National nuclear forensics libraries: a case study on benefits and possibilities for identification of sealed radioactive sources

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Abstract

A National Nuclear Forensic Library (NNFL) is a useful nuclear forensics tool which consists of information and subject matter expertise on radioactive and nuclear (RN) materials produced, used or stored within a State. If RN material is found out of regulatory control the NNFL can be used as part of a nuclear forensics investigation to help identify whether or not the material is consistent with a country's national holdings. In previous work, a number of signatures which can be useful to identify sealed sources of ^{241}Am were investigated. To validate the measurement results, an official query concerning information about two of the previously investigated ^{241}Am sources was sent to the United States Department of State, the international point-of-contact (POC) for the U.S. NNFL. The aim of this work is to show how data obtained in a characterization of a radioactive source can be used in conjunction with an NNFL to investigate the history of a source out of regulatory control.

Keywords National nuclear forensic library · Nuclear forensics · Radioactive sources

Introduction

Nuclear security deals with both nuclear and other radioactive (RN) materials and is an area of interest and ongoing concern in the international community. One aim of nuclear security is to prevent and deter the malicious use of RN materials [1]. Nuclear forensics, a subdiscipline of forensic science dealing with the examination of RN material, can be an important component of a nuclear security regime. Nuclear forensics consists of a wide variety of analytical methods and instruments which, often in combined use and after analysis of a subject matter expert, may help to determine signatures or characteristics of the materials, i.e. the identity of the materials, how, when and where the materials were made, and their intended use [2, 3].

One tool that can be used as part of a nuclear forensics investigation and help determine the history of RN material found out of regulatory control is a National Nuclear Forensics Library (NNFL) [2, 3]. As previously stated [4]:

An NNFL consists of scientific and technical, regulatory, administrative and other sources of data and information on RN materials produced, used or stored within a State; and is complemented by subject matter expertise to support the examination of RN material samples, to evaluate data and information, and to draw conclusions regarding material provenance.

Information about RN material, obtained either from analysis of the material or from certificates, can be used to populate an NNFL. At the Nuclear Security Summit in 2016, the U.S. announced that [5]:

A country's NNFL will likely contain sensitive or proprietary information and thus NNFL content and access must be protected by the government. At the same time, exchanging queries about the material information or expertise maintained in a library—without divulging detailed information on library or database content—is an effective means to facilitate a more comprehensive approach to using materials information to support an investigation. [...] The U.S.

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Department of State is the national POC for the U.S. NNFL and is ready to receive queries through diplomatic channels.

^{241}Am is an alpha-emitting radionuclide that is often used in industrial applications in e.g. density and level gauge instruments. ^{241}Am is also used in ionizing smoke detectors. In combination with beryllium, ^{241}Am can be used as neutron sources for well logging and moistures gauge instruments. In previous work, possible signatures of sealed ^{241}Am gamma sources were investigated using gamma spectrometry as the measurement technique [6]. Among the investigated signatures were impurities and age. During these investigations, it was determined that the sources emitted neutrons, which is a sign that there are low-Z impurities such as sodium contained within the sources. These impurities are most likely originating from the construction material of the source. These elements can undergo (α, n) reactions, which may result in the emission of characteristic gamma photons. Other impurities are isotopic such as ^{243}Am , which is a result of the difficulty to chemically separate isotopes of the same element. Peaks from ^{239}Np , a daughter product of ^{243}Am , were observed in the spectra in some of the sources. Another investigated signature was the age, or separation date, of the sources. The age, in this respect, is the time that has passed since the americium was last chemically separated and can be determined by the relation between the activities of ^{233}Pa , a daughter product of ^{241}Am , and ^{241}Am , which both can be found in the gamma spectrum. According to our previous studies, the ages of the examined sources varied between 12 and 44 years. For one of these sources the age was known and agreed well with the calculated value.

In support of efforts to validate the results of the previous work, the United States provided an opportunity to make an official inquiry to the U.S. NNFL using the model process announced at the 2016 Nuclear Security Summit [5]. Two

of the sources from the previous study were chosen for this inquiry.

The aim of this work is to show how measurement data can be used in combination with an NNFL to help investigate the history of a sealed source. The aim is also to show how the NNFL can benefit from comparing the NNFL data to measured data to establish whether the data in the library is relevant to its task. Moreover, this work also points out some possible characteristics of radioactive sources that may be useful to include in an NNFL and that the level of detail in an NNFL may differ depending on the needs of the State.

Materials and methods

Source descriptions

Two of the sources from the previous study were chosen for this work. A query to the U.S. NNFL was initiated by submitting a completed query template [7] together with images of the sources and the previously published paper to the U.S. NNFL international point-of-contact (POC) at the U.S. Department of State. A selection of the technical information provided to the U.S. NNFL as part of the query can be found in Table 1. This information was used to build the search process described below. The number of encapsulations in Table 1 refers to how many layers of protective barriers there were. Double encapsulation decreases the risk of contamination but single encapsulation decreases the shielding of the source, which can be useful for low energy nuclides [8]. The purpose of the query was to investigate whether the U.S. NNFL could provide any information of similar sources with similar labelling and known history that would help to verify the separation dates.

Table 1 Information provided in the NNFL query

Forensic signature	Source 1	Source 2	Source of information
Isotope	^{241}Am	^{241}Am	Image, measurement
Nominal activity	185 GBq (5 Ci)	185 GBq (5 Ci)	Image
Label	AI 1234 AB	T.R.C. 5678 CD	Image
Labeling location	Capsule Lid (or bottom)	Capsule Lid (or bottom)	Image
Number of encapsulations	Assumed single	Assumed single	Estimation
Outer capsule dimensions, diameter $\times$ thickness	45 $\times$ 8 mm	45 $\times$ 8 mm	Measurement
Source shape	Disk	Disk	Image
Wall thickness dimension	5 mm	5 mm	Estimation
Material dimension diameter	35 mm	35 mm	Estimation
Separation date	January 6 1982 $\pm$ 2 y	July 18 1972 $\pm$ 2.6 y	Measurement

The sources have been given fictional labels apart from the first letters AI and T.R.C., respectively, to conceal their true identity. Measurements refer to [6]. The separation date refers to the date at which the last chemical purification was performed

Description of US NNFL

The U.S. National Nuclear Forensics Library serves as the centralized U.S. Government (USG) capability to catalog and interpret forensic data for RN materials used, produced, and stored within the United States. When a nuclear security event involving material out of regulatory control occurs, the U.S. NNFL helps answer:

- Is the unknown sample consistent with U.S.-origin material?
- Is the unknown sample consistent with imported nuclear material held in the United States?

As stated in [5]:

When submitting queries to the U.S. NNFL, the USG requests that the government submitting the query provide details about the material in question and a statement that the query is in support of a law enforcement or regulatory investigation. These details may include categorization information such as, but not limited to, a physical description of the material (amount, color, physical form, etc.), a radiological assessment (type/s of radiation present, total activity, dose rate, etc.), a description of any containment or packaging, and copies of any accompanying documentation or identifying information. Additional characterization information such as analysis details, isotopics, and chemical composition may also be requested, but are not always required.

The U.S. NNFL is supported by rigorous laboratory capability to generate data through forensic examinations of RN materials. Data from forensic analyses are stored in the U.S. NNFL, a federated system of databases. For the Sweden query, the Radiological Sealed Source Library, the radiological material component of the U.S. NNFL, was used.

Radiological sealed source library analysis

A series of search parameters was used to interrogate the library and narrow the number of possible sources. An initial search based on label information did not return any useful results. Therefore an iterative search process was constructed such that the library was firstly searched with respect to the isotope ^{241}Am and a maximum activity of 185 GBq. The hits from this search were then searched with respect to outer dimensions of the source including a 10% uncertainty in thickness and diameter.

Results and discussion

The search results were compared to each of the sources independently. A summary of the search process can be seen in Fig. 1.

First, the database was carefully searched for the labels 1234 AB and 5678 CD on the sources (the labels used in this paper are fictitious to conceal the identity of the

Fig. 1 A schematic picture of the database search

sources). This search did not result in any hits, and it was assumed that the labels on the sources are serial numbers rather than model numbers, as the library did not contain information on these serial numbers. In the next search, the library was downselected based on the isotope and maximum activity. This search returned 14 possible model numbers, manufactured by 12 different companies. In order to further reduce the number of possible sources, the results were compared to the outer dimensions, including a 10% uncertainty in dimensions. This search resulted in two source models:

- Am1.G66 manufactured by BEBIG Isotopentechnik und Umweltdiagnostik GmbH
- AMC.50 manufactured by Amersham Plc and QSA Global, Inc.

The wall thickness of the sources was compared to the information on this parameter in the two library sources. The wall thickness of AMC.50 is 2.5 mm which is half the thickness compared to the estimated wall thickness of the two sources investigated. Since the wall thickness was an MCNP simulated parameter, the AMC.50 was not excluded despite the differing thickness. Am1.G66 did not have any information on wall thickness. The fact that this model did not have any information on wall thickness was, however, not a reason for exclusion.

The labeling scheme for the two downselected models were compared to the two investigated sources. While there was no resemblance between the Am1.G66 model and the two sources, the labeling scheme for the other model, AMC.50, showed consistence to the source labeled AI 1234 AB. The labeling scheme for the AMC.50 model should include nuclide, activity, serial number and the letters AI (representing the manufacturer Amersham International) on the capsule lid. The other source, T.R.C 5678 CD shows the same consistency to the library entry, with the exception that the label T.R.C. is different from AI. (*Note: The U.S. NNFL contains information on the labeling schemes for the Am1. G66 and AMC.50 models because they were licensed for production or distribution in the U.S.*)

To further investigate whether or not this inconsistency fits with historical events, the background of Amersham International was explored. Research into the company revealed that the Radiochemical Centre (TRC) site in Amersham, was formed as a part of the UK Atomic Energy Agency, UKAEA, in the late 1940s. [9] In 1966 Amersham Plc. installed a cyclotron and began producing radioisotopes for nuclear medicine and advancing their encapsulation technology at the site. Following the Atomic Energy Authority Act 1971, UKAEA transferred its functions into companies. Among these companies, The Radiochemical Centre Limited (TRC Ltd.) was formed. In 1982 TRC Ltd.

was privatized and the name was changed to Amersham International Limited.

Forensic signatures identified for Source 1 are consistent with the forensic signatures associated with Amersham International's source model AMC.50, with the exception of the estimated wall thickness. The measured separation date of Source 1, January 1982, is consistent with the engraving AI on the capsule lid and the information in the NNFL. Since the separation was done in January 1982, the source itself is most likely younger and therefore falls within the date range of 1982 to 1998 when Amersham International was manufacturing ^{241}Am sources of similar form to Source 1.

Similar to Source 1, the forensic signatures identified for Source 2 are also consistent with Amersham International's AMC.50 source model number, with the exception of the estimated wall thickness and the manufacturers engraved initials. Source 2 has a similar labeling location and scheme (activity, nuclide, manufacturer's initials, and serial number) except it has the initials T.R.C. instead of AI. T.R.C. is consistent with an abbreviation for The Radiochemical Center, but this labeling system could not be confirmed using information within the NNFL. Assuming The Radiochemical Centre, the predecessor to Amersham International, produced Source 2, the measured separation date of July 1972 is consistent with the manufacturer's existence from 1966 to 1982. This thorough examination of the history of Amersham International is an example of the need for subject matter experts who can interpret the data present in a nuclear forensic investigation and connect the measurements to the information contained in the NNFL.

In addition to information on the labeling scheme, the U.S. NNFL also contains details regarding the chemical form of the AMC.50 source model. According to the library, the chemical form of the americium source is americium oxide mixed with powdered silicon, sodium and magnesium. This corresponds well with the two investigated sources as measurements showed presence of both sodium and magnesium as well as silicon in these sources [6].

Conclusions

This work has shown that by comparing measured characteristics of sealed sources with entries of similar sealed sources in an NNFL, it is possible to find potential matches in the library that could help identify the origin of an unknown source. The labeling of the source together with the dimensions and chemical composition could, in this case, be used for narrowing down the number of possible sources in the library to one single source model. Also, by following the history of the production company, it appears most likely that the investigated sources have the origin that has been

suggested in this work. It cannot, at least, be ruled out that the sources originate from the identified manufacturers. It may be noted, however, that in other cases the information from radioactive source suppliers may not be as readily available due to e.g. nondisclosure agreements and other proprietary concerns.

This work also shows that the information in the NNFL is relevant for its purpose, since it is possible, in this case, to identify a manufacturer using information about dimensions, radioactive isotope and nominal activity. Depending on the intended purpose and utility of a NNFL, the level of information detail in the library may need to vary. If it is only necessary to identify a manufacturer, basic manufacturing characteristics may be sufficient, whereas, if the intent is to identify an individual source, it could be important to include regulatory information such as serial numbers. If it is also important to identify an individual source if the serial number for some reason is missing or unavailable, the library needs to be populated with data that can be obtained by measurement, such as the age of the source and elements and isotopes other than the nominal radioactive material, that are characteristic for a specific source. This is, after all a matter of priorities; the work needed to characterize sources in a library to the extent that each and every source in the library can be identified under a vast number of scenarios may be extensive. Therefore, the level of detail needed to make the library useful for each country needs to be determined for each individual NNFL.

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Declarations

Conflict of interest The authors declared that there is no conflict of interest.

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