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Application of Nuclear Criticality Safety to Early Earth Age Uranium

Norm Schwers and John Miller

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Introduction

- This review discusses the natural criticality events which occurred in the Oklo/Gabon area and compares that to some currently known uranium deposits
  - Natural reactors were proposed in 1956: Oklo region reactors discovery in 1972
- Natural decay of uranium; U^{235} and U^{238} enrichment and quantity/mass
- Method for identification of the age of the deposit: decay to lead U^{238} -> Pb^{206} and U^{235} -> Pb^{207}
  - U -> Pb decay chains are well defined and geologists determined the Earth to be 4.54 billion years old (Ba)
  - This review calculated various U mass values, and evaluates various criticality parameters going back to 4.54 Ba

Uranium deposits in nature

- Reports include 14 different rock and mineral formations of U deposits
  - Most are silica oxides and uranium oxides of various forms
- Known enrichment in all known U deposits is (0.72%) consistent across earth with one exception (Oklo region)
- Most reports of decay products (i.e., are very low) reportedly due to lead leaching
- Cosmic materials show significantly lower concentrations than Earth crust
- Mantle lava is lower than the quantities in the crust

Uranium Enrichment

- The effect of going back in time increases the mass as well as the enrichment
- Inverted time from today

Time (yrs)	^{235}U	^{238}U	Ratio	U Total	Enrichment
0	1	137.9	137.9	138.9	0.720%
1.0E+06	1.00	138	137.79	138.92	0.721%
7.04E+08	2.00	154	76.91	155.81	1.284%
1.00E+09	2.68	161	60.16	163.71	1.635%
2.00E+09	7.17	188	26.24	195.22	3.671%
3.00E+09	19.2	220	11.45	238.79	8.033%
4.00E+09	51.3	256	4.99	307.80	16.682%
4.54E+09	87.4	279	3.19	366.24	23.860%

  - $T_{1/2}$ U^{238} 4.468 billion years (1X $T_{1/2}$ = 2 times current content)
  - $T_{1/2}$ U^{235} 703.8 million years (6.4 X $T_{1/2}$= 84 times current content)
- ANSI/ANS 8.1 subcritical limit for Saturated solution U_3O_8 is 0.96% which corresponds to about 3.5E8 years

Lead – Primordial or Radiogenic

- Lead in nature:
  - 14 ppm in the Earths crust and uranium is 2.7 ppm (ratio U:Pb = 0.19)
  - Reports that propose the Earth's age list the U:Pb ratio as 7.5 to 8.2
  - Chart of Nuclides Pb^{204}=1.4%, Pb^{206}=24.1%, Pb^{207}=22.1%, Pb^{208}=52.4%
  - Inconsistencies of Earth's Pb ppm and the isotopic content in the crust.
- If all of the Pb^{206} and Pb^{207} are radiogenic, then the starting point would have had to be at an enrichment nearly 50% (nearly 6 Ba)

Lead – Primordial or Radiogenic

- Reportedly the U:Pb ratio adjacent to U deposits is low due to lead preferentially leached away from the uranium deposits.
  - Example of lead leaching preferentially into a system is from the Flint MI water project. Change water supply, changes pH and electrical potential, other chemicals affect any passivating layer
  - Plausible for an open system, however from a criticality safety standpoint, water for leaching would significantly boost reactivity.
- Assume some Pb^{206} and Pb^{207} is primordial AND if some lead leaches out of the system: creates significant errors in the estimates of the age of the earth

Oklo Reactors

- In Oklo reactors, different quantities of U are involved in each of the 16 natural reactors
  - Some Oklo deposits have been fully mined (some very near surface)
  - U concentration between 0.1% up to 10% (remainder being rock)
  - Enrichment at estimated time of Oklo is 3.7% (2 Ba)
- Reportedly operated at 100kW for 1 million years
  - Steady state reactor operation is hard without control mechanisms
    - Likely a moderator expansion and expulsion from the reactor system
  - Cycling at higher powers as water ingress is more likely. Heat and cracking could allow more paths for water into the rock
- Why did Oklo reach criticality and others didn't?
  - Mass and Concentration are not reported nearly as high as the MacArthur River uranium deposit

Canadian Mine: MacArthur River

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- Evaluated because it is high grade ore; concentration is >17% and mass is 580,000 MT discovered in 1988 (after Oklo)
  - U tailings data shows Pb < 1%, but based on decay data, the lead could be approximately >60%.
  - Materials identified in tailings are primarily quartz, calcium sulphate, and illite (clay-like substance).
  - Compare 580,000 MT to ~125 tonnes U in a BWR
  - Compare high enrichments in early Earth to Oklo estimated to be 3.7%
  - Problem uncertainties
    - There is no consistent marker to prove when the uranium ore concentrations came together. This affects enrichment and mass.
    - Assumption: sandstone mixed into system

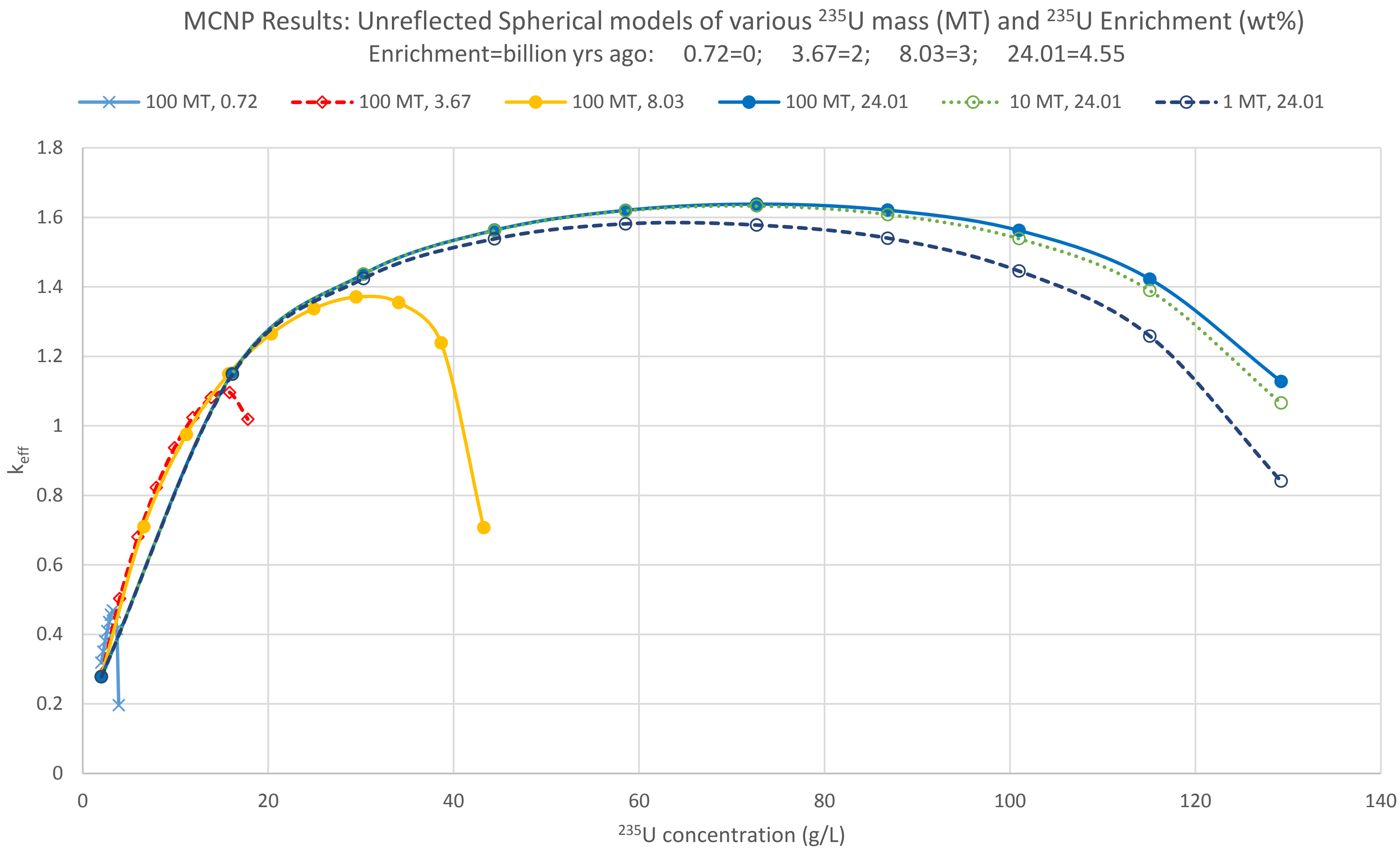
Criticality Safety factors

- Moderator – anticipated to be low, but water would fill pores
  - If low, how did it leach out lead uniformly across all forms of uranium deposits– mines in Canada are 100-450 m deep
- Concentration – uncontrolled, but may have varied.
  - Some mines in Canada >20% by mass
- Enrichment – remarkably consistent across the globe. Reduced slowly over time.
- Mass – tremendous mass in the hundreds of thousands of metric tons and would have been much more (2.6 times today's value)
- Geometry – Varies from site to site
- Absorption – would vary by materials in each mine
- Reflection – rock, uranium, metals, sandstone, etc.

MCNP Calculation and Analysis

- Information provided here evaluated three mass levels for various past times (higher enrichment)
  - System as modeled is U_3O_8 missed with SiO_2
  - Water added into porosity region
- The K_{eff} eigenvalues are not real.
  - The values prompt critical (keff = 1.007) are unlikely given that the water likely seeps in slowly and gets expelled due to fission heat.
- The high eigenvalues at early earth age is inconsistent with a lack of natural criticality. Something prevented the natural criticality.
- Criticality is more likely for early age uranium due to higher enrichment, higher mass, and wider range of favorable moderation values.

MCNP Results



Flooded left and dry right

Conclusions

- There is an indeterminate amount of lead Pb^{206} and Pb^{207} that is primordial
- Some U and Pb leaching out of the system results in U:Pb ratios that are inaccurate for deposit age estimation
  - The lack of Pb in U deposits could be from reduced chronological time frame, or from natural causes (leaching from open system)
  - Leaching requires water and water increases uranium reactivity: making criticality more likely
- U mass and enrichment are significantly higher in earlier chronological time.
  - Natural criticality is easily achieved in high concentration ore
  - The lack of criticality is evidenced by the consistent uranium enrichment