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The use of SEM-EDX for the identification of uranium compounds in swipe samples for nuclear safeguards

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ABSTRACT

Environmental swipe sampling for safeguards purposes has been used by International Atomic Energy Agency since 1997 and is a powerful tool to detect undeclared materials and activities. This work describes the use of SEM-EDX, as an initial screening technique, in real-life swipe samples for identifying and characterizing uranium particles. The swipe samples were collected from a conversion plant at IPEN/CNEN, Brazil. The relative error of atomic percentage found in two different particles on the swipe samples, in comparison with UF4 CEA standard, range from 1.1% to 1.2% for fluorine and 4.6% to 5.0% for uranium. The values demonstrate the viability of SEM-EDX technique, as a previous analysis and as a support for other techniques, to identify some chemical and morphological characteristics in uranium particles.

1. INTRODUCTION

The safeguards system applied by the International Atomic Energy Agency (IAEA) is an important element of the global nuclear non-proliferation regime.

Since 1996, swipe samples have been used by the IAEA as a routine component of safeguards inspections. Swipe samples can be one of the most powerful techniques for verifying the absence of undeclared nuclear activities in States under safeguards agreements(1-6).

Swipe sample analysis can be broadly subdivided into individual particles analysis and bulk analysis. The main techniques for individual particle analysis are Secondary Ions Mass Spectrometry (SIMS), Fission Track Thermal Ionization Mass Spectrometry (FT-TIMS), and Laser Ablation Inductively Coupled Plasma Mass Spectrometry (LA-ICP-MS). These can reveal important nuclear signatures such as isotopic ratios of nuclides, impurities and the age of nuclear compounds(7-12).

The Secondary Electron Microscope with Energy-Dispersive X-Ray analyzing system (SEM-EDX) can be particularly useful in the initial identification of uranium in swipe samples, and might be appropriate to identify and characterize uranium particles (13-15).

The aim of this work is to show how the SEM-EDX technique may be helpful as support for other techniques, identifying and characterizing uranium particles in a real-life swipe samples from a conversion plant of the nuclear fuel cycle at the Nuclear and Energy Research Institute – IPEN/CNEN, São Paulo, Brazil.

2. EXPERIMENTAL

Instrumentation, Materials and Methods

Morphological and elemental analysis of uranium particles analysis was carried out by Scanning Electron Microscope, SEM, (TM3000 TableTop, Hitachi), with 15kV acceleration capability, magnification range 15x to 30000x, resolution 30nm and with high-sensitive semiconductor Backscattered Electrons (BSE) detector.

Chemical characterization was performed by energy dispersive x-ray spectroscopy (EDX), which uses a Silicon-Drift Detector (SDD) of 30 mm² and has a resolution 135 eV, by a system QUANTAX 70. The acquisition time for EDX was 200 seconds.

The standard UF_4 CEA (*Commissariat a L'Énergie Atomique*) was used to obtain the reference spectrum by EDX.

Swipe sampling area:

IPEN/CNEN operates a MTR research reactor, IEA-R1, which uses uranium enriched up to 20% of ^{235}U as nuclear fuel. This material is produced at the Nuclear Fuel Center department (CCN, IPEN/CNEN)[(10)]. The swipe samples were collected in a conversion plant which converts UF_6 in an intermediate compound, UF_4, necessary to produce the U_3Si_2-Al alloy used as a nuclear fuel in IEA-R1.The particles present in the swipe samples were recovery by using a carbon adhesive tape and fixed on aluminum supports that fit into the SEM.

3. RESULTS AND DISCUSSION

EDX analysis was performed using five independent replicates at the same sample point and the results are presented as an average of the replicates. Only uranium and fluorine were selected for the data acquisition.

Initially the EDX analyses were performed on the UF_4 CEA standard. The atomic % values founded were: 19.6 % for U and 80.4 % for F. These values are in good agreement with the expected chemical composition of UF_4. The EDX spectrum and morphological characteristics are show in Figure 1.

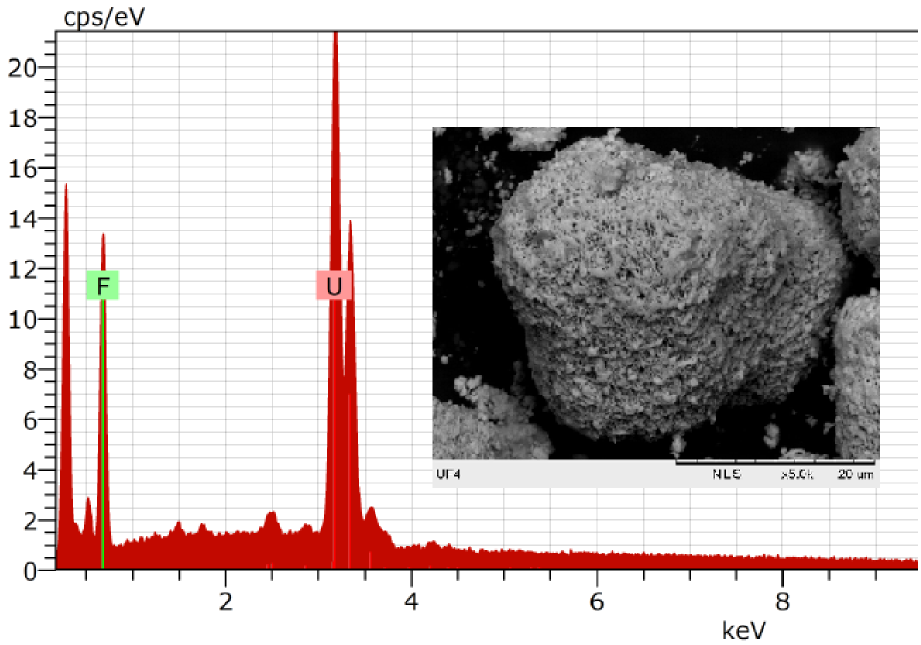

Figure 1: EDX spectrum for UF_4 CEA standard and its morphological characteristics.

Then, the EDX analyses were carried out on the real-life swipe samples and the results are shown in Table 1.

Table 1 – Results obtained by EDX analysis for two different particles and the standard UF_4 CEA.

	Sample				
	UF_4 CEA	**Particle 1**	**Relative error %**	**Particle 2**	**Relative error %**
Atom % U	19.6	18.7	4.6	20.6	5.0
Atom % F	80.4	81.3	1.1	79.4	1.2

OBS: The relative error is relative to the values obtained for UF_4 CEA standard.

It is important to notice that the relative error of atomic % found for the particles on the real-life swipe samples, in comparison with UF4 CEA standard, range from 1.1% to 1.2% for fluorine and 4.6% to 5.0% for uranium. These errors are in good agreement with the error of the technique.

Figure 2 shows the morphological characteristics, obtained by SEM, of two different particles found in the collected sample and its similarity with the UF_4 standard (Figure 1).

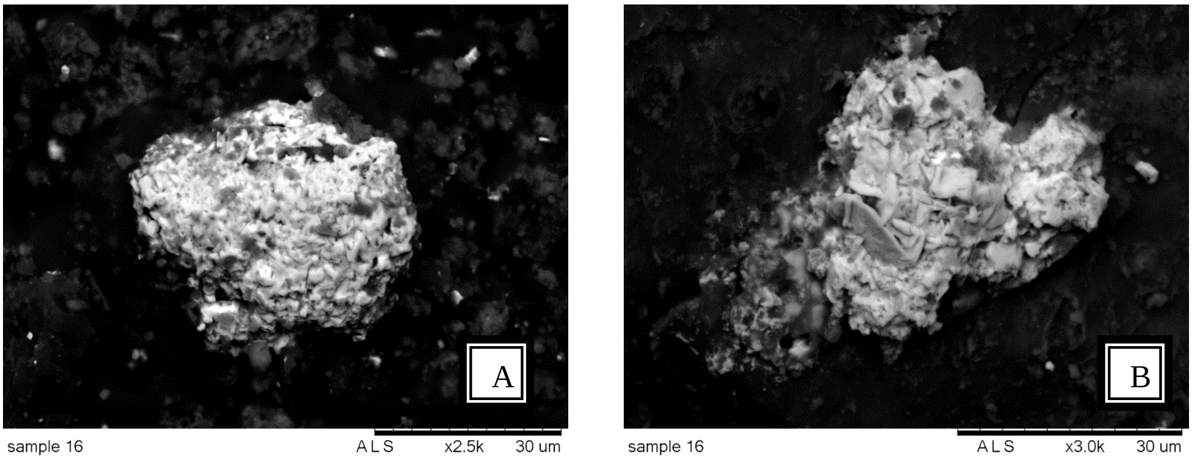

Figure 2: Image of two particles found on the real-life swipe sample collected at conversion facility.

The morphological characteristics and the results of atomic % suggest that the particles A and B are UF_4 compounds.

Figure 3 shows the EDX spectrum of particle A found on the swipe sample.

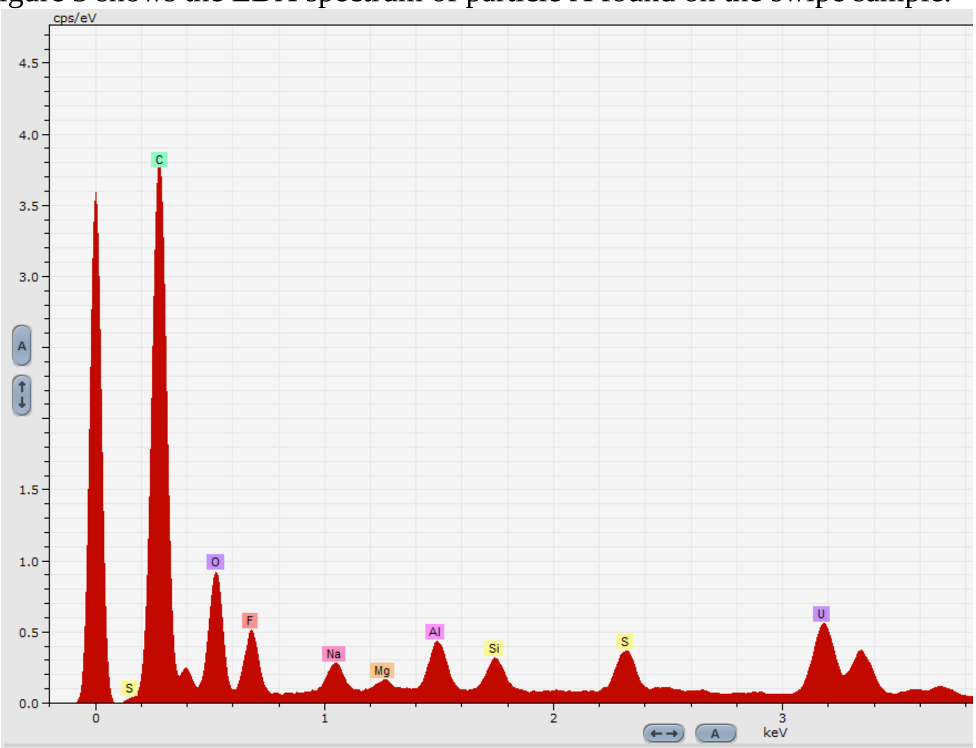

Figure 3: The EDX spectrum of particle A found on the swipe sample.

As it can be observed in the Figure 3, the EDX spectrum of a real-life particle found in a swipe sample is complex, being composed of several elements. Such complex spectrums composed of several elements are inherent in environmental sampling. In this case, Aluminum is possibly derived from the support, silicon from the detector, and carbon and oxygen from the atmosphere of the microscope chamber.

4. CONCLUSIONS

Annually circa of one thousand analysis of swipe samples are performed for safeguards purposes. As a consequence new methodologies and techniques are always been tested not only to face this analytical demand as well as to improve the quality of the results

Analytical techniques that can reveal nuclear information in swipe samples are important in the safeguards field and various techniques have been developed to obtain this information, known as signatures.

In this context, the use of SEM-EDX as a previous analysis can be very helpful to identify some chemical information and morphological characteristics in uranium particles.

This study demonstrated the capacity of the SEM-EDX technique to identify, in a real-life swipe samples, the main constituents of UF4 as well as its morphological characteristics.

Thus the SEM-EDX can be useful technique as an initial screening analysis and as supporting for other more precise techniques.

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