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Application of Nuclear Criticality Safety to Early Earth Age Uranium

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INTRODUCTION

Application of Nuclear Criticality Safety methods to early earth age uranium results in uncontrolled nuclear criticality events. This review considers the parent nuclides of the radioactive decay process for uranium as well as evaluation of the lead (Pb) daughter products. Most studies of the age of the earth use the $^{235}U/^{238}U$ ratios as well as the daughter products of radiogenic lead.

While the half-life values are not in question, this paper evaluates the concentration of Uranium soil-rock mixtures and the uranium isotopic ratios at various times going back to the early earth. Mass and enrichment are highest at the earliest times. With increased uranium, it is anticipated that the concentration of uranium in the soil is also at its highest. The current age of the earth is reported to be approximately 4.55 billion years old. Mass and enrichment are the factors most affected by time. Other primary factors of nuclear criticality safety are generally independent of time.

Concentration and Mass

Concentration is the weight percent of uranium among all other materials in a given analyzed sample. The half-life of ^{238}U is 4.47 billion years and ^{235}U is 704 million years. The current ratio of ^{235}U to ^{238}U is 1:137.88 (an ^{235}U enrichment of 0.72x wt.%). Working backwards in time, the ratio at 4.55 billion years would have been approximately 88.3 times the current ^{235}U mass and increases to 24.0 wt.%. The ^{238}U would have also increased by a factor of 2.02. Increasing enrichment from 0.72% to 24%, should increase reactivity and supposedly more natural criticality events should have occurred. However, other criticality safety factors are affected by the reverse chronology evaluation. Mass and concentration are simultaneously affected by the reverse chronological effect. The uranium concentration is evaluated as an assumption that within an open system, there will be a wide variety of concentrations of rock and sedimentary formations where uranium is found. Just as half-life decay reduces the uranium materials, the half-life buildup will double the fissile material for each half-life. It is however, more complicated, because the decay of uranium decays to radiogenic lead.

Given the time since the earth was formed, uranium would have decayed to radiogenic lead. However, there is a lack of lead identified in the mill tailings. If the ratio of lead increases from earth formation to

current times, the concentration of lead should have increased by 2.66 of the original nuclide value from both the decay of ^{235}U and ^{238}U. Given, standard decay process to radiogenic lead, the concentration of lead in the surrounding materials should be 1.66 times that of uranium.

Some mines have a uranium concentration below 1% by weight and others are greater than 20%. The World Nuclear Org estimates Canadian mining mass to be greater than 580,000 metric tons at approximately 20wt% uranium. Given a current concentration 20%, the ^{238}U would have more than doubled and the ^{235}U would be 88 times as large.

For this paper, the criticality event is significantly more likely with the higher uranium concentration. Given a concentration of 20% Uranium as found in some mines, the concentration at earth formation should be significantly higher to a maximum of greater than 40% theoretical maximum. This assumes that the concentration of the earth formed around the uranium is approximately constant. Published estimates of uranium in the universe, sun, and earth's oceans are 3-4 orders of magnitude below the estimates of uranium in earth's crust. When considering the earth's crust, it is important to note that the material is not localized in one region. The top 10 uranium producing mines in the world are spread out on five continents.

Lead Analysis

Uranium is found in more than a dozen different types of earth including sand, sandstone, feldspar, granite, etc. This document reviews a couple of the mixtures sandstone and granite. In each of the documents reviewed, there is a lack of lead identified in the mill tailings. One was identified as in the low hundreds of PPM. If the decay is accurate and there is no natural separation, then there should be greater than 300,000 PPM. Neither the EPA nor the US NRC websites list lead as a significant issue in uranium mill tailings. Some documents report that the lead leaches from the uranium. It is recognized that there would likely be some leaching in soil, and less through rock formations. Without lead being in the mill tailings, leaching would be consistent between all different material formations in all locations around the world. From a nuclear criticality perspective, any water leaching would also be a significant contributor to a nuclear chain reaction.

Additional Criticality Safety Factors

There are additional criticality safety factors which vary depending on the material. Loose packed soil is less effective reflection than the more dense materials. Thus far in the research, we have modeled only SiO2. Other sediments could add significantly to reactivity if they are moderating materials such as carbon based materials.

Interaction/heterogeneity would only be a factor if there were migrations of pockets of uranium based material that were slowly moving together. It is not clear that interaction is a factor and is not evaluated. Mass and enrichment start out at their highest values at the formation of the earth and decrease slowly with time up to the present. Mass and enrichment were discussed above and are important factors in

nuclear criticality. Moderation would be necessary in any system that is leaching lead from the other materials, but would also be a positive reactivity addition. There are many materials which could be neutron absorbers. However, it is unlikely that the materials which are absorbers would be consistent across all materials and formations. Also, it seems that if the uranium were imbedded in essentially neutron poisons, that that effect would carry forward in some level into the uranium processing and would be detrimental to the overall process.

For this analysis, the focus was on 100MT uranium at concentration of 20%. For many uranium mines, this would be well beyond a bounding concentration. However, the largest mines have significantly more mass than this (>580,000 MT in one uranium deposition compared to 100 MT modeled). Given this model is bounded by nature, it was determined that geometry and volume are insignificant criticality parameters.

MCNP Calculations and Analysis

In this nuclear criticality review, The LANL Monte Carlo N-Particle transport code MCNP was used to evaluate the reactivity of certain systems. The variables of this review were based on mass, enrichment, concentration and moderation. The driving change in this review was time as it impacted enrichment. Mass values of 1MT, 10 MT, and 100 MT uranium were modeled. The primary factors in evaluating criticality potential in that evaluation is the mass, enrichment, and moderation of the sample. Given that the concentration of the uranium mines report 20% concentration and hundreds of thousands of MT, this is still a low percentage of some of the larger mines. The reactivity values at 4.55 Ba is well in excess of a critical mass due to the uranium mass, concentration, moderation, and enrichment.

Summary

There has only been one naturally occurring reactor (Oklo) identified historically, there has to have been other factors that prevented uncontrolled nuclear criticality events. The concept of a naturally occurring reactor was originally documented (Kuroda) in 1956 using reactor theory or the infinite multiplication constants. The Oklo reactor was discovered 1972 following a routing check on enrichment by a uranium worker. Many other papers have evaluated the Oklo phenomena and addressed issues to identify what caused that uranium deposit to go critical. Consideration of the lack of lead as a potential indicator of the age of the earth as being a possible factor. Also, the higher concentration uranium deposits (>15%) were discovered in the 1990s, and reevaluation of the overall effect on a natural criticality were not considered.

The high reactivity levels and the low quantity of radiogenic lead identified in uranium tailings, tends to favor a significantly shorter time period or a highly efficient naturally occurring leaching process. A shorter time period would reduce uranium mass, enrichment. Given even a small quantity of moderator would allow an uncontrolled nuclear criticality for high concentration uranium deposits for enrichment between 3 and 8 percent ^{235}U.

The evaluation and analysis of the nuclear criticality safety factors should be evaluated further to document the actual uranium ore grade, and Pb constituents. Identification of the quantity (PPM) of radiogenic lead couples with the NCS factors could be a more useful tool for determining the age of the earth

Further calculations could be considered to determine the impact of different rock formations and materials where uranium is located. An evaluation of the natural leaching of uranium and its decay by-products to associate the effect of radiogenic lead or other materials.

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