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Application of the angle measure technique as image texture analysis method for the identification of uranium ore concentrate samples: New perspective in nuclear forensics

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ABSTRACT

The identification of interdicted nuclear or radioactive materials requires the application of dedicated techniques. In this work, a new approach for characterizing powder of uranium ore concentrates (UOCs) is presented. It is based on image texture analysis and multivariate data modelling. 26 different UOCs samples were evaluated applying the Angle Measure Technique (AMT) algorithm to extract textural features on samples images acquired at 250× and 1000× magnification by Scanning Electron Microscope (SEM). At both magnifications, this method proved effective to classify the different types of UOC powder based on the surface characteristics that depend on particle size, homogeneity, and graininess and are related to the composition and processes used in the production facilities. Using the outcome data from the application of the AMT algorithm, the total explained variance was higher than 90% with Principal Component Analysis (PCA), while partial least square discriminant analysis (PLS-DA) applied only on the 14 black colour UOCs powder samples, allowed their classification only on the basis of their surface texture features (sensitivity > 0.6; specificity > 0.6). This preliminary study shows that this method was able to distinguish samples with similar composition, but obtained from different facilities. The mean angle spectral data obtained by the image texture analysis using the AMT algorithm can be considered as a specific fingerprint or signature of UOCs and could be used for nuclear forensic investigation.

1. Introduction

Nuclear forensic science, or nuclear forensics, is a relatively young discipline which evolved as the need for analysing interdicted nuclear or radioactive material arose. In the event of a discovery of illicit trafficking of nuclear material, questions such as"what is the material?',"How it was produced?',"Where did it come from?' have to be answered as soon as possible. Typically, an investigation involves several measurable parameters, also termed as signatures or fingerprints; these can be understood as physical, chemical, isotopic characteristics of the nuclear materials that could collectively help to identify its origin [1–4].

Uranium ore concentrates (UOCs), or *"yellow cakes"*, are the precursors of nuclear fuel. These materials are often traded in large quantities and, therefore, diversions or thefts can happen. In fact, nuclear forensic investigations have been reported on this type of materials [5–7]. Depending on their processing history, these *"yellow cakes"* possess different compositions and it is therefore possible to differentiate or distinguish them from each other. Various measurable quantities associated with yellow cake compositions have been reported. These include analysis of major isotopes of uranium [8,9], and minor isotopes such as thorium [10], lead [11–13], strontium [12], sulphur [14] and neodymium [15]. Other minor constituents such as rare-earth elements [8,11,13,16–18], non-volatile organics [19] and anionic impurities [18,20,21] have also been analysed. Spectroscopic techniques such as infrared [21], near infrared reflectance [22,23], laser-induced breakdown [24] and Raman [25,26] of *"yellow cakes"* have also been applied.

A less explored signature is the correlation between the morphological characteristics of different UOCs, obtained using image

Abbreviations: AMT, Angle Measure Technique; C, Calibration; CV, cross-validation; I, intensity; IA, image analysis; PCA, Principal Component Analysis; PLS-DA, partial least square discriminant analysis; RMSE, root mean square error; UOCs, uranium ore concentrates

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analysis (IA) technique, and the processing or production history. IA represents a valid tool for the scientists that has been widely used in a wide range of fields such as medicine, agriculture, food, remote sensing, etc. [27–30] and that is currently gaining popularity in the nuclear field [31–33]. This technique allows the rapid definition of morphological and intensity characteristics of an object or of a complex structure in terms of size, shape, colour or surface properties (surface texture). Several qualitative and quantitative methods have already been applied to images of nuclear materials for different purposes [34–36]. Keegan et al. [7], for instance, described qualitatively the morphology of samples from two different UOCs; the physical examination was conducted using both optical and electron microscopy and the samples revealed a difference in the microstructure of smaller grains; in particular, in one case the smaller grains exhibited rougher, 'more textured' morphology, while in the other the grains were smoother showing 'less textured' morphology. This difference in microstructure, as explained by Keegan, might be evidence of a different processing history for the two materials. Recently, Ho et al. have demonstrated that IA can be used as an analytical technique to identify and classify several different UOCs samples on the basis of their morphology characteristics [37].

IA offers great advantages as it is highly accurate, reproducible, consistent and rapid [27,38,39]. In IA different levels of image processing can be used for feature extraction and pattern recognition. Starting with the simplest operations (low level of processing), it may include qualitative analysis by visual inspection, intermediate level processing or complex operations (high level processing) involving multivariate approach in the image data elaboration (see Fig. 1 in supplementary materials).

Among the different methods, image texture analysis is gaining interest with scientists and different image texture analysis methods have already been used for powder characterization [38,40]. This is because in an image where the entire field of view of the powder sample is acquired these methods provide information not only on the individual particles but also on the bulk powder environment (i.e. how the particles are arranged together). Traditional IA for powder characterization is focused on single particle analysis instead and this approach requires time consuming steps for sample preparation, e.g. for dispersion of the powder before microscopic imaging [7,40].

The aim of this work was to evaluate the ability of the image texture analysis to identify different powders of UOCs from their surface appearance (surface topography, surface texture). The Angle Measure Technique (AMT) [41,42] algorithm was applied, as part of the image texture analysis methods, to 26 UOCs from 26 separate facilities located in various countries. The surface appearance of the powder samples of the UOCs is influenced by the composition and the process technology [7], which give rise to different chemical-physical properties such as grain size, shape, homogeneity, etc., and to the capacity to create agglomerates of different dimension. All these features were evaluated by the assessment of the Image Texture.

2. Materials and methods

2.1. Powder samples

Table 1 illustrates the main parameters associated with the UOCs samples used in the present work. A total of 26 UOCs samples were selected in function of their composition and the technology used for the production and, furthermore, 14 samples (labelled with "*" in the table) were also characterized by a black colouring of their powders. The samples were prepared in the following way: a few hundred milligrams of powder were placed in a clean petri-dish; a layer of adhesive carbon (sticky on both sides) was applied onto of a 12.5 mm SEM sample disk holder; the holder coated by the double sticky tape was put in contact with the sample powder. The holder was gently pressed onto the sample until sufficient powder adhered to the tape. The samples were subsequently coated with 10 nm gold to ensure conductivity in view of the Scanning Electron Microscopy examination.

Table 1
Information regarding 26 UOC samples investigated in this study.

Sample	Facility[a] and abbreviation	Country	Composition[b] and abbreviation	Known processes
1	South Alligator **(ALL)**	Australia	Uranyl hydroxide **(UH)**	U ppt with MgO
2	Atlas **(Atl)**[*]	USA	Mixed oxide **(Mix)**	U ppt with H_2O_2
3	Chevron Hill **(ChH)**	USA	Mixed (ADU+oxide) **(Mix)**	U ppt with NH_3
4	Congo **(Con)**	Belgian	Uranyl hydroxide **(UH)**	Not known/available
5	South Dakota **(Dak)**	USA	Mixed (oxide+hydroxide) **(Mix)**	U ppt with MgO
6	ESI **(ESI)**	Canada	ADU **(ADU)**	U ppt with NH_3
7	ESI **(ES2)**	USA	ADU **(ADU)**	Not known/available
8	El Dorado **(ElD)**	Canada	ADU **(ADU)**	U ppt with NaOH
9	Federal American Partners **(FAP)**[*]	USA	U_3O_8 **(U3O8)**	U ppt with NH_3
10	Heng Yang **(HeY)**[*]	China	UO_2 (+ U_3O_8) **(UO2)**	Not known/available
11	Key Lake **(KeL)**[*]	Canada	U_3O_8 **(U3O8)**	U ppt with NH_3, calcined at 750 °C
12	Mary Kathleen **(MaK)**[*]	Australia	U_3O_8 **(U3O8)**	U ppt with NH_3
13	Mulberry **(Mul)**	USA	ADU **(ADU)**	Not known/available
14	Nufcor **(Nuf)**[*]	South Africa	U_3O_8 **(U3O8)**	Not known/available
15	Olympic Dam **(OlD)**[*]	Australia	U_3O_8 **(U3O8)**	U ppt with NH_3, ADU dried and calcined at 750 °C
16	Palabora **(Pal)**[*]	South Africa	U_3O_8 **(U3O8)**	U ppt with NH_3
17	Petromics **(Pet)**[*]	USA	Mixed oxide **(Mix)**	Not known/available
18	Queensland **(Que)**[*]	Australia	U_3O_8 **(U3O8)**	U ppt with NH_3
19	Rabbit Lake **(RaL)**	Canada	Uranyl peroxide **(UP)**	U ppt with magnesia and H_2O_2
20	Ranstad **(Ran)**	Sweden	Sodium diuranate **(SDU)**	U ppt with NaOH
21	Rössing **(Rss)**[*]	Namibia	U_3O_8 **(U3O8)**	U ppt with NH_3
22	South Uranium Plant **(SUP)**[*]	South Africa	U_3O_8 **(U3O8)**	U ppt with NH_3, calcined at 490 °C for 6 h
23	Somair **(Som)**	Niger	Sodium diuranate **(SDU)**	U ppt with NaOH
24	Spisak Black **(SpB)**[*]	Yugoslavia	Uranyl hydroxide **(UH)**	Not known/available
25	Techsnab **(Tec)**[*]	Russia	Mixed (U_3O_8+ADU) **(Mix)**	Not known/available
26	United Nuclear **(UnN)**	USA	ADU **(ADU)**	Not known/available

[a] 14 samples that are physically black in colour are denoted with an *.
[b] The compositions are assumed from their corresponding infrared and/or Raman spectra.