spectrometry training in the frame of the Residential Assignment Program in Hungary

The program incorporates the following topics:

— Generalized introduction to nuclear forensics supporting law enforcement investigations and nuclear security vulnerability assessments. Overview of the IAEA NSS-2G document.
— Introduction to national laws, legal instruments and evidence handling relevant to nuclear and other radioactive material out of regulatory control, as well as crime scene management. The assignment includes a practical introduction to the different techniques common in the nuclear forensic laboratory: gamma-spectrometry, neutron techniques, mass spectrometry, radiochemistry, X-Ray diffraction, X-ray fluorescence analysis and scanning electron microscopy.
— Exercises and detailed training: radiological crime scene management (how-to-collect the sample at the scene, proper official packaging and labelling, chain-of-custody, in-field categorization), physical characterization of the samples in a glove-box; gamma-spectrometric measurements using different types of detectors (planar, coaxial, well-type ones performed also in low-background chamber) and different type of

nuclear materials (e.g. natural, low-enriched and highly enriched uranium fuel pellets and powders, or uranium ore concentrates) as samples. An investigation of shielded materials by neutron tomography. Detailed analysis using SEM/EDX and X-Ray Diffraction systems. Techniques for sample preparation and special chemical separation methods, as well as ICP-MS measurements (precise isotope ratio and impurity analysis).
- Orientation to regulations pertaining to nuclear security relevant to the Hungarian Atomic Energy Authority on regulation connected to nuclear security and nuclear forensics.
- Overview of evidence handling techniques and traditional forensic investigations in Hungary.
- Visiting of other laboratories with demo measurements (TEM, portable XRF, LIBS and neutron tomography) and other facilities like Hungarian Police Traditional Forensics Institute, Paks Nuclear Power Plant or other Hungarian Nuclear Reactors.

Model case: determination of relationship among "unknown" nuclear samples found at "crime scenes" of the participants following injects and questions by law enforcement. Participants learn how to work in a team; how to plan and lead the analysis following hypothetical seizure scenarios and injects, model analytical data obtained; how to find relevant questions and answers. The exercise is followed by more injects where participants have their samples for nuclear forensics measurements, more traditional forensics, crime scene evidence, and analytical data are also given for further discussions.

The main steps of the Team Exercise are:

- Establishment of an analytical plan for a hypothetical seizure in teamwork, with the help of the nuclear forensics experts of MTA EK.
- Following a full nuclear forensics examination on a real timescale (2 months) from radiological crime scene investigation, evidence collection, and in-field categorization through laboratory measurements until origin assessment using the National Nuclear Forensics Library system of Hungary
- Findings of investigation: all the participants have their own sample which is similar or different from other samples of other participants. At the end of the program a special task for the participants is to investigate any relationship among the samples and determination of their origin using the Hungarian NNFL system.

The MTA EK program provides scientific background and technical experience for the participants and basic information and knowledge transfer. The main objectives of the program are to provide information on the technical infrastructure of a nuclear forensics laboratory, how to apply analytical techniques for nuclear forensics purposes, and how to gain experience in a full nuclear forensics examination (from crime scene until the origin assessment). This program also provides information on how to interpret the analytical results for nuclear forensics purposes (special way of thinking and experience), how to establish and use an NNFL, and how to share this information (ITWG, GICNT activities, joint publications, etc.). It also addresses the specific scientific interests of the participants, so although it has a planned, detailed, structured schedule, it is nevertheless flexible.

2.2. Joint Research Centre of the European Commission Residential Assignment

The JRC of the European Commission in Karlsruhe, Germany, hosted Andrei Apostol, a non-destructive analysis expert from the Horia Hulubei National Institute of Physics and Nuclear Engineering (IFIN-HH) in Romania, to participate in a scientifically challenging source characterization project. The objective of this work was to characterize californium neutron sources by determining their age and isotopic composition. These sources are used for a variety of purposes in industry, medicine, nuclear safeguards and geology. There have been incidents when californium neutron sources were found out of regulatory control. In such a situation, characterization of these sources and nuclear forensics analysis are required for tracing their history back to the place of origin or eventually to the point of loss of regulatory control [2].

High-resolution gamma spectrometry was used to determine the age and isotopic composition of californium sources during this Residential Assignment. The methods were tested using 5 californium reference sources at JRC-Karlsruhe. The method for age determination is based on measuring the activity ratio of a long-lived and a short-lived fission product of 252Cf. The isotopic composition was determined by directly measuring the activity of 249Cf and 251Cf and of the short-lived fission products that are in equilibrium with 250Cf and 252Cf. It has been demonstrated the gamma spectrometry can be successfully used to characterize californium sources for nuclear forensic purposes [3].

During the Residential Assignment, it was also investigated how the incomplete knowledge of the 250Cf/252Cf activity ratio affects this age-dating approach [3]. The work performed during this assignment are a benefit both nuclear safeguards and nuclear forensic experts. Given a unique nature and limited number of production facilities of californium sources, the non-destructive gamma spectrometric and modelling approach described in this work might provide significant information for the potential case investigation involving 252Cf sources. More information about the results of this Residential Assignment can be found in the resulting scientific publication [3].

2.3. Lawrence Livermore National Laboratory – Department of Energy

LLNL is part of a national laboratory complex which supports peer-to-peer collaborations with international partners in the area of nuclear forensics. These collaborations are often facilitated through the U.S. Department of Energy's Office of Nuclear Smuggling Detection and Deterrence (DOE/NSDD). To date, many peer-to-peer collaborations have been supported by LLNL including joint sample analyses, staff exchanges, and workshops. It is through this peer-to-peer engagement mechanism with NSDD that a collaboration was initiated with Argentina, a country with decades of experience in research and development in the nuclear energy field. Argentina has well maintained facilities with advanced instrumentation, technical experts, and research knowledge of all areas of the fuel cycle. This peer-to-peer engagement objective was designed to pursue mass spectrometric analyses of a certified reference material (CRM) 125-A under the stewardship of LLNL nuclear forensics analysts [4]. The radio chronometry reference material CRM 125-A is a 4.2% enriched uranium dioxide pellet (UO2) that is certified for uranium isotopic composition as well as a model purification date based on the 230Th/234U (daughter/parent) chronometer [5]. The research model for the first IAEA Residential Assignment held in the U. S. was framed in the context of this on-going joint sample analysis engagement between DOE/NSDD and the National Atomic Energy Commission CNEA in Argentina.

Marta Bavio, a mass spectrometrist from CNEA, was selected by the IAEA as the resident-scientist. The goal was for her to obtain a deepened understanding of the radio chronometry and trace elemental analysis techniques applied to nuclear forensics samples during her 3-month Residential Assignment with the nuclear forensics group at LLNL. Marta was immersed in the day-to-day operations of a nuclear forensics laboratory. Marta's Residential Assignment at LLNL centred around characterizing the trace element composition, isotopic composition, and model-age determination of CRM 125-A. Marta took the necessary training to be fully compliant with the health and safety regulations of LLNL's chemical and mass spectrometry laboratories. This allowed for a hands-on experience with the most direct observations and results gathered by Marta, herself. Fig. 3 shows Marta conducting chemistry in a LLNL nuclear forensics laboratory and the LLNL scientists who supported the IAEA Residential Assignment.

FIG. 3. (left) Marta Bavio examining CRM 125-A solutions. (right) The team of LLNL scientists who supported the IAEA Residential Assignment and Marta Bavio (centre).

Marta's Residential Assignment was designed like a nuclear forensics examination in that the sample under examination, CRM 125-A, followed an analytical plan that collected data using non-destructive techniques followed by destructive techniques. Marta worked with subject matter experts to collect morphological data including scanning electron and optical microscopy and physical measurements. Next, Marta dissolved subsamples of CRM 125-A for destructive analyses. She did this work according to the standard operating procedures at LLNL, with input from her experiences at CNEA. Marta learned how to calibrate isotopic spikes using multiple calibrants and determine the isotopic composition of uranium, thorium, and protactinium by performing isotope dilution by mass spectrometry in order to calculate the Th/U and Pa/U model-ages. Because CNEA has access to both single- and multi-collector mass spectrometers, part of the Residential Assignment was to determine model-ages using both types of instruments. Scientists from CNEA and LLNL were interested in determining the model-age precision achievable for single- vs. multi-collector instruments.

We performed 230Th/234U and 231Pa/235U analyses of CRM 125-A using three different instruments at LLNL: (1) Thermo Scientific iCAP-Q quadrupole single-collector inductively coupled plasma–mass spectrometer (Q-ICP-MS), (2) Thermo Scientific Element XR high resolution single-collector ICP-MS (HR-ICP-MS), and (3) Nu Plasma HR multi-collector ICPMS (MC-ICP-MS). The same sample aliquots, isotopic tracers, QC reference materials, and data reduction algorithms were used in the collection and processing of data generated by each instrument, allowing for direct comparison of model-age results amongst the three mass spectrometer designs [7]. A subset of the results from Marta's Residential Assignment can be seen in Fig 4. The Pa/U model-ages are plotted relative to the [Th/U] certificate model-age of CRM 125-A. All trace element, isotopic, and model-age results Marta gathered were consistent with both certificate values and with historical data collected at LLNL on additional subsamples of CRM 125-A. These results are important for the radio chronometry community in order to establish quality control reference materials for the 231Pa-235U chronometer.

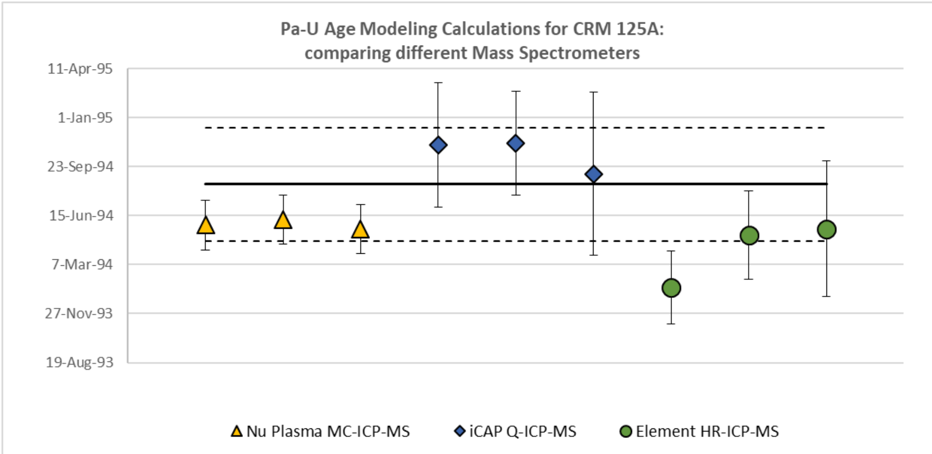

FIG. 4. Final 231Pa/235U model-ages for CRM 125A; a comparison of results collected from three different instruments: Nu Plasma MC-ICP-MS, iCAP Q-ICP-MS, and Element HR-ICP-MS.

The Residential Assignment sought to enhance the skills and confidence of a scientist from CNEA in performing isotopic and elemental concentration measurements by mass spectrometry in support of a nuclear forensics examination. Marta wrote a final report for IAEA and gave a presentation that summarized her findings and lessons learned during the Residential Assignment. The analytical work performed by Marta and the nuclear forensics analysts at LLNL led to participating in a panel discussion at the IAEA Technical Meeting on Nuclear Forensics in April of 2019. Their work also led to participating in the RANC conference in Budapest, Hungary in May of 2019. Finally, this work was published in the Journal of Radioanalytical and Nuclear Chemistry Journal in October 2019. Another tangible outcome to-date is LLNL scientist's support of Marta Bavio in the pursuance of her PhD in nuclear forensics from university in Argentina. For more details on the technical contributions of this Residential Assignment, see [6].

3. CONCLUSIONS

The IAEA Residential Assignment fosters new or existing nuclear forensics collaborations between the host organization and the home organization of the resident scientists. All resident scientists emphasized that in addition to advancing their analytical skills in the area of nuclear forensics, their experiences in the IAEA Residential Assignments resulted in professional and personal connections that will help them build a long-term international network of nuclear forensics colleagues. Ultimately, the IAEA Nuclear forensics Residential Assignment is a novel approach to sustainable nuclear forensics capacity building that aims to benefit all parties involved.

ACKNOWLEDGEMENTS

The authors of the paper would like to acknowledge the guidance and support of David K. Smith and Jerry Davydov of the IAEA Department of Nuclear Safety and Security. Lawrence Livermore National Laboratory would like to acknowledge the support of U.S. Department of Energy's Office of Nuclear Smuggling Detection and Deterrence. This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344. Releasable to external audiences, LLNL-CONF-796917.

REFERENCES

[1] IAEA, Division of Nuclear Security, Department of Nuclear Safety and Security. Residential Assignment for Human Capacity Building in Nuclear Forensic Analytical Measurements. Additional Information Sheet.

[2] APOSTOL, A., ZSIGRAI, J., BAGI, J., MAYER, K., Investigation of parameters for nuclear forensic characterization of 252Cf sources. European Commission, Karlsruhe, 2017, JRC109568
[3] APOSTOL, A.I., ZSIGRAI, J., BAGI, J. et al., J Radioanal Nucl Chem (2019) 321: 405. https:/doi.org/10.1007/s10967-019-06628-0
[4] KIPS, R., TREINEN, K., LINDVALL, R., BAVIO, M., KRISTO, M., Nuclear Forensics Capacity Building at LLNL through Peer-to-Peer Engagements. (2018) Institute of Nuclear Materials Management proceedings.
[5] New Brunswick Laboratory, Department of Energy, Certified Reference Material 125-A. 1997
[6] TREINEN, K, SAMPERTON, K., LINDVALL, R., WIMPENNY, J., GAFFNEY, A., BAVIO, M., BARANSKY, E., WILLIAMS, R., Journal of Radioanalytical and Nuclear Chemistry,(2019)Evaluating uranium radiochronometry by single-collector mass spectrometry for nuclear forensics: a multi-instrument investigation, DOI: 10.1007/s10967-019-06832-y, pg.1-14.