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ELEMENTAL COMPOSITION OF AIRBORNE PARTICULATES

IN URANIUM MINING AND MILLING OPERATIONS*

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ABSTRACT

Airborne particulates were collected through filters in occupational areas of the uranium mining and milling complex located in Pocos de Caldas, Brazil. The filters were analyzed by microPIXE (particle induced x-ray emission) combined with Rutherford Backscattering (RBS) of the incident protons. The results are discussed in the paper.

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1. Introduction

There is increasing interest in the elemental composition of particulates present in uranium mining and milling operations because lung diseases, including lung cancer, might be due to toxic elements present in the respirable fraction of such particulates. Routine air monitoring is underway in occupational areas of the Brazilian uranium mine and mill industrial complex in Pocos de Caldas as part of a radiation protection program. This program includes the sampling of airborne particulates in the open pit mine, and in the precipitation, drying and packaging areas of the uranium mill, as described elsewhere (1). Two types of sampling are routinely used: (i) conventional air filtration for determining airborne uranium concentrations, and (ii) collection of suspended dust using cascade impactors for the characterization of particle size distribution of uranium in aerosols to estimate the activity median aerodynamic diameter (AMAD) for future dose calculations.

This paper deals with a procedure associated with the first type of sampling and analysis. Testing uniform distribution of elements on the filter surface is important because collected filters are usually divided in two or more parts for independent analysis by radiochemical methods. The PIXE technique (2) was used in combination with RBS of protons to test the adequacy of this common practice in the nuclear industry.

2. Experimental

Sampling procedures to collect airborne dust particles in occupational areas of uranium mines and mills have been described elsewhere (1). The filter samples are usually divided in two parts for analysis of their uranium content by two independent radiochemical techniques. A test for the uniform

distribution of uranium and other elements was accomplished by microPIXE and RBS analyses of a series of irradiated sites on a filter exposed to airborne dust in a uranium mine, and a filter exposed in a uranium mill. For the filter exposed to airborne dust from the uranium mine, 24 irradiated sites 20 to 200 μm were analyzed, covering a total distance of about 1 mm on the filter. The filter exposed to airborne dust in the drying area of the uranium mill was irradiated in 6 sites 200 μm apart.

A 20 X 200 μm microbeam is obtained by focusing the proton beam from the 3.5 MV Van de Graaff accelerator at Brookhaven National Laboratory (BNL) with an electrostatic quadrupole triplet. The target holder can move in the horizontal and vertical directions with steps as small as 5 μm . RBS protons are detected by an annular particle detector, and characteristic x-rays emitted by the irradiated portion of the sample are simultaneously detected by a lithium-drifted silicon (Si-Li) detector. A schematic representation of the experimental arrangement for the microPIXE-RBS combination at BNL was presented elsewhere with a detailed description of the data acquisition and analyzing facilities (3). A discussion of calibration procedures using the microPIXE-RBS combination was also published elsewhere (3).

The calibration for the filter analyses was made by irradiating the National Bureau of Standards (NBS) SRM-610. The x-ray spectrum of the NBS/SRM-610 is shown in Figure 1a with peak identification. Experimental data on the number of counts per backscattered proton per ppm were obtained from the irradiation of the NBS/SRM-610 for 10 elements, and a least square fit curve on exponential functions is represented with the experimental data in Figure 1b. The concentrations of detected elements in the irradiated volume of the filter were obtained by dividing the net number of x-rays under the characteristic x-ray peak of the element, normalized by the number of

backscattered protons, by the calibration factor in $\frac{\text{counts}}{\text{protons ppm}}$ for the particular element, as given in Figure 1b.

3. Results and discussion

Figure 2a shows the x-ray spectrum of irradiated site number 1 in the filter exposed to airborne dust in a uranium mine, while Figure 2b shows the x-ray spectrum of irradiated site number 13 in the same filter. Despite the similarity of these two spectra, noticeable differences in some peaks can be observed.

Table 1 shows the local concentrations of some of the elements detected in these filters and in filters exposed to airborne dust in a yellow cake drying area of a uranium mill. Some of the elements detected in the filters were also found in lung tissues of former uranium miners and millers collected at autopsy (4). The ratios of the elemental concentrations (site 1/site 13) of the uranium mine filter presents variations from 0.01 for Zr up to 1.12 for Mn. Since neither of these elements appear in the spectrum of a nonexposed filter shown in Figure 2c, one can expect that, for example, the low ratio (0.01) for Zr is due to the fact that a much larger amount of this element was deposited in site 13 than in site 1. If this nonuniform distribution also applies for U, the practice of cutting the filter in half to be analyzed by independent methods should not be continued, since it is likely that this element is not uniformly distributed throughout the filter surface. However, ratios of elements such as K, Ca, Ti, Fe, Ni, Zn, and Sr are difficult to interpret, since these elements appear distinctly in the x-ray spectrum of the nonexposed filter shown in Figure 2c. Nonuniform distribution of elements in the filter itself may cause some of the ratios shown in Table 1.

Figure 2d shows the x-ray spectrum of irradiated site 2 of a filter exposed to airborne dust in the drying area of the uranium mill. This spectrum can be considered typical for that area, because most ratios of elemental concentrations determined for different sites of the same filter are near unity, meaning essentially that the elements are part of the airborne dust in the area. However, it is interesting to note that the ratio for uranium in the two sites of the yellow cake filter is only about 0.5, meaning that some airborne dust particulates are richer in uranium than others. X-ray spectra of freshly-produced yellow cake, not shown in this paper, indicate that PIXE analysis can also be helpful in identifying undesirable impurities.

4. Conclusions

1. The common practice in some monitoring operations of the uranium industry of cutting filter samples in half to be analyzed by independent methods for their uranium content should be abandoned because nonuniform distribution of elements can give misleading results.
2. The combination of microPIXE and RBS techniques is a helpful tool for examination of elemental concentrations of airborne dust particulates collected in filters.
3. Filters with more uniformly distributed elemental composition should be used for reliable elemental concentration determinations.
4. Undesirable elements and impurities in yellow cake can be easily detected by using the techniques described in this paper.
5. Particle size distribution and elemental composition of suspended dust particulates commonly found in occupational areas of the uranium industry can usually be determined by using PIXE, microPIXE and RBS techniques.

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Table 1. Local concentrations and their ratios of irradiated sites of filters exposed to airborne dust in occupational areas of a uranium mine and a uranium mill (drying area) as determined by combining microPIXE with Rutherford Backscattering of protons.

Element	Z	Local Concentrations ($\times 10^{-3}$ ppm)				Ratio	
		Uranium Mine		Uranium Mill		Uranium Mine	Uranium Mill
		Site 1	Site 13	Site 5	Site 2	Site 1/Site 13	Site 5/Site 2
K	19	13.04 $\pm$ 0.04*	49.5 $\pm$ 0.1	22.87 $\pm$ 0.07	23.58 $\pm$ 0.09	0.263 $\pm$ 0.001**	0.970 $\pm$ 0.005
Ca	20	0.74 $\pm$ 0.01	2.51 $\pm$ 0.03	35.25 $\pm$ 0.05	31.93 $\pm$ 0.04	0.295 $\pm$ 0.005	1.104 $\pm$ 0.002
Tl	22	0.594 $\pm$ 0.003	0.645 $\pm$ 0.005	8.58 $\pm$ 0.01	7.77 $\pm$ 0.01	0.921 $\pm$ 0.009	1.104 $\pm$ 0.002
V	23	-----	-----	4.32 $\pm$ 0.01	4.07 $\pm$ 0.01	-----	1.061 $\pm$ 0.004
Cr	24	-----	-----	0.268 $\pm$ 0.004	0.206 $\pm$ 0.004	-----	1.30 $\pm$ 0.03
Mn	25	1.613 $\pm$ 0.005	1.440 $\pm$ 0.006	-----	-----	1.120 $\pm$ 0.006	-----
Fe	26	3.053 $\pm$ 0.005	11.30 $\pm$ 0.01	0.308 $\pm$ 0.002	0.329 $\pm$ 0.002	0.2702 $\pm$ 0.0005	0.936 $\pm$ 0.008
Ni	28	-----	-----	0.032 $\pm$ 0.001	0.040 $\pm$ 0.001	-----	0.80 $\pm$ 0.04
Cu	29	0.0112 $\pm$ 0.0006	0.055 $\pm$ 0.002	-----	-----	0.20 $\pm$ 0.01	-----
Zn	30	0.059 $\pm$ 0.002	0.281 $\pm$ 0.004	27.83 $\pm$ 0.03	27.42 $\pm$ 0.02	0.210 $\pm$ 0.008	1.015 $\pm$ 0.001
Sr	38	-----	-----	0.004 $\pm$ 0.007	-----	-----	-----
Zr	40	0.010 $\pm$ 0.007	0.98 $\pm$ 0.02	-----	-----	0.010 $\pm$ 0.009	-----
Pb	82	0.008 $\pm$ 0.002	0.069 $\pm$ 0.003	0.019 $\pm$ 0.002	< 0.010	0.12 $\pm$ 0.03	-----
U	92	-----	< 0.005	5.05 $\pm$ 0.05	10.04 $\pm$ 0.06	-----	0.503 $\pm$ 0.006

* Error due to counting statistics only.

** Propagated error from local concentrations.

FIGURE CAPTIONS

Figure 1. a) X-ray spectrum of NBS/SRM-610. b) Sensitivity curve fitting experimental data obtained by irradiating the NBS/SRM-610 with 2.5 MeV protons.

Figure 2. a) X-ray spectrum of irradiated site #1 of a filter exposed to airborne dust in a uranium mine. b) X-ray spectrum of irradiated site #13 of the same filter referred to in Fig. 2a. c) X-ray spectrum of a nonexposed filter. d) X-ray spectrum of irradiated site #2 on a filter exposed to airborne dust in a yellow cake drying area.

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