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Development of Nuclear Forensics Capabilities at CNL

Canadian Nuclear Laboratories (CNL) has a long history of research and development in nuclear fuels as well as expertise in handling and characterization of radioactive materials. CNL has been leveraging its knowledge base and specialized facilities to support the enhancement of Canadian nuclear forensics capabilities. Most of this work is conducted through projects within the Federal Nuclear Science and Technology (FNST) Work Plan, a program established and managed by Atomic Energy of Canada Limited to facilitate work at CNL to contribute to the Government of Canada's health, science, innovation and climate change objectives. In addition, CNL also performs work in partnership with Government of Canada departments and agencies under the auspices of the Canadian Safety and Security Program, administered by Defence Research and Development Canada's Centre for Security Science, which funds science and technology initiatives to enhance Canada's preparedness for prevention of and response to potential threats.

Nuclear forensics (NF) uses a wide range of techniques to characterize materials in the context of an interdiction and subsequent possible legal or national security investigation. Considerations are given to safety, preservation of evidence and ultimately may include a wide range of non-destructive and destructive analyses in the context of nuclear security event involving nuclear and/or radioactive material. Materials of interest may include nuclear materials as well as radioactive sources used for medical and industrial purposes. CNL's program of work on nuclear forensics focuses on several areas, including development of new methodologies, determination of signatures of nuclear materials and radioactive sources, and providing support to the national NF capability.

Signatures of Radioactive and Nuclear Materials

Signatures of radioactive and nuclear materials can provide investigators with useful information about materials such as its origin, fabrication process, use, or history. Thus, one area of work for CNL is to investigate novel signatures of radioactive sources, irradiated and unirradiated fuels, and irradiated reactor alloys.

Age dating of radioactive sources is an important component of nuclear forensics as it aids in the determination of the provenance and origin of the materials. Cesium-137 is one of the most common radionuclides used in sealed radioactive sources. For 137Cs radiochronometry, the separation of 137Cs and 137Ba prior to analysis of 137Ba is required as the stable daughter products must be measured by mass spectrometry. The manual separation techniques are advantageous for ease of set-up in hot cell facilities, however they are typically time intensive and separation efficiencies can be operator dependent. CNL evaluated a radiochronometric method for 137Cs sources using Ion Chromatography (IC) for the separation of barium and cesium which would allow for automation of the separations and potentially a reduction in time and operator error. The performance of the IC separation was investigated through the use of inactive standard materials and analysis performed by inductively coupled plasma – mass spectrometry (ICP-MS). The study found that while separations of cesium and barium can be achieved by IC even in complex radioactive solutions, the limitation of the method's operating concentration range is not compatible with the concentrations of 137Ba in the low activity (or diluted) sources that can be handled outside a hot cell. The dilutions required to enable sample handling outside of the cell often brings the Ba concentrations into the very low ppb range, in which, at these levels interferences from natural occurring Ba in the environment can become problematic. Any method developed for radiochronometry needs to be robust for various complex matrices, large concentration ranges and suitable for various installation requirements (e.g., fume hoods and hot cells).

CNL has extensive information gained from post-irradiation examination (PIE) of nuclear fuels. Signatures and trends in PIE data have been analyzed using human computed techniques by investigating correlations with physical meanings. However, it is expected that statistical methods, such as principal components analysis, can be utilized to determine additional correlations which enable attribution. The work involves scoping the best approach to applying statistical methods to fuel signatures using available measured data, not only in the PHWR industry but also in the LWR industry. The feasibility of using simulated data is being explored as well as data pre-treatment automation, to minimize necessary labour. Algorithms are being developed and applied to determine the level of confidence in attributions.

Part of the NF work at CNL is the investigation of isotopic signatures of nuclear fuel materials of interest to Canada including power reactor, research reactor and other experimental fuels. Fuel types that have been investigated previously include uranium-based and mixed-oxide fuels, and the current focus is research reactor fuel with thorium-based fuels being considered for future work. An investigation into possible signatures of unirradiated fuels has also been initiated.

CNL has experience measuring nuclide ratios to estimate the fluence of reactor alloys for supporting ageing management and life extension. These techniques are being adapted for application to nuclear forensics. The focus is on materials that may be sampled during the facility operating life (e.g., structural materials, control or absorber rods, scrape samples, etc.). Materials of potential application include aluminum, stainless steel, zirconium, and Inconel alloys. Various samples of irradiated reactor alloys from reactors NRU and NRX with a known operating history and original chemical compositions are available for analysis and method development. Simulations of their operating history will be done using reactor physics codes, and samples of these materials will be analyzed using mass spectrometry to validate the isotopic ratios of selected elements of interest obtained from the simulations. Once validated, the goal is to use these isotopic ratios to "back-calculate" the irradiation history of the material which can then be compared to the operating history of the reactor and thereby validate the methodology.

Novel Non-Destructive Techniques

There is a need to develop capabilities to identify the origin of undeclared nuclear materials through improved detection methods, specifically novel non-destructive techniques (NDT) and characterization methods for powders, in the context of nuclear forensics. CNL has been supporting the Canadian effort in nuclear forensics including participation in national and international exercises that have used a variety of non-destructive and destructive techniques as well as specialized knowledge such as nuclear fuel expertise. The non-destructive techniques used at CNL include a variety of instruments in surface science (e.g., SEM/EDX, TEM, XPS, Auger, and SIMS), material science (e.g., neutron imaging, activation, and diffraction) and radiochemical analysis (gamma spectroscopy, mass spectrometry). A number of these techniques were applied to an unknown legacy material to enable full accountancy and safe, secure disposition. Subsequent verification of the NDT results through destructive analysis and mass spectrometry supported the value of the NDT analysis. CNL is currently looking to systematically evaluate the current state of various NDTs and explore the potential of existing and emerging techniques in the context of supporting nuclear forensics. Opportunities for collaborations with Canadian institutions is also being explored to expand the techniques available to CNL for nuclear forensics applications.

Some materials of interest to NF are found in powder form, such as uranium ore concentrates, uranium dioxide powder, radioactive sources (e.g., CsCl and SrCl salts), and post-detonation debris particles. As new fuels are developed for advanced reactors, such as small modular reactors, novel characterization techniques may be required for powders (e.g., fuel salts). There are a number of techniques that can be applied, including particle size measurement and morphology characterization techniques. Collaboration with international organizations, which can be in the form of benchmarking of the different techniques, is being planned.

Conclusion

CNL is continuing development work on new and innovative methodologies as well as leveraging its knowledge base and specialized facilities to support the enhancement of Canadian nuclear forensics capabilities. Work is being conducted on a wide variety of materials of interest including nuclear materials and radioactive sources used for medical and industrial purposes that may be found pre- or post-dispersion. CNL's program of work on nuclear forensics focuses on several areas, including development of new methodologies as applied to radiochronometry, determination of signatures of nuclear materials and radioactive sources, and application of non-destructive techniques.

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