Identification of unknown nuclear fuel by impurities and physical parameters

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(Received July 25, 2000)

Analytical parameters sufficient for identification of nuclear material of unknown origin have been assayed for a number of examples of uranium dioxide fuel. These parameters comprise isotopic composition of uranium and the morphology features of a fuel pellet. However, in practice, it occurs that a sample of an industrial material corresponds frequently to the specification limits of several suppliers. In order to narrow further a range for potential sources, additional analytical parameters are necessary, such as fuel pellet impurity levels and surface roughness. In this study we utilise results from the following nuclear analytical techniques: thermal-ionization mass spectrometry, glow discharge mass spectrometry and profilometry. By way of example, we demonstrate how these results serve to differentiate the sources of confiscated, vagabond nuclear materials.

Introduction

During the last decade law enforcement agencies have been faced with a number of seizures of vagabond nuclear material. Although these materials have mainly involved low-enriched uranium, they still contravene non-proliferation regulations. Therefore, the state authorities where the material has been found must prosecute those involved. To provide necessary evidence for prosecution, identification of a given intercepted material and its intended use must be established.

In the context of such investigations, the following categories of information are required: (1) place of production, and (2) intended use of the material.

Existing nuclear analytical techniques have been used or modified to answer these requirements. The systematic application of such techniques is now commonly known as Nuclear Forensics.1,2

A logical approach to identifying an unknown object is to compare typical patterns with matching historical data. For purposes of nuclear fuel identification, databases have been implemented,3,4 containing characteristic information for a variety of fuel types. However, the scope of the parameters available for such databases depends not only on the material type and origin but the year of production because the list of characteristic parameters and parameters under control is changed periodically. In any event, the analyses conducted on unknown materials should replicate the parameters contained in the historical database, and the analytical techniques to reproduce them have to be established and should be updated regularly to benefit from modern developments.

Data recorded and used for quality control of material production are of special significance for identification. Although such data is almost always commercially sensitive and not published in open literature, they are mandatory and, in turn, require setting up of sophisticated techniques in the laboratories charged with the analytical measurements.

In order to identify the production plant and thus the last known legal owner of a seized nuclear material, the protocol discussed here follows a step-by-step diagnosis using advanced technologies of analytical practice. The procedure for each investigation is flexible rather than being bound to fixed analytical schemes. By comparing the “fingerprints” to the database parameters in each step, the number of possible sources of a seized material is narrowed; and further analysis steps are decided upon from previous observations. In addition, each analysis step has to comply with health-physics requirements of handling nuclear material and avoid any unnecessary destruction of a sample.

Scope of the study

This paper describes, for selected cases of seized low-enriched uranium fuel, the determination of parameters sufficient to identify the origin and intended use of the material.

In this study the isotopic enrichment, geometric dimensions and impurities were found to be the most useful characteristic parameters for identification. Uranium isotopic composition in a material directly points to intended use. Together with geometric dimensions (pellet height, diameter and possible hole diameter), it is specific for a reactor type and thus immediately narrows the range of the possible production method. Such information is also published in open literature.5 Thus, these parameters are well suited as a first step in the systematic identification of unknown nuclear material.2,6
It has been hypothesized that parameters like impurities and surface roughness of the material give further exploitable evidence for linking a given unknown sample to a particular production plant and fabrication process. Published variations between different production plants in specified maximum fuel pellet impurities are negligible; but, because different fabrication methods are used, differences in actual impurities are expected. Therefore, these differences have been proposed for the identification of unknown samples. However, the surface roughness of pellets is a parameter that has not been included in the Institute for Transuranium Elements (ITU) database until now.

Although the surface roughness of a pellet is mainly dependent on the grinding procedure used on the fuel pellet, differences in surface roughness between different pellets, polished with the same polishing medium, have been observed. Differences may be due to variations in density and grain size of the individual pellets. This may, in turn, be dependent on powder properties of the starting material and preparation technique of the uranium-oxide pellets. Variations among different batches from the same plant have also been observed. Thus, future comparisons of measured fuel pellet surface roughness with entries in the database might not only indicate the production plant but can also give information on the production batch. Unlike data regarding isotopic enrichment, there is little information on these parameters published. However, such data are available from archives of production plants.

Apart from additional contamination or damage prior to seizure, identification of seized nuclear material is complicated by the fact, that it may be “scrap”, i.e., material which does not comply with the quality-control requirements of a production plant. Hence, some of the determined parameters may lead to ambiguities or even may give contradictory information about a material’s origin.

**Experimental**

**Isotopic composition and geometric dimensions**

The first step to narrow the range of potential origins of a given sample is to determine its geometric dimensions and isotopic composition. The geometric dimensions of the uranium pellets are determined using a micrometer in a standard way. In order to achieve high accuracy, the isotopic measurements are performed with a Finnigan Mat 261 thermal ionization mass spectrometer (TIMS). This device is equipped with seven Faraday collectors, fixed suitably for uranium and plutonium isotopic-ratio measurements. The uranium isotopic composition is measured by means of a routine total evaporation analysis method described previously.10

**Impurities**

Glow discharge mass spectrometry (GDMS) does not require chemical handling of the solid samples and offers a simple analysis method to determine impurities of fuel pellets. Moreover, with this method the risks of the contamination of the samples are small.11,12

Impurities compiled in this work have been determined with high resolution GDMS, VG 9000. The instrument is in a glove box and consists of a direct current glow-discharge ion source coupled to a double-focusing mass spectrometer of reverse (Nier-Johnson) geometry. This provides high transmission (>75%) and sensitivity when operating at a high mass-resolving power (4000 with 10% valley definition). Ion detection is accomplished by a dual-detection system consisting of a Faraday cup and transverse-mounted Daly detectors. The glow discharge was supported by high-purity argon. Impurities of sample 10 were determined using a secondary-cathode technique.13 The discharge voltage during the measurement is typically 1.2 kV at a current of 1 mA. The pressure of the ion-source chamber is 3·10⁻⁴ bar. Under these conditions the sample surface is pre-sputtered for 45 minutes. Parallel measurement is performed on the opposite side of the pellet.

**Surface roughness**

To measure surface roughness, a Veeco Sloan Technology Dektak 8000 measuring system is used. It is equipped with a videocamera system, a 12.5 μm diamond stylus and is fully computer controlled. Arithmetic average roughness (CLA or Rₐ), Arithmetic-average deviation (AA), and minimum roughness of the surface are calculated (Rₐ,min) by the Dektak 8000 software.

The pellets are measured with an operating force of 30 mg on the stylus and a 30-second scan time. The samples are scanned over a length of 2000 μm. A scan resolution of 0.222 μm/sample is achieved with these parameters. Ten parallel scans are recorded within a band of 300 μm from three different sides of the cylinder surface of the pellet. A steel reference material, with a minimum and maximum roughness of 2.52 μm and 2.57 μm, respectively, is measured under the same conditions.

**Results and discussion**

The relevant data from the first analysis phase of this work are the isotopic compositions and geometrical dimensions of the fuel pellets as compiled in Table 1, together with citations of suggested reactor types.3,5