Analysis of UOC for nuclear forensics using Scanning Electron Microscope

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Abstract
Nuclear forensics involves the analysis of nuclear material for possible provenance determination using various analytical tools that are available for such analysis. In this study, Scanning Electron Microscopy (SEM) combined with Electron Dispersion Spectrometer (EDS), were used to determine the signatures of uranium ore concentrates (UOCs) samples for nuclear forensic applications. SEM and SEM / EDS provided substantial information on the UOC's morphology and elemental composition. Distinct qualitative and quantitative difference are present for the different UOC's. The UOC's surface consists of agglomeration made up of homogenous spherical particles, irregular shaped particles and plate like bulky particles. Average particle size ranged between 0.1 – 0.2 µm. EDS analysis of all the samples showed they contained a consistent 70 weight % of uranium and a stoichiometric formula closest to the molecule of UO_4. This technique can thus be used to distinguishing and fingerprinting UOC's originating from different mine.

1. Introduction

Nuclear forensics is a new discipline of forensic science. It is defined as *the examination of nuclear or other radioactive material, or of evidence that is contaminated with radionuclides, in the context of legal proceedings under international or national law related to nuclear security* [1]. Nuclear forensics provides material signature such as isotopic abundances, elemental concentrations, physical and chemical forms, morphology and physical dimensions that may be used to link a material, either nuclear or other radioactive (non-nuclear, such as those used for medical imaging), to individuals, locations, or processes, date of production and on the intended use [2, 3].

The Uranium Ore concentrate (UOC), commonly known as yellow cake is the main component in the uranium fuel cycle production. It is produced by various process which involves crushing, grinding and leaching the uranium ores or recovered as a by-product of other products, such as copper or phosphoric acid. Several cases involving theft of UOC's are recorded in the IAEA incident trafficking database [4]. Morphological signature is a comparatively new topic in nuclear forensics and refers to the size, crystalline structure and shape of particles. It is complementary signature to isotopic and elemental compositions [5]. The particle shape parameter is one of the most useful morphological characteristics for material differentiation [6].

The focus for much of the development and success of nuclear forensic investigation is to provide rapid capability for the characterisation of materials in scenarios where a bulk quantity has been

discovered or seized. A Scanning Electron Microscopy (SEM) equipped with Electron Dispersive Spectrometry (EDS) has the capability to provide one of the most rapid and reliable microscopy- based direct analytical techniques for measuring particle size, morphology and composition, however it requires accurate sample preparation [7]. EDS can also provide rapid qualitative analysis of elemental composition with a sampling depth of 1–2 microns, whilst x-rays might also be used to represent maps or line profiles, showing the elemental distribution in a sample surface [8].

SEM has been successfully applied in nuclear forensics to investigate the shape, appearance and particle size of various nuclear material from the nuclear fuel cycle [6, 9, 10]. Morphology of uranium pellets intended for the graphite moderate reactor was identified [11], high enrich uranium power origin was identified [12] and at the Munich airport, a sample consisting in a mixture of uranium and plutonium analysed by SEM revealed different grain sizes, leading to the conclusion that the materials were coming from different processes of formation [13]. The aim of the study was to resolve nuclear forensics signatures based on morphology and compositional analysis of the UOC's samples from Namibia and South Africa uranium mines.

2. Materials and Method

Three (3) UOC samples were obtained from Uranium mines in Namibia and South Africa. The powdered samples were mounted on an aluminium stub using double sided carbon tape. The mounted sample were coated with carbon to enhance conductivity and prevent charge build-up during SEM imaging. Secondary electron images of the samples were obtained in an FEI Quanta FEG 250 field emission gun SEM operating at an accelerating voltage of 15 kV. Oxford Energy Dispersive Spectrometer (EDS) operating with the Inca software was used for the compositional analysis. The quantitative measurement of the imaged morphological features was performed using Image J 1.52 a software and the distribution plotted in excel.

3. Results and Discussions

3.1 Morphology Characteristics

In an attempt to identify forensic signatures indicative of the origin of the material, qualitative as well as quantitative SEM image analysis of surface characteristics of the UOC's were carried out. The respective SEM images of the samples can be seen in Figures 1A – C below. Each row of images represents a sample taken at different levels of magnification X 13 000, 50 000 respectively. The UOC from mine A consists of massive agglomerates made up of homogenous spherical particles of size range 0.050 – 0.234 μm.

The UOC from mine B is a heterogeneous matrix consisting of spherical particles sandwiched between plates like bulky particles of size in range 0.041-0.799. The third UOC from Mine C shows the finest textures homogenous spherical particles in the range of 0.037-0.115. The morphological structures are summarised in Table 1.

The order of particle size is as follow: Mine A > Mine B > Mine C meaning Mine C has smallest particle sizes. The particles size distribution of the UOC's is shown in Figure 2 a – c. There is notable difference in the texture, size and shape of the particles from the different mines, this is mainly attributed to the different processing mechanism such as dissolution, extraction, ion exchange and precipitation [3].

Table 1: Summary of the morphology characteristics.

Mine ID	**Texture**	**Average particle size (nm)**
A	Homogenous spherical particles	92 ± 9.10
B	Heterogeneous platelets spherical particles	89 ± 8.45
C	Homogenous fine grained particles	69 ± 6.45

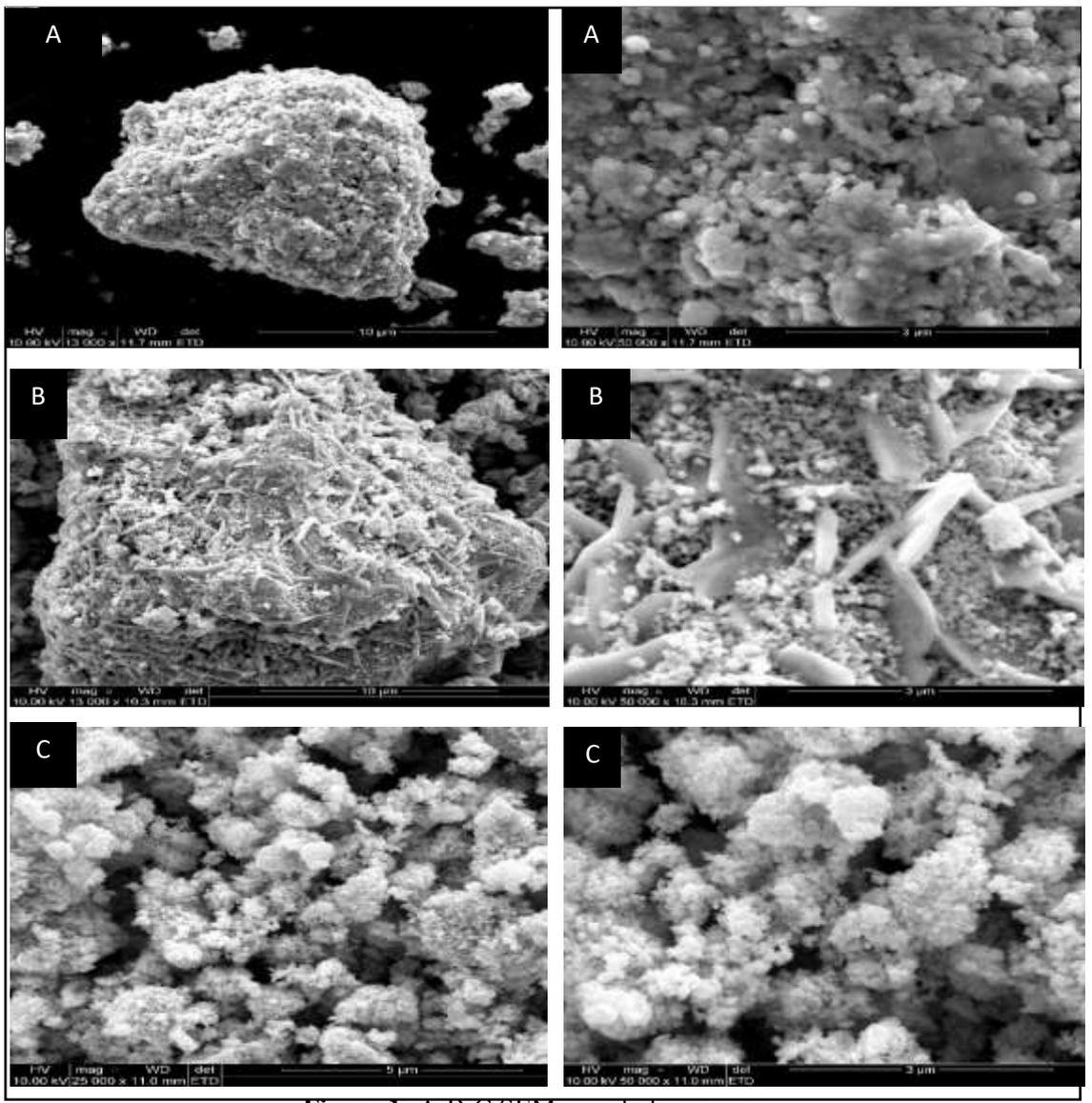

Figure 1: A-B-C SEM morphology images.

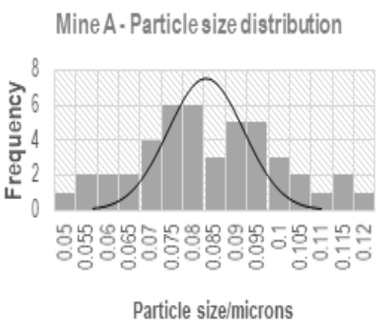

Figure 2.1: SEM images, histogram distributions and lognormal fits of mine A

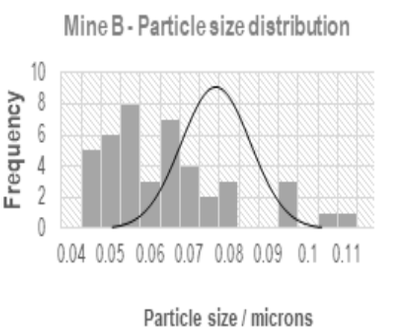

Figure 2.2: SEM images, histogram distributions and lognormal fits of mine B

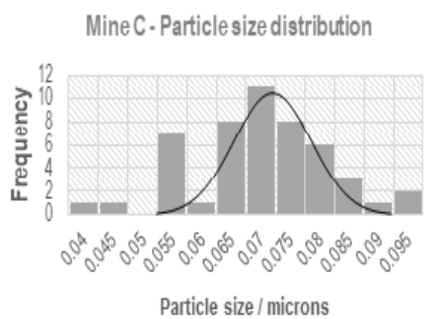

Figure 2.3: SEM images, histogram distributions and lognormal fits of mine C

There is difference in the particle size distributions as seen in Figure 2 above. Mine A particles shows almost symmetric distribution where most of the observed particles close to the mean 0.1 µm. Few particles are further away from the mean in both directions, Mine B distribution is skewed to the right most particle size is between 0.05and 0.09 µm. Mine C particles are left-skewed as most of the particle size clustered on the left side of the histogram. The differences in the distribution indicate that the mean particle size are different and distinct for each mine.

3.2 Elemental composition

Elemental composition was performed with the EDS and the obtained spectrums are depicted in Figures 3.1- 3.3 below. The major peaks observed were those of U and O, C with some minor peak of Na. The element carbon on the spectrums is a result of carbon coating prior to the analysis. Mine A contains additional element impurities Na which is due to the processing mechanism of the mine.

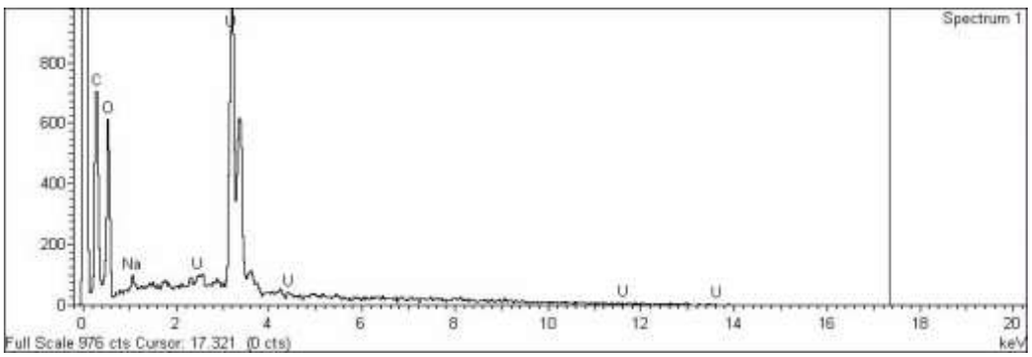

Figure 3.1: EDS spectrum of mine A, UOC.

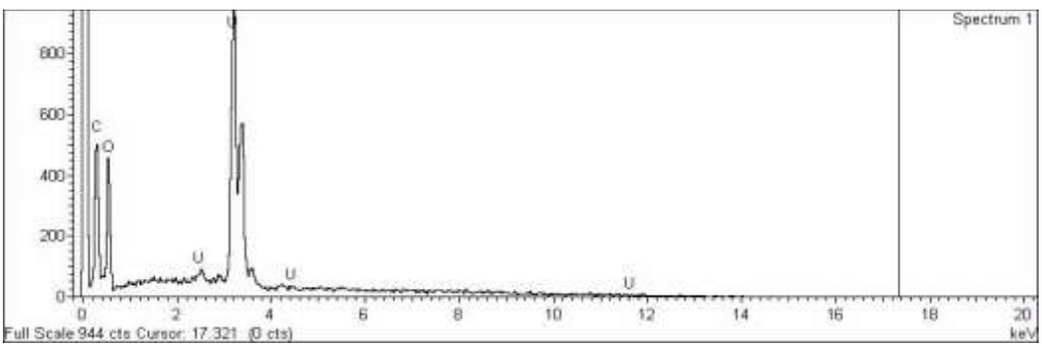

Figure 3.2: EDS spectrum of mine B, UOC.

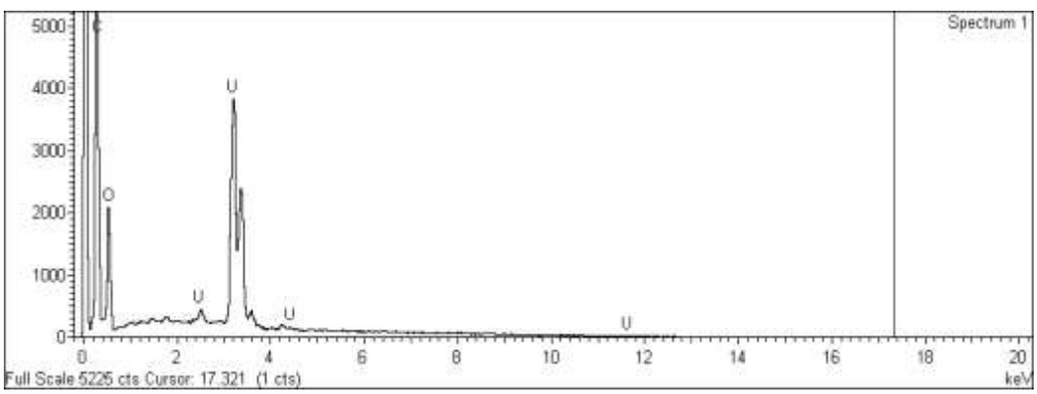

Figure 3.3: EDS spectrum of mine C, UOC.

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Results of the semi quantitative analysis of the UOC's using SEM / EDS are shown in Table 2.

Table 2: Elemental composition (weight (wt. %) and atomic (at %)).

	U		O		Na		Formulae
Mine ID	wt%	at%	wt%	at%	wt%	at%	Based on at% ratio
Mine A	73.84 ± 0.68	16.1 ± 0.50	25.32 ± 0.77	81.98 ± 0.77	0.85 ± 0.11	1.92 ± 0.28	UO_4
Mine B	77.64 ± 0.21	18.92 ± 0.18	22.36 ± 0.21	81.08 ± 0.18	-	-	UO_4
Mine C	76.56 ± 0.63	18.02 ± 0.53	23.44 ± 0.63	81.98 ± 0.53	-	-	UO_4

The results of wt % confirm that the samples of the mine are Uranium Oxide compounds constituting on over 70% of Uranium. The at% ratio analysed are closest to the molecules of UO_4 which constitute of 20%U and 80% O.

4. Conclusion

It has been demonstrated that SEM is a useful tool for possible signature of UOC origin assessment. The morphology analysis was able to distinguish particle's texture, shape and size of the UOCs indicative that they are of different mines. Impurities related to the production process were observed from the EDS results and the weight percentage indicates samples contains 70% of uranium. Future work would be to use a more quantitative approach to investigate if there are any crystalline difference between such samples.

Acknowledgement

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