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Progress Report

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**Oklo—Natural Fission Reactor Program**

**January 1—March 31, 1979**

University of California



**LOS ALAMOS SCIENTIFIC LABORATORY**

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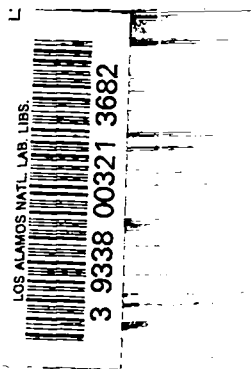
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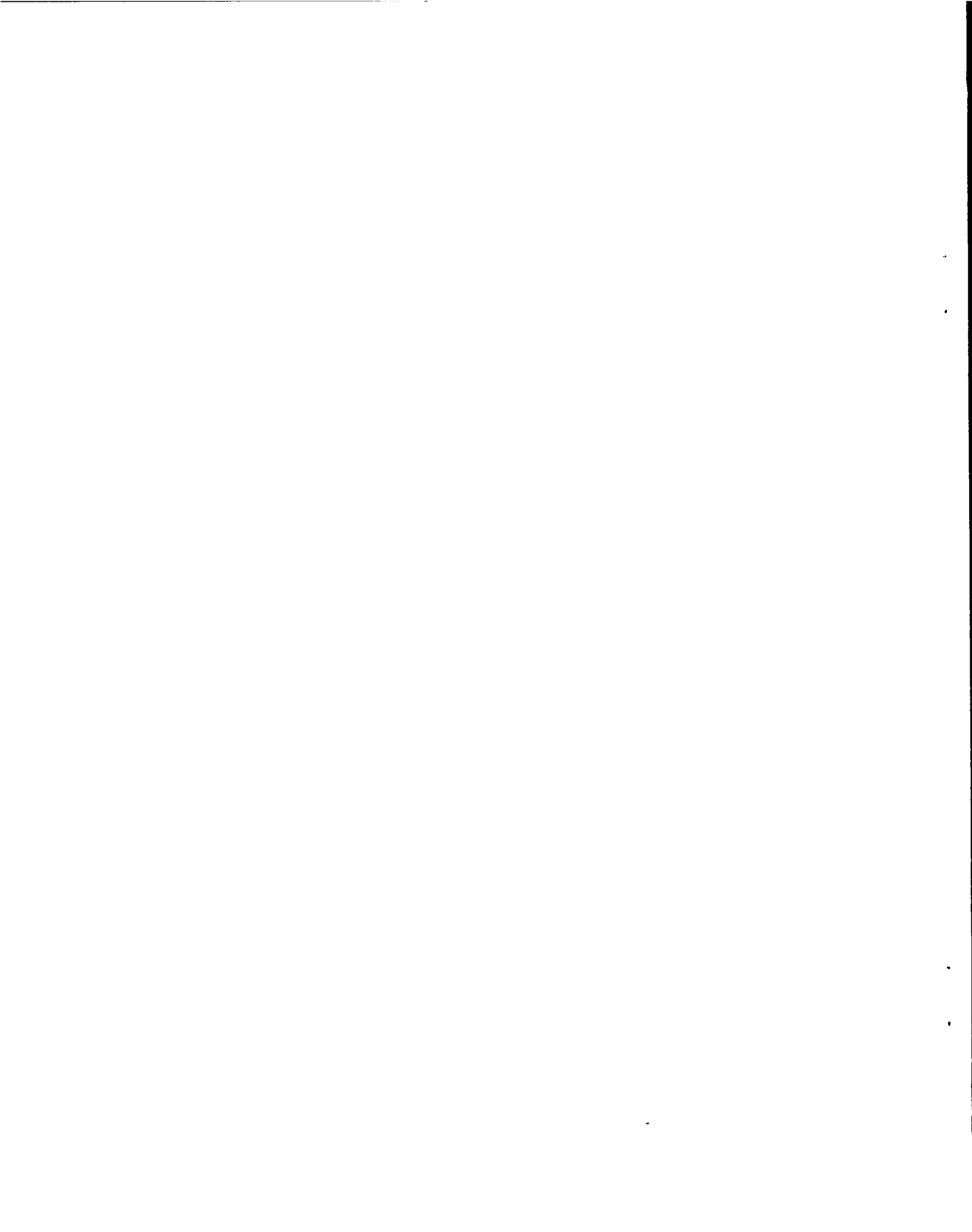
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# Oklo-Natural Fission Reactor Program

January 1—March 31, 1979

Alexander J. Gancarz





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OKLO-NATURAL FISSION REACTOR PROGRAM

January 1--March 31, 1979

by

Alexander J. Gancarz

ABSTRACT

The U-Pb chronologic portion of a comparative U-Ru U-Pb age dating study is complete. The data for the Cluff Lake Uranium Deposit indicate multiple episodes of uranium mineralization. Concentration of uranium started at  $1.3 \times 10^9$  years ago followed by intense pitchblende mineralization at  $1.05 \times 10^9$  and  $0.8 \times 10^9$  years ago. Important technical advances in Ru mass spectrometry have been made. It appears that the analytical difficulties hampering the U-Ru part of the study have been resolved. Additional analyses of Oklo uranium ore samples confirm that lead loss is diffusion controlled. Isotopic analyses of Pb in a conglomerate layer underlying the Oklo deposit indicate that this layer is a major Pb transport path and repository.

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I. INTRODUCTION

This report covers activities of the following individuals.

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II. PROGRAM OBJECTIVE

Funding for this program, which includes the United States' participation in the international investigation of the Oklo natural fission reactor phenomenon, commenced in fiscal year 1975. The first formal report of progress in this program was the annual report for fiscal year 1976, issued in November 1976. The Oklo phenomenon refers to the occurrence of self-sustaining fission chain reactions in a series of very rich uranium ore pockets located in an extensive Precambrian pitchblende deposit in Gabon, West Africa. The uranium formed critical masses about  $2 \times 10^9$  years ago. The duration of criticality was several hundred thousand years. During this time, approximately 6 tonnes of uranium were fissioned. Studies of the Oklo phenomenon have shown that many fission products were retained at the sites where they were generated. Principal objectives of the present investigation are identification of the migration paths of some of the mobile fission products and reconstruction of the paleohydrology and transport history.

Lead was chosen as the first element to be investigated for tracing transport paths. Although it is not a fission product, it is the decay product of U and previous work has shown that  $\sim 70\%$  of the radiogenic lead is missing from the Oklo reactor site. In addition, the common lead background is low and procedures for both elemental and isotopic analyses are relatively easy to perform. The second element to be used for tracing transport paths is ruthenium. Development of a reliable isotopic analysis procedure for ruthenium appears to be near completion. Developmental efforts were described in the previous quarterly report.

III. ACTIVITIES DURING THE CURRENT QUARTER

A. Comparative U-Pb and U-Ru Dating Study

An important aspect of developing the U-Ru dating technique is to compare U-Ru ages with U-Pb ages. Our comparative study is being done on the Cluff Lake Uranium ore deposit in Saskatchewan, Canada. We have completed a preliminary U-Pb chronologic study on the D ore body. The data and interpretations



are summarized in an extended abstract of a paper, Chronology of the Cluff Lake Uranium Deposit, Saskatchewan, Canada, to be presented at the International Uranium Symposium, Sydney, Australia, in June 1979. The abstract follows.

The Cluff Lake Uranium Deposit, northwestern Saskatchewan, Canada, is located at the southern edge of the Carswell structure, a window through the sedimentary Athabasca Formation into the underlying metamorphic basement complex. Like many of the other occurrences of U in Saskatchewan, the mineralization at Cluff Lake occurs near the contact of the basement rocks and the Athabasca Formation. The U mineralization is associated with concentrations of Te, Se, Au, Co, Ni, Cu, Pb, As, and S [1].

There are several U ore bodies at Cluff Lake. The mineralization of the D ore body occurs predominately in the Athabaskan rocks with only minor mineralization in the basement rocks. The D ore body contains massive pitchblende mineralization which is generally associated with bituminous material. Large excesses of either bituminous material or graphite without U mineralization are not observed.

This paper presents U-Pb and Pb isotopic data for rock samples from the D ore body and from other areas in the Carswell structure, which delineate the chronology of the deposit. This investigation is part of a more general comparative study of U-Pb and U-Ru chronologies.

There are several important constraints on the time of mineralization of the Cluff Lake deposits. They must have formed subsequent to the deposition of the Athabasca Formation. Recent Rb/Sr data on Athabaskan samples yield ages of 1.35 to 1.43 AE [2]. The mineralization of the D ore body predates the formation of the Carswell structure since faults inferred to be associated with its formation offset mineralized zones. K-Ar data on the Cluff Lake breccia inferred to be contemporaneous with the development of the Carswell structure, yield an age of 0.478 AE [3]. U-Pb mineralization ages of 1.05 to 1.15 AE have been reported [4].

U and Pb concentration data and Pb isotopic data are presented in Table 1. Lead abundances were determined by atomic absorption spectroscopy and have 10 percent uncertainties. U abundances were determined by delayed neutron counting of neutron irradiated samples. They have 5 percent uncertainties. Samples 10.33, 10.34, and 15.01 - 15.35 are from the D ore body and samples 15.39 - 15.41 are non-ore rocks from within the Carswell structure.

With one exception, the rock samples from the D ore body are mineralized pelitic sandstone of the Athabasca Formation. Sample 15.35 is a sample of mineralized basement rock. The samples are from drill holes or trenches made in conjunction with the exploration program of Amok, Ltd. and were supplied by Dr. K. Tapaninen. The one isotopically anomalous sample, 15.02, is in effect a surface sample, as it was collected from the contact between mineralized sandstone and overlying recent glacial gravel and sand. The data are included, but are not considered in the subsequent discussion.

The ore samples were selected to have a wide range of U concentrations and as shown in Table 1 vary from 0.5% to 62% U.  $^{204}\text{Pb}$  is very abundant with concentrations ranging from 0.5 to 325 ppm (average 50 ppm), which, assuming normal crustal Pb isotopic abundances correspond to total common Pb concentrations ranging from 34 ppm to 2.25% (average 0.35%). Therefore, the rocks are highly enriched in common lead.

Three rocks from within the Carswell structure were selected because of their high concentrations of Pb relative to U. U concentrations range from 0.5 to 600 ppm. Again, assuming normal crustal Pb isotopic abundances, the  $^{204}\text{Pb}$  concentrations correspond to 19 to 820 ppm total common Pb.

All of the ore samples are discordant showing relative loss of Pb with respect to U. Assuming an age of  $\sim 1$  AE, the average loss of radiogenic  $^{206}\text{Pb}$  is 65%. No samples with excess radiogenic Pb relative to U have yet been observed in the vicinity of the D ore body. Two of the three rocks from outside the D ore body, but from within the Carswell structure, are discordant with relative Pb loss. The third sample, collected  $\sim 18$  km NNW from the D ore body, shows an 8% enrichment of Pb relative to U.

TABLE 1: CLUFF LAKE U-Pb DATA

Sample	Pb g Pb/g rock	U g U/g rock	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{204}\text{Pb}/^{206}\text{Pb}$	$\mu$
10.33	$7.61 \times 10^{-2}$	$5.8 \times 10^{-1}$	$0.041787 \pm 4$	$0.088149 \pm 5$	$0.001062 \pm 1$	$6.98 \times 10^3$
10.34	$1.39 \times 10^{-1}$	$6.2 \times 10^{-1}$	$0.11390 \pm 1$	$0.12522 \pm 1$	$0.002920 \pm 1$	$1.63 \times 10^3$
(Repeat)			$0.11393 \pm 1$	$0.12532 \pm 1$	$0.00292 \pm 4$	$1.63 \times 10^3$
15.01						
(1st leach)	$5.32 \times 10^{-3}$	$1.30 \times 10^{-1}$	$0.061519 \pm 4$	$0.091557 \pm 5$	$0.001540 \pm 2$	
(2nd leach)	$3.41 \times 10^{-3}$	$1.31 \times 10^{-2}$	$0.10525 \pm 4$	$0.11087 \pm 2$	$0.002687 \pm 2$	
(Total)	$8.73 \times 10^{-3}$	$1.43 \times 10^{-1}$	$0.078028$	$0.098848$	$0.001973$	$8.45 \times 10^3$
15.02	$6.53 \times 10^{-4}$	$2.69 \times 10^{-2}$	$0.03549 \pm 1$	$0.061527 \pm 8$	$0.000810 \pm 2$	$4.80 \times 10^4$
15.04	$6.832 \times 10^{-3}$	$1.07 \times 10^{-1}$	$0.01506 \pm 3$	$0.07808 \pm 3$	$0.00029 \pm 1$	$5.43 \times 10^4$
15.15						
(1st leach)	$1.12 \times 10^{-2}$	$5.71 \times 10^{-1}$	$0.01747 \pm 1$	$0.07857 \pm 1$	$0.000427 \pm 3$	
(2nd leach)	$9.47 \times 10^{-3}$	$1.83 \times 10^{-2}$	$0.011796 \pm 1$	$0.077031 \pm 3$	$0.000287 \pm 1$	
(Total)	$2.07 \times 10^{-2}$	$5.89 \times 10^{-1}$	$0.01486$	$0.07786$	$0.000363$	$7.38 \times 10^4$
15.15	$4.84 \times 10^{-2}$	$6.07 \times 10^{-1}$	$0.013341 \pm 3$	$0.077282 \pm 6$	$0.000300 \pm 2$	$3.92 \times 10^4$
15.18						
(1st leach)	$7.28 \times 10^{-3}$	$4.80 \times 10^{-1}$	$0.009582 \pm 1$	$0.078057 \pm 5$	$0.000221 \pm 1$	
(2nd leach)	$7.09 \times 10^{-3}$	$2.56 \times 10^{-2}$	$0.007566 \pm 2$	$0.07764 \pm 1$	$0.000184 \pm 1$	
(3rd leach)	$2.32 \times 10^{-2}$	$1.16 \times 10^{-3}$	$0.007341 \pm 1$	$0.077533 \pm 4$	$0.000166 \pm 1$	
(Total)	$3.76 \times 10^{-2}$	$5.07 \times 10^{-1}$	$0.007817$	$0.07765$	$0.000180$	$6.99 \times 10^4$
15.25						
(1st leach)	$2.85 \times 10^{-4}$	$4.81 \times 10^{-3}$	$0.07700 \pm 3$	$0.09148 \pm 2$	$0.00163 \pm 3$	
(Residue)	$8.46 \times 10^{-5}$	$9.79 \times 10^{-5}$				
(Total)	$3.70 \times 10^{-4}$	$4.91 \times 10^{-3}$				
15.31	$4.52 \times 10^{-3}$	$4.58 \times 10^{-2}$	$0.066935 \pm 1$	$0.094759 \pm 4$	$0.001633 \pm 1$	$6.22 \times 10^3$
15.35	$1.26 \times 10^{-3}$	$1.02 \times 10^{-2}$	$0.05148 \pm 16$	$0.082423 \pm 24$	$0.001112 \pm 4$	$7.12 \times 10^3$
15.39	$1.80 \times 10^{-3}$	$6. \times 10^{-7}$	$2.6916 \pm 3$	$0.48249 \pm 6$	$0.028155 \pm 6$	$4.33 \times 10^{-2}$
15.40	$7.00 \times 10^{-4}$		$2.5077 \pm 3$	$0.8015 \pm 1$	$0.05094 \pm 5$	
15.41	$4.35 \times 10^{-4}$	$6. \times 10^{-4}$	$6.5859 \pm 6$	$0.15800 \pm 2$	$0.005144 \pm 8$	$1.80 \times 10^3$

There do not appear to be among the U-Pb data any systematic relationships yielding well-defined chronologic information. The youngest  $^{206}\text{Pb}/^{238}\text{U}$  model age is 0.234 AE. Consequently, U and/or Pb within the D ore body must have been redistributed more recently than 234 m.y. ago. This is younger than the 478 m.y. age for the formation of the Carswell structure, and the event leading to the formation of the Carswell structure does not appear to be recorded in the U-Pb systems of the analyzed ore samples.

In contrast to the U-Pb data, the Pb isotopic data yield considerable chronologic information. Pb data for the three rocks from outside the D ore body form a straight line on a plot of  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{204}\text{Pb}/^{206}\text{Pb}$  intersecting the  $^{207}\text{Pb}/^{206}\text{Pb}$  axis at a value corresponding to an age of  $1.33 \pm 0.03$  AE. This age is similar to, although slightly younger than, the Rb/Sr deposition age of the Athabasca Formation [2]. This line also intersects a normal ( $\mu \sim 9.4$ ) Pb evolution curve at values corresponding to ages ranging from 1.3 AE to 1.5 AE, indicating normal U/Pb ratios in the basement complex rocks prior to 1.33 AE. Thus, it appears that within 50 to 100 m.y. after the deposition of the Athabasca Formation, U and Pb were redistributed, resulting in the concentration of U in rocks in the Cluff Lake region.

The very rich U ore samples 15.15 and 15.18 (U > 50%) and a leach on sample 15.06 all contain very radiogenic Pb with  $^{206}\text{Pb}/^{204}\text{Pb} \sim 4000$ . These data indicate an age for the extensive massive pitchblende precipitation of  $1.05 \pm 0.03$  AE. Sample 10.33 and 10.34 are also very rich ore samples, but contain large quantities of common Pb, 71 and 293 ppm of  $^{204}\text{Pb}$  respectively, and  $^{206}\text{Pb}/^{204}\text{Pb} \sim 500$ . As indicated by their high  $^{207}\text{Pb}/^{206}\text{Pb}$  values, they contain significant initial Pb which is quite radiogenic. Assuming this initial Pb evolved from 1.33 AE to 1.05 AE implies an average  $\mu$  (i.e.  $^{238}\text{U}/^{204}\text{Pb}$ ) in the time interval of  $\sim 1000$ . Samples 10.33 and 10.34 were also analyzed for Ru and the U-Ru data yield an age of 1.5 AE [5]. This is older than the U-Pb age of 1.05 AE for the massive pitchblende mineralization. These samples, however, may contain not only initial Pb, but also initial Ru and thus, the U-Ru ages are artificially old.

The remaining analyzed samples are all relatively poor in U, with concentrations ranging from 14.3% to 0.49%. A two part leaching experiment on sample 15.01 produced Pb with  $^{207}\text{Pb}/^{206}\text{Pb}$  values ranging from .09156 to 0.11087. These data lie on a line which intersects the  $^{207}\text{Pb}/^{206}\text{Pb}$  axis at a value corresponding to 0.80 AE. The other lower grade ore samples lie along this line. These data suggest yet another period of disturbance at  $\sim 800$  m.y. with the production of lower grade ore. These lower grade ores do contain a radiogenic initial Pb. Assuming that this initial Pb evolved from 1.33 AE to 0.80 AE implies an average  $\mu$  of  $\sim 500$ . Assuming evolution from 1.05 AE to 0.80 AE implies an average  $\mu$  of  $\sim 900$ . These values of  $\mu$  (500 to 1000) in comparison to the values in the rich U ores (1,600 to 70,000) suggest that the U in lower grade ores is not remobilized U from the rich ore, but rather new U transported in from outside the Cluff Lake region.

In summary, the U-Pb data indicate a complex history of mineralization starting with the deposition of the Athabasca Formation. These data do not, however, help to resolve whether the source region of the U was the metamorphic basement complex or the Athabasca Formation sediments. The major massive mineralization occurred at a 1.05 AE with a second episode of mineralization at

800 m.y. and a U-Pb disturbance at times younger than 234 m.y. Multistage mineralization is also indicated by petrologic [6] and U-Pb isotopic studies [7] at Rabbit Lake, Saskatchewan. This deposit, like Cluff Lake, occurs at the contact between the basement rocks and the Athabasca Formation, as do many other of the U deposits in northern Saskatchewan, suggesting that the processes and general time scales of mineralization identified at Cluff Lake may be common to the other deposits.

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#### B. Development of a U-Ru Dating Technique

The difficulty with the U-Ru part of the comparative study has been in the quantitative assay of ruthenium and of ruthenium isotopic abundances, both at the nanogram level. Thermal ionization mass spectrometry is being used to measure the isotopic abundance and isotope dilution with a  $^{96}\text{Ru}$  spike is being

used for quantitative assay. The work this quarter resulted in marked improvement of Ru isotopic analysis. The mass spectrometry no longer appears to be a serious limitation. Modification of an ion source has provided one to two orders of magnitude increased sensitivity for ruthenium, while greatly reducing the molybdenum spectral interferences.

Previous mass spectrometric development showed a significant increase in Ru sensitivity when a sample of a few nanograms was loaded onto a resin bead, which in turn, was mounted on a Re filament. Mo interferences were still significant and precluded precise analyses of nanogram size Ru samples. This increased sensitivity was partially offset by increased low level hydrocarbon ion spectral interferences. Mo emission from the Re filament was significantly lowered by shortening the filament from .250" to .050". This gave one-fifth as much filament facing the source and reduced the interference by a comparable amount. Even with this improvement, there was too much Mo in the spectrum to allow reliable measurement of  $^{100}\text{Ru}$ , which is the isotope used to distinguish natural Ru from fission Ru. There was an improvement in the peak shape on the mass spectrometer with samples analyzed with this short filament, but no increase in sensitivity.

This quarter improved Ru sensitivity has been achieved by altering the Z axis focusing portion of the mass spectrometer ion source. In general, ion sources are masked in the Z axis to provide an opening slightly higher than the height of the filament. We masked the Z axis to correspond to the short filament. The first three plates were masked to .120", leaving .035" clearance above and below the .050" filament. The optimum voltage settings on the Z focusing and deflection plates in the ion source changed radically, while those for the other plates changed little.

Several effects were immediately obvious. These are:

- 1) A 20 to 50 fold increase in Ru sensitivity which is much greater than that achieved with use of the resin bead.
- 2) Reduced Mo interferences from the Re filament.
- 3) Greatly increased sensitivity for other elements such as Zr present as impurities in the sample.
- 4) A further improved peak shape.

The altered Z focusing properties suggest that much or all of the improved sensitivity comes from improved Z axis focusing in the sources. The higher

drawout potential on the filament may also play some role. The actual sensitivity increases are difficult to measure because the optimum filament temperature appears to be lower for this source configuration. The sensitivity increase for Ru is between 20 and 50.

Improved chemistry is required to take maximum advantage of this increased sensitivity. Efforts to miniaturize the chemistry are continuing. Zr is more of a problem than it was previously, which suggests that its sensitivity increase may be even greater than that for Ru.

With this modified ion source the Ru peak shape is equal to that obtained with uranium and plutonium. This allows for much improved reliability in the peak stepping data collection mode. Precision is improved and accuracy is more repeatable with the better peak shape.

This redesigned source shows such an improvement that the resin bead method is not being used at this time. This eliminates the difficulties of getting a high percentage of the Ru loaded onto one or two resin beads and physically transferring the beads to a filament, and avoids the low level of hydrocarbons in the spectrum.

Several samples of Ru separated from the rich U ore deposits of Cluff Lake have been analyzed and precision ranges from 0.25% to 0.50% for the major nuclides (99, 101, 102, 104). The measurements of the  $^{100}\text{Ru}$  nuclide are sufficiently accurate for the subtraction of natural Ru. Further development will probably improve this somewhat, but the present technique should be adequate for the testing of the age dating technique. These data have not been completely calculated yet, but the data from three filament loadings of a natural plus spike mix of Ru are reported in Table I. These data have not yet been bias corrected, and each loading is  $25 \times 10^{-9}$  of total Ru. This gives about  $8 \times 10^{-9}$  grams each of 102 and 96, and proportionally less of the other nuclides.

### C. Pb Transport from the Oklo Uranium Deposit

U-Pb and Pb isotopic analyses on all the additional U-ore and most of the non-ore Oklo samples obtained in France in the fall of 1978 are complete. These data along with our previous analyses of Oklo samples indicate the following:

1. The escape of Pb from uraninite grains, including not only the Pb produced by U decay, but also Pb incorporated into grains at the time of formation, is diffusion controlled.

TABLE I  
Ru RATIOS ON NATURAL + SPIKE MIX (25 ng TOTAL Ru)

Filament	Count Rate/Sec.	104/102	101/102	100/102	99/102	96/102
1	$4 \times 10^4 - 10^5$	.5831	.5446	.4080	.4162	.9685
2	$4 \times 10^4 - 10^5$	.5858	.5432	.4089	.4146	.9703
3	$3 \times 10^4 - 5 \times 10^4$	.5835	.5460	.4117	.4184	.9753
		avg. .5841	.5446	.4095	.4164	.9714
		S.D. .0015	.0014	.0019	.0019	.0035

2. Some ore samples are enriched in Pb relative to U, and this extra Pb has an isotopic composition indicating that it is the Pb which has diffused from uraninite grains.
3. The conglomerate which stratigraphically underlies the Oklo ore deposit is a transport path and major repository of the Pb from the ore deposit.

The data from which these conclusions are drawn are in Table II and illustrated in Figures 1 and 2. Fig. 1 is a graph of  $^{207}\text{Pb}^*/^{206}\text{Pb}^*$  versus  $^{238}\text{U}/^{206}\text{Pb}^*$ .

TABLE II  
OKLO U-Pb DATA

Sample	Pb g Pb/g rock	U g U/g rock	$^{208}\text{Pb}/^{206}\text{Pb}$	$^{207}\text{Pb}/^{206}\text{Pb}$	$^{204}\text{Pb}/^{206}\text{Pb}$	$\mu$
KN 5	$1.3 \times 10^{-3}$	$3.0 \times 10^{-3}$	$0.02148 \pm 1$	$0.13566 \pm 7$	$0.000462 \pm 1$	$5.0 \times 10^3$
KN 6	$3.50 \times 10^{-3}$	$1.13 \times 10^{-2}$	$0.01353 \pm 1$	$0.12326 \pm 4$	$0.000238 \pm 2$	$1.3 \times 10^4$
KN 56	$2.35 \times 10^{-3}$	$5.9 \times 10^{-3}$	$0.02119 \pm 1$	$0.13138 \pm 4$	$0.000425 \pm 2$	$5.9 \times 10^3$
KN 61	$1.39 \times 10^{-3}$	$2.8 \times 10^{-3}$	$0.05772 \pm 1$	$0.14594 \pm 2$	$0.001160 \pm 2$	$1.8 \times 10^3$
KN 76	$2.06 \times 10^{-3}$	$5.9 \times 10^{-3}$	$0.011869 \pm 3$	$0.115840 \pm 8$	$0.000186 \pm 1$	$1.5 \times 10^4$
KN 187	$6.72 \times 10^{-3}$	$3.47 \times 10^{-2}$	0.008928	$0.10718 \pm 1$	$0.000178 \pm 1$	$2.8 \times 10^4$
SC 0 - 2261	$7.28 \times 10^{-4}$		$0.08784 \pm 1$	$0.15413 \pm 2$	$0.000886 \pm 1$	
SC 50 - 2273	$2.89 \times 10^{-3}$		$0.07982 \pm 2$	$0.14277 \pm 4$	$0.000945 \pm 1$	
KN 230 - 2460b	$1.6 \times 10^{-3}$	$3.9 \times 10^{-4}$	$0.01671 \pm 3$	$0.13829 \pm 11$	$0.00037 \pm 1$	$6.5 \times 10^2$
SC 48 bis/2288	$9.5 \times 10^{-4}$	$2.0 \times 10^{-4}$	$0.11329 \pm 2$	$0.16210 \pm 2$	$0.001973 \pm 4$	$1.2 \times 10^2$
F <sub>B</sub> Oklo pelite whole rock pyrite						

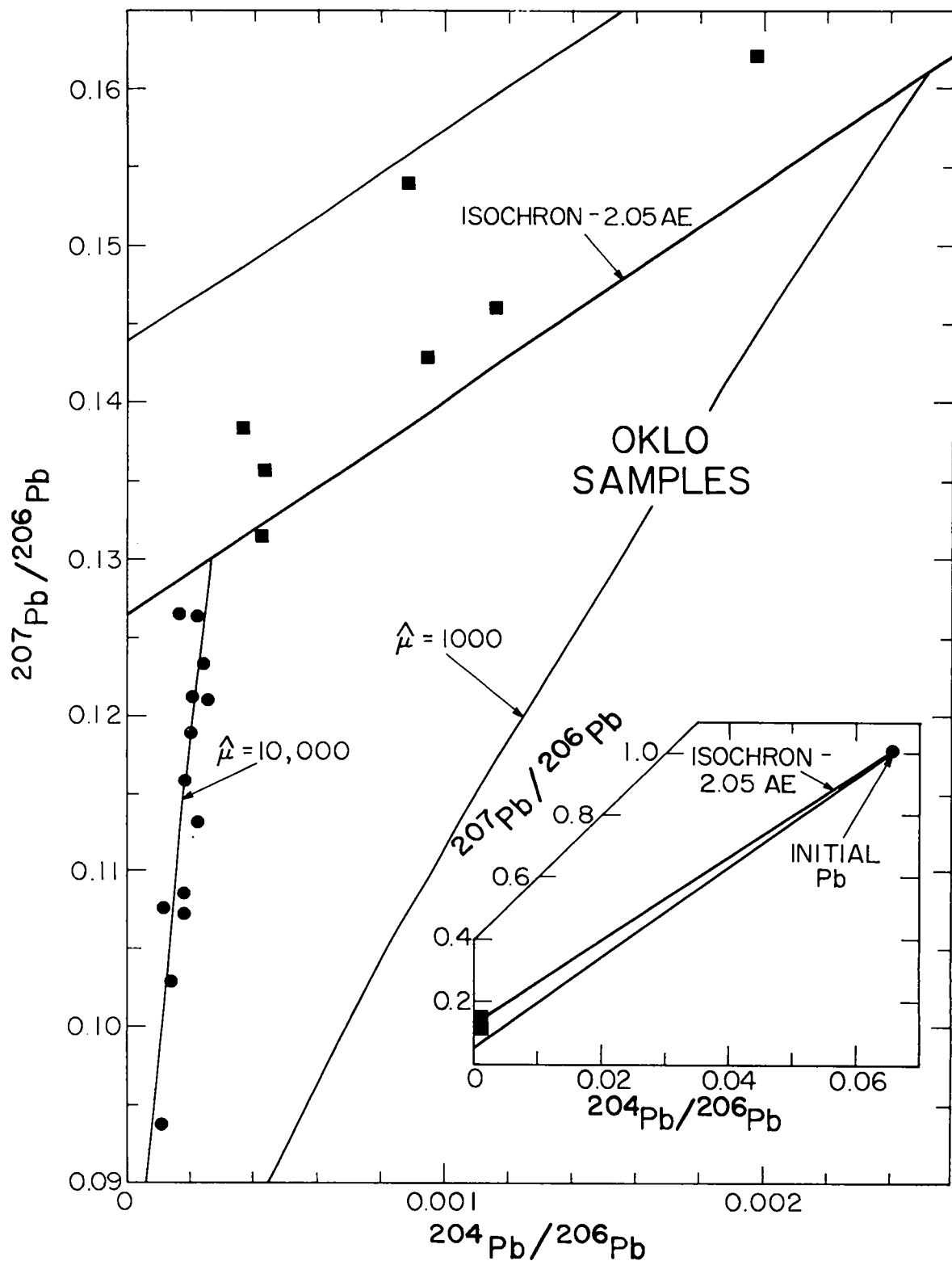


Fig. 2. Plot of  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{204}\text{Pb}/^{206}\text{Pb}$  for Oklo samples. The circles are for samples which show Pb loss relative to U and the squares are for samples which show relative Pb gain.



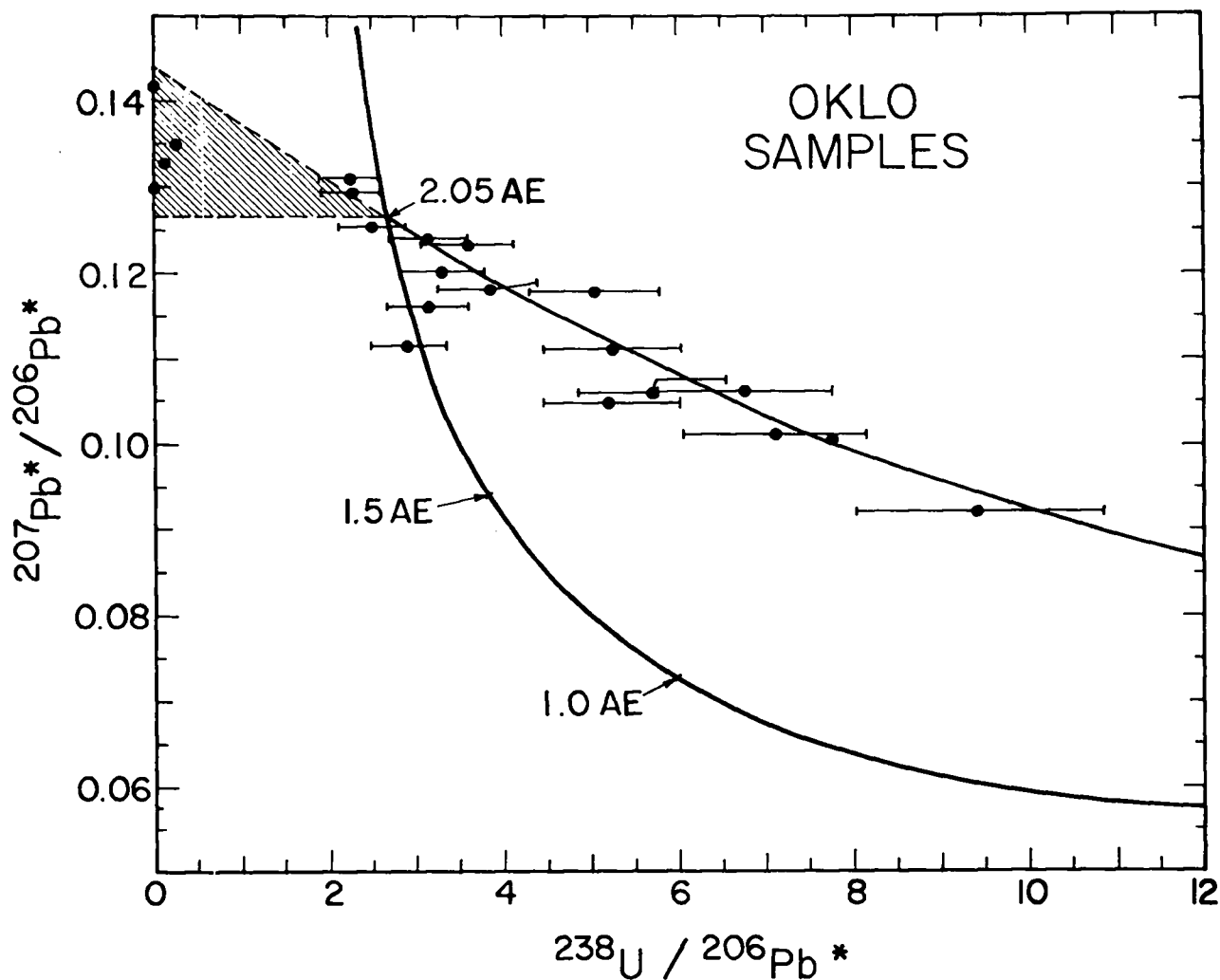


Fig. 1. Graph of  $^{207}\text{Pb}^*/^{206}\text{Pb}^*$  versus  $^{238}\text{U}/^{206}\text{Pb}^*$  with data for Oklo samples.

The \*'s indicate only Pb produced by in situ U decay (i.e. radiogenic Pb), and not Pb incorporated into grains at the time of formation. The amount of initial Pb is determined by the amount of  $^{204}\text{Pb}$ , which has no long-lived precursor. The lower curve is concordia, and the upper curve is the continuous volume diffusion trajectory. The data to the right of the concordia curve, i.e. in the direction of Pb loss, in general lie on the diffusion curve indicating this is the mechanism of loss of the Pb produced by U decay. Fig. 2 is a graph of  $^{207}\text{Pb}/^{206}\text{Pb}$  versus  $^{204}\text{Pb}/^{206}\text{Pb}$ . On this plot, the initial Pb is not subtracted from the total Pb. The data on Fig. 1 which show relative Pb loss are plotted as the circles on this figure. They lie on the line labelled  $\hat{\mu} = 10\ 000$  which is the continuous volume diffusion trajectory for Pb. Therefore, it appears that not

only is the radiogenic Pb (i.e. Pb\*) lost by a diffusion mechanism, but also the initial Pb is lost by this mechanism.  $\hat{\mu}$  is  $^{238}\text{U}(\text{today})/^{204}\text{Pb}(\text{time of formation})$ .  $\hat{\mu}$  for normal crustal rocks is about 10. Therefore, the relative concentration of U to Pb during the formation of the Oklo deposit was about 1000:1.

The shaded area on Fig. 1 is the area where data for samples which have gained diffused radiogenic Pb relative to U plot. This is an area rather than a line since the  $^{207}\text{Pb}^*/^{206}\text{Pb}^*$  of the diffused component is variable depending on the value of the effective diffusion coefficient ( $D/a^2$ ). Similarly, on Fig. 2, the region above the line labelled isochron is the area where data for samples which have gained diffused radiogenic and diffused initial Pb relative to U plot. The data for Oklo samples with excess Pb all lie in these two regions. The conclusion is, therefore, that this excess Pb is the component diffused out from uraninite grains.

These data do not specify how once out of the uraninite grains, the Pb was transported into these Pb enriched samples. However, samples SC 0, 50, 48, 6 and KN 230 are all from the basal conglomerate under the Oklo deposit and have excess Pb. The presence of this diffusion component indicates that the basal conglomerate is a major transport path and repository for Pb from the uranium deposit.

#### D. Pb Transport Around the Key Lake Uranium Deposit

All of the samples collected at Key Lake last quarter arrived in Los Alamos. They have been unpacked and catalogued. We are beginning to prepare samples for U-Pb analysis.

#### E. Conclusions

A U-Pb chronology indicating multiple episodes of U mineralization has been established for the Cluff Lake U deposit. Additional data on Oklo samples confirm that Pb is lost by a diffusion mechanism. A major Pb transport path and repository has been identified at Oklo.

#### F. Communications and Publications

##### Talks

R. J. Vidale "Oklo A Natural Fission Reactor", January 31, 1979  
- Joint Chemistry-Physics Department Seminar, Case-Western  
Reserve University, Cleveland, Ohio.

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R. J. Vidale "Oklo A Natural Fission Reactor", January 31, 1979  
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R. J. Vidale "Oklo - A Natural Fission Reactor", February 1, 1979  
- University of Michigan, Distinguished Women Lecture Series, Ann Arbor, Michigan.

D. G. Brookins "The Significance of the Oklo Uranium Deposit to Problems of Radioactive Waste Repositories in Geomedia, February 19, 1978, University of British Columbia, Vancouver, B. C., Canada.

### Publications

A. J. Gancarz Chronology of the Cluff Lake Uranium Deposit, Saskatchewan, Canada. Submitted to: International Uranium Symposium on the Pine Creek Geosyncline, June 4-8, 1979, Sydney, Australia.

## IV. PROBLEMS

There do not appear to be any severe problems at this time.

## V. ACTIVITIES PLANNED FOR NEXT QUARTER

The main emphasis in the next quarter will be to establish a chronology for the Key Lake U deposit, determine the amount of Pb loss, and try to establish preferential directions of Pb transport. In addition we will continue the Ru isotopic analyses of the Cluff Lake ore samples.