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Department of Natural Resources
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A Rapid Radiometric Analysis For Equivalent Uranium

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A B S T R A C T

The laboratory of the Alaska State Division of Mines and Geology has inaugurated a program of routinely checking for radioactivity of virtually all samples submitted by the public for identification or analytical work. This report describes a routine radioactivity test for "significant" radioactivity and a semiquantitative equivalent uranium* analysis. The routine survey analysis for "significant" radioactivity is made directly on crushed and ground samples in their paper sample bags. This study shows that a five second test in our lead shielded, end-window, laboratory geiger counter has a minimum detection limit of 0.05% equivalent uranium. A semiquantitative radiometric analysis which has an analytical range between 0.01% and 2.0% equivalent uranium is also described. The semiquantitative analysis uses about three grams of powdered rock sample pressed at 12 tons/sq. inch gage pressure into a 1¼" pellet. The sample's radioactivity is determined directly from this pellet. The radioactivity from the sample is compared to that from analyzed standards. The precision of the analysis was found to be 10% to 15% of the value when taking 100 to 200 total counts, which requires about one minute counting time on a 0.1% uranium ore sample.

I N T R O D U C T I O N

The laboratory of the Alaska State Division of Mines and Geology handles hundreds of samples per year by public request for identification and analytical work. Because of the simplicity of the radioactivity test, the nondescript appearance of some uranium minerals, and because very few prospectors routinely make the test, it is thought to be justified for the laboratory to test for radioactivity essentially all submitted samples.

The results of the semiquantitative analysis described are reported in terms of equivalent uranium (eU). The analysis is a measure of the total radioactivity from the sample as detected by our instrument compared to the total radioactivity detected from a sample of known uranium content. However, the bulk of the radioactivity from an uranium ore sample is from uranium daughter products rather than from the uranium itself. The eU analysis presupposes the uranium is in equilibrium with its daughter products. The eU analysis may be defined as the amount of uranium, in equilibrium with its daughter elements, in an unknown sample that gives the same counting rate as that from a known sample (Johnson, 1954, p 238). Thus, the radiometric analysis is not specifically an analysis for uranium, but is an analysis for radioactivity. It is expressed in terms of the amount of uranium which may be present to give rise to that amount of radioactivity. Samples that are found to contain significant radioactivity can be quantitatively analyzed by X-ray spectrography specifically for uranium.

*The term "equivalent uranium" implies that this is a broad spectrum radioactivity test and is not specific for uranium.

In making a radiometric analysis it is important to realize that the process of nuclear fission is a random one. In any random process the number of events that occur in unit time is not necessarily the same as the number of events that occur in the next unit time. The fractional difference between a single measurement and the average of a large number of measurements is the statistical error. The probable error between two measurements can be stated as the standard deviation, σ, which is proportional to the $\sqrt{N}$ where N is the number of events, counts in our case. σ may be stated as relative or percentage error, termed the coefficient of variation (γ), where $\gamma = \frac{\sigma}{\text{average value}} \times 100$. In random processes the precision (reproducibility) of the measurement obtained is related only to the number of events recorded, and not in any way to the time required to monitor the events. Therefore, to obtain equal precision of all measurements the constant should be "fixed counts", selected to achieve the desired precision, and the measured variable should be the amount of time required to accumulate the counts. Furthermore, the background, which is composed of cosmic and any other extraneous radiation sources and instrument electronic noise, is a separate variable and should be established as a separate counting rate. Sometimes these counting rates are very low and require long counting times for high precision. To indicate the magnitude of these counting times the following table shows the number of counts needed to obtain the stated relative precision:

Number of Counts Accumulated	Relative Error %
10	32
100	10
1,000	3
10,000	1

Since background and near-background counting rates may be on the order of less than ten counts/min., high precision at these rates require considerable time to accumulate the required counts. Efficiency in counting strategy has been described by Evans (1954, p 22-24). Review of his paper is useful.

E Q U I P M E N T

The principal instrument used in this radiometric analysis is a Model RC1A lead-shielded, end-window geiger counter manufactured by the Nucleonic Corporation of America, Brooklyn, New York. The Model RC1A counter includes a count accumulating device. This provides for establishing a counting rate by dividing the accumulated number of counts by the time required to accumulate the counts. Cosmic and other extraneous sources of radiation are small because of the heavy lead shield around the counter.

The rock sample is pulverized to principally minus 200 mesh by any suitable grinder. We use a Braun pulverizer after reducing the sample to minus ¼" with a jaw crusher.

The amount of radioactivity measured from a sample depends on several factors which include the volume of sample, the sample-to-counter geometry, and the concentration of the radioactive elements. A very rapid, yet adequate, method of controlling these factors for a semiquantitative analysis is to prepare the sample as though for an X-ray spectrographic analysis. The powdered sample is placed in a 1¼" diameter thin-walled aluminum cup (Spec - cap*). The cup and contained sample are both pressed into a durable pellet using 12 tons/sq. in. pressure in a Carver Model 20000-82 hydraulic press. Samples and standards are both similarly prepared so the counting rates of the samples can be compared to the counting rates of the analyzed standards. This pressed pellet method standardizes the sample-to-geiger-tube geometry and reasonably well reproduces sample volumes. For simple survey work to identify samples with "significant" radioactivity powdered sample in paper bags of approximate constant volume provides adequate control.

An important characteristic of a geiger-miller tube is the impressed voltage. The detector sensitivity is dependent on the voltage, but is not linearly dependent. The curve of counter sensitivity versus increasing voltage forms a steep slope, followed by a plateau, and then another steep slope. For good operating stability it is necessary to use a voltage in the mid-range of the plateau portion of the curve. In this voltage range only small changes in sensitivity result from normal voltage fluctuation. At present the optimum operating voltage for our counter is 1130V. This voltage can vary as vacuum tubes and other electronic components age, and the voltage must be checked from time to time.

E X P E R I M E N T A N D R E S U L T S

ROUTINE SURVEY ANALYSIS

Our need is for a survey analysis to detect significant quantities of radioactivity in a minimum of testing time. Standard samples of powdered ore of known uranium content that are similar in volume to normal laboratory samples were used. The standard samples are simply powdered ore in paper sample bags each weighing approximately 200 grams and are similar to the powdered routine laboratory sample. These standard samples ranged in U values from 0.005% to 2.0%. The standards were successively placed in the counting chamber and counted for one minute. Background counting level was established with no sample in the counting chamber. Each standard sample was run three times. The results are shown in table 1.

It is seen from the table that the 0.05% U sample which we accepted as the lower practical U level to be of interest for ore, gave a counting rate about eight times the background count. Using a minimum five second counting time, background should yield four to six counts.

*Spec Industries, Inc., Metuchen, N. J., 08841, Catalog No. 3619.

To test the routine five second count procedure background, 0.01% U, and 0.05% U samples were each counted ten times for five second intervals with the results shown in table 2. The standard deviation of the five second count at 0.05% U is 1.1 count, the mean is 4.7. With background being zero or one count we have 99% confidence of detecting 0.05% U above background with a five second count. The above data confirm the validity of the five second count.

TABLE 1

COUNTING RATE ON APPROXIMATELY 100 GRAMS
OF POWDERED ANALYZED STANDARDS

Sample	Counting Rate Counts/Min. 3 One Min. Runs	Comment
No Sample	6, 10, 8	Background
0.005%U	12, 13, 13	
0.01%U	17, 15, 18	
0.05%U	54, 58, 59	
0.1%U	97, 90, 89	
1.0%U	537, 539, 524	
2.0%U	519, 554, 504	Above 1% the counter was "choked"*

*"Choking" means that the closeness of arrival of radioactivity pulses was higher than the electronic curcuitry of the instrument could resolve and register as individual counts. If coincident arrival becomes sufficiently high the geiger tube can go into a state of continuous discharge and no counts are registered.

TABLE 2

TOTAL COUNTS OBTAINED IN 5 SECONDS ON
APPROXIMATELY 100 GRAMS OF 0.05% U

Background (No Sample)	0.01%U	0.05%U
1	2	3
1	2	5
0	1	5
0	2	4
1	1	6
1	1	4
1	2	7
2	2	4
1	2	4
1	2	5

The following procedure is recommended for the routine rapid radioactivity test:

1. Turn on geiger counter and allow 15 minutes to warm up.

2. With a 1% eU standard in the counting chamber confirm the proper counter plateau voltage. This will be between 1100 and 1200V; at this time 1130V is used.

3. Move "Count" switch to off position and press the "Reset" button to negate any partial counts in the counter circuitry.

4. Set count accumulator to zero.

5. Lift the "Count" switch to begin the count. Accumulate counts for five seconds and read the accumulated counts. If the accumulated count value is three or more set the sample aside for further testing, otherwise the sample can be reported as containing less than 0.05% equivalent uranium.

6. For those samples yielding three or more counts in five seconds a new count of one minute duration comparing to one minute counts on background and one minute counts on each of the 0.01%U and the 0.05%U should be made. If this test indicates radioactivity on the order of 0.05% eU or greater the sample should then be run by the semiquantitative radiometric procedure for eU.

SEMIQUANTITATIVE EQUIVALENT URANIUM ANALYSIS

Powdered samples of analyzed uranium ores were prepared by loosely filling thin walled Spec - Caps level full with standard sample. The cup and contained sample were than pressed into a thin 1¼" diameter pellet. The pressing was done by placing the cup and sample in a suitable die and pressing at 12 tons/sq. in. gage pressure in a hydraulic press.

The counting rate on each of the standards was obtained using one minute counting time. Table 3 shows these data.

To obtain a measure of precision five pellets were pressed from the 0.1%U sample. Each of the five pellets were counted three times for one minute intervals. These data are shown in table 4.

TABLE 3

COUNTING RATE DATA OBTAINED FOR THE SEMIQUANTITATIVE EQUIVALENT URANIUM ANALYSIS

Sample	Counting Rate, Counts/Min. (Three Runs)	Comment
No Sample	6, 10, 8	Background
0.005%U	9, 12, 11	
0.01%U	11, 12, 9	
0.05%U	21, 22, 18	
0.1%U	33, 28, 34	
1.0%U	227, 211, 228	
2.0%U	394, 395, 390	

TABLE 4

PRECISION STUDY DATA OF 0.1%
URANIUM SAMPLE

Pellet	Counting Rate, Counts/Min.			
	Run 1	Run 2	Run 3	Average
1	33	29	27	30
2	29	31	34	31
3	24	36	30	30
4	35	27	31	31
5	35	31	29	32
Average Counting Rate	31	31	30	

There is approximately equal variation among pellets and among separate runs on a single pellet which probably means the counting error is as great as the sampling error. The standard deviation on Run two, for example, is

$$= \frac{\sqrt{x^2}}{N-1} = \frac{45}{4} = 3 \text{ counts/min.}$$ (x = individual deviations from the mean). A measure of the analytical error can be made by subtracting the eight cpm (counts/min.) background from the 31 cpm for 0.1% U, which gives 23 cpm/ 0.1% U. The 3 cpm standard deviation is then calculated to equal 0.01% U. An analysis in the 0.1% U concentration range, as indicated from the above test, would yield a precision of 10% of the reported value. This is an acceptable semiquantitative analysis for our application. The precision could be improved by taking a larger number of counts. After confirming significant eU the sample can be analyzed by X-ray spectrography for a more precise and specific value.

The recommended procedure for the semiquantitative analysis is as follows:

1. Reduce the sample to principally minus 200 mesh.

2. Fill a Spec-Cap with loose powder to level full.

3. Press the cap and sample into a pellet at 12 tons/sq. in. gage pressure in a suitable die and hydraulic press.

4. Allow the counter to warm up and stabilize for 15 minutes.

5. Establish the geiger plateau voltage, probably 1100 to 1200V.

6. Determine the counting rates from the prepared standards by accumulating no fewer than 100 counts, preferably 200 to 500 counts.

7. Determine the counting rates on the unknown samples, accumulating total counts equal to those taken on the standards.

8. Compare the counting rates obtained on the unknowns to an analytical curve drawn from uranium concentration versus counting rate obtained on the standards.

9. It is advisable to analyze each unknown sample in duplicate. The value obtained is reported as "equivalent uranium" with an estimated precision of 10% of the value.

REFERENCES

Johnson, Donald Haskall, Radiometric Prospecting and Assaying; in Nuclear Geology, edited by H. Faul, John Wiley and Sons, N. Y., 1954, p 238.

Evans, Robby D., Instruments and Techniques of Detection and Measurement; in Nuclear Geology, edited by H. Faul, John Wiley and Sons, N. Y., 1954, p 22-25.