



## Nuclear forensic analysis of an unknown uranium ore concentrate sample seized in a criminal investigation in Australia



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### ABSTRACT

Early in 2009, a state policing agency raided a clandestine drug laboratory in a suburb of a major city in Australia. During the search of the laboratory, a small glass jar labelled “Gamma Source” and containing a green powder was discovered. The powder was radioactive. This paper documents the detailed nuclear forensic analysis undertaken to characterise and identify the material and determine its provenance. Isotopic and impurity content, phase composition, microstructure and other characteristics were measured on the seized sample, and the results were compared with similar material obtained from the suspected source (ore and ore concentrate material). While an extensive range of parameters were measured, the key ‘nuclear forensic signatures’ used to identify the material were the U isotopic composition, Pb and Sr isotope ratios, and the rare earth element pattern. These measurements, in combination with statistical analysis of the elemental and isotopic content of the material against a database of uranium ore concentrates sourced from mines located worldwide, led to the conclusion that the seized material (a uranium ore concentrate of natural isotopic abundance) most likely originated from Mary Kathleen, a former Australian uranium mine.

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### 1. Introduction

In May 2009, officers from a state policing agency in Australia requested that the Australian Nuclear Science and Technology Organisation (ANSTO) provide identification and provenance of an unknown radioactive powder sample seized in a raid on a clandestine drug laboratory. The sample was found and transported to ANSTO in a small glass jar (Fig. 1). No radioactive contamination was detected on the outside of the jar.

The analytical sequence initiated by ANSTO generally followed that proposed by the International Atomic Energy Agency’s (IAEA) model action plan (MAP) [1,2]. For this investigation, mostly comparative signatures were used; that is, comparisons were made between the material characteristics of the seized sample and those same characteristics for samples of known origin. In this way, analytical data were interpreted to provide information to the state police on the material’s

identification and provenance. Previous research has been conducted by ANSTO and others into the assessment of various signatures for origin determination of uranium ore concentrates (UOC). Particularly valuable measurements for attributing UOCs have included isotopic ratios of Sr, Nd, and Pb, the level of elemental and anionic impurities, and the pattern of rare earth element concentrations [3–7]. Lawrence Livermore National Laboratory (LLNL) has been working in this field for some time and has developed an extensive database of analytical measurements on UOC samples from around the globe (as well as an archive of physical samples) with which to compare unknown UOC samples. They have developed the Discriminant Analysis Verification Engine (DAVE) algorithm, based upon the iterative application of partial least squares discriminant analysis (PLS-DA), designed to assist matching of the characteristics of an unknown UOC with signatures contained in the database [8–10]. A sub-sample of the seized material was sent to LLNL for comparison against the uranium sourcing database using DAVE, confirmation of the analytical results obtained at ANSTO, and additional characterisation of the material as required.

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**Fig. 1.** Glass jar labelled “Gamma Source” containing radioactive material received by ANSTO.

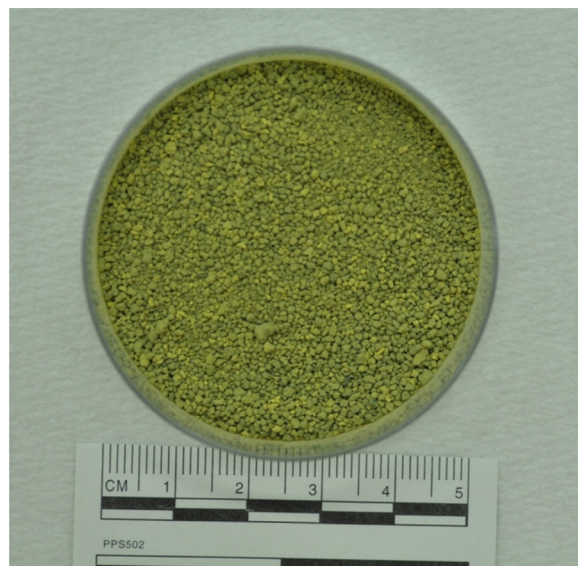
## 2. Materials

An unknown radioactive powder sample was provided to ANSTO by an Australian state policing agency. It was given the identification NSR-F-130509, which is used throughout this paper. The sample was contained in a small, clear glass jar with a metal screw-top lid (Fig. 1), identical to a Kraft Vegemite™ jar. The glass jar contained 41.95 g of what appeared to be a free flowing, green granular powder (Fig. 2). The material appeared to be homogeneous, with no visible indications of other materials or impurities. After initial analysis at ANSTO, a 10 g aliquot of this material was sent to LLNL.

Preliminary elemental analysis of the seized sample indicated that the rare earth element (REE) pattern was a close match to a previously analysed UOC sample sourced from Mary Kathleen. Therefore, a uranium ore and a UOC sample from Mary Kathleen uranium mine were also characterised using some of the described techniques. The UOC (which was black in colour) was believed to have been produced in 1977, while the ore sample was mined in late 1980.

## 3. Characterisation techniques

A summary of characterisation techniques used to examine the seized material is provided in Table 1. High resolution gamma-ray spectrometry (HRGS) measurements were conducted for initial ‘categorization’ of the seized sample (i.e. to provide insight into the nature of the material and identify risks to health and safety). HRGS was performed on the ‘as-received’ sample at ANSTO using a Canberra instrument with an HPGe detector. The acquisition period was 12 h and the resulting spectrum was analysed using the



**Fig. 2.** Green coloured radioactive material designated NSR-F-130509.

‘multi-group analysis for uranium (MGAU)’ software (version 3.0) to determine the approximate uranium isotopic abundance. LLNL performed gamma spectrometry using in-house HPGe gamma spectrometers. Spectra were acquired for 8 h for initial categorization and then 3 days for confirmation. All spectra were analysed using an in-house version of the GAMANAL software.

Physical examination of the samples was conducted using both optical and electron microscopy. At ANSTO, the microstructure of NSR-F-130509 was characterised using scanning electron microscopy (SEM, JEOL JSM 6300), while elemental composition was measured using energy dispersive X-ray spectroscopy (EDS, Noran Instruments Voyager Series IV X-ray microanalysis system). At LLNL, the seized powder and an exemplar of UOC produced at the Mary Kathleen uranium mine were analysed using an FEI Inspect F SEM/EDS. LLNL prepared the materials for SEM/EDS analysis by one of two methods: surface transfer of the as-received sample or ultrasonication in solvent to promote disaggregation of the material.

The phase composition of the samples was measured using X-ray diffraction (XRD). ANSTO used a Siemens Kristalloflex D500 spectrometer in conjunction with the X’Pert HighScore Pro™ package from Panalytical. LLNL used a Bruker D8 Discover MR XRD and Bruker software. X-ray fluorescence (XRF) was used to determine major elemental content (i.e., elements greater than approximately 0.05 wt.%) [11]. ANSTO analysed pressed powders

**Table 1**

Summary of characterisation techniques used to examine the seized uranium material (and UOC from Mary Kathleen (MK) Uranium mine).

Analytical technique	Measured parameter
High resolution gamma-ray spectrometry	Initial categorization, approximate uranium isotopic abundance, activation and fission products
Optical microscopy	Physical characterisation—particle size and morphology
Electron microscopy (SEM/EDS)	Particle size; microstructure and elemental composition
XRD	Phase composition
XRF	Elemental composition (high content impurities)
Vis/NIR reflectance spectroscopy	Chemical information with no sample preparation; colour, CH, OH and NH bonds
Solid-phase microextraction (SPME) and GC-MS	Volatile and semi-volatile organic compounds
Loss-on-heating	Moisture content (to 140 °C for 2 h) and content of thermally removed species (to 750 °C for 12 h)
MC-ICP-MS (plus chemical separation)	U assay and isotopic analysis
ICP-MS	Age determination and Sr and Pb isotopic analysis
Neutron activation analysis (NAA)	Elemental impurities (including REEs)
Ion chromatography	Elemental impurities in MK ore sample
Elemental analyser	Anions (F <sup>-</sup> , Cl <sup>-</sup> , Br <sup>-</sup> , NO <sub>3</sub> <sup>-</sup> , SO <sub>4</sub> <sup>2-</sup> , PO <sub>4</sub> <sup>3-</sup> )
	Stable isotopes of C, N, O and S

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