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Criticality Safety Problems Related to Storage of Highly Active Liquid Waste

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ABSTRACT

The geometries of liquid waste storage tanks are not generally safe against criticality. Normally, this does not cause problems as fissile materials exist in nitric acid solution only as depleted uranium or in insignificant concentration of the originally reprocessed inventory of plutonium. However, if sedimentation of solid particles would occur, the deposited material would cause criticality safety problems. Particularly, non -horizontal installation of the storage tanks would increase the Eigen value. The effect of the storage tank inclination and the presence of transplutonium elements on the criticality safety are investigated using the NCNSRC code packages. The results are compared well with a similar German published results.

Key Words: High radwaste / Criticality safety/ Waste Storage/

INTRODUCTION

During the past years, large number of experimental and theoretical work had been performed to improve the basis for establishing criticality control on high level radioactive waste to help ensure freedom from nuclear accidents in chemical or metallurgical operations or on storage locations. Factors affecting criticality have been studied, with one of the principal purposes being to establish accurate critical sizes and masses over a range of significant parameters from experiments in selected assemblies. As it is not always feasible to establish experimentally all the data points that one might wish to obtain, calculations which are correlated to experiments , establish more firmly the critical dimensions and masses between measured points.

Moreover, calculations are used to assess criticality conditions for cases with specific conditions and parameters.
In the national center of nuclear safety and radiation control NCNSRC, criticality safety for transported or stored fuel were subject to a number of studies [1-9]. Most of these studies were concerned with more or less reactor fuel for research or MTR reactors type. With growing interest in fissile materials in laboratories, the previous criticality safety studies are extended in the present study to cover high level waste not necessarily research reactor waste.

There are many factors to be considered in determining safe operational limits for chemical and metallurgical handling fissile material . Process variables, control philosophy , environmental conditions , interaction effects, etc. must be taken into account; but all of these considerations, to a greater or lesser extent, are founded on the criticality parameters of a single homogeneous units.

The basic unit of all criticality data is the isolated spherical core consisting of an idealized mixture of a single fissile isotope and moderator, usually water. In such a system the number of variables is reduced to a minimum and thus a common ground is established for the comparison of the critical parameters of one particular isotope with those of another. Further this simple system would be the basis against which the effect of additions of other materials can be measured.

Following this approach, in the present work such a simple system is adopted .
The different cases studied using one dimensional spherical geometry are:
1- Different uranium enrichment;
2- The uranium concentration in UO2-water mixtures;
3- Plutonium 240 concentration in Pu metal water mixtures
4- Critical systems of plutonium nitrate solutions
5- The concentration of nitric acid in Pu nitric acid solutions
6- The presence of neutron poisons in the form of metals.
However in order to study the effect of storage tank inclination on criticality three dimensional geometry were adopted.

CALCULATION PROCEDURE

The first step in criticality calculation is to generate cross sections suitable for the type of calculation in question. For the purpose of the present calculation the cell parameter code WIMSD4 [12] was used to generate multigroup cross section set for each case listed above. It is well known that even the multigroup microscopic cross section well be affected by the spectrum variation as a result of varying the fissionable material density or fissionable material to hydrogen atomic ratio or to the presence of neutron poisons. As a result one has to generate a new cross section set for each value of the above parameters. The cross section has then to be transferred to ANISN [13] or TRITON [14] format using the routines in the NCNSRC codes library [10].

The code ANISN is then used for all spherical system calculations, While the three dimensional diffusion code TRITON is used to calculate the problem of tank inclination. Both search option (critical radius, zone thickness search) and straight forward criticality calculation are used in ANISN calculation.

CALCULATION AND RESULTS

Criticality calculations for water reflected spheres of UO2 water mixtures for two different enrichments (5% and 10%). In this calculation the total uranium concentration was varied in the range .1 - 10 g/cm3 . The results are shown in fig. 1.
The minimum critical mass of the total uranium was found to be 8.6 kg for concentration 1g/cc and enrichment 10%. While the minimum critical mass in case of 5% enrichment was found to be 45 kg total uranium for concentration .94% g/cc.

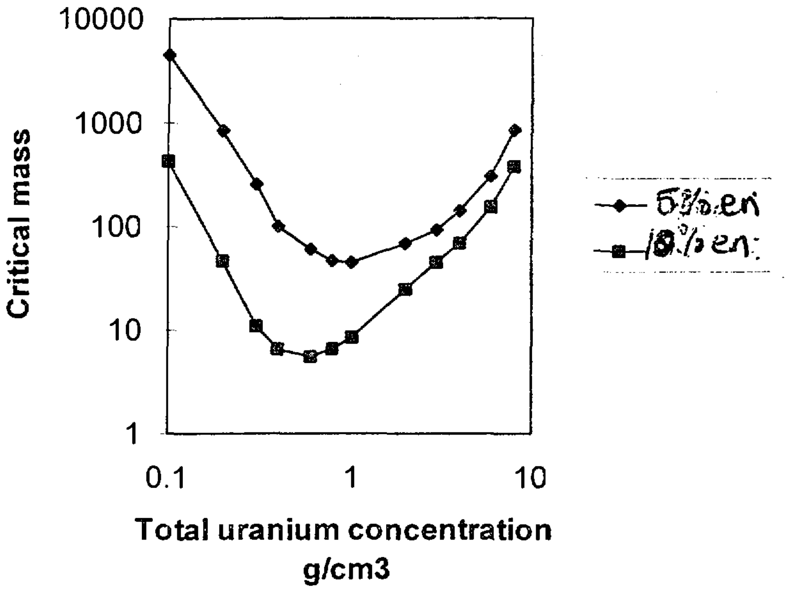

Figure 1: Critical mass of water reflected spheres of 5% and 10% enriched UO2 H2O mixtures

The above results were compared with earlier experimental and theoretical calculation by Chamles [13] .In Chamles calculation the four groups diffusion theory were used. The difference between the present and the earlier results amounts to less than 1% in most cases.

Plutonium systems

The criticality calculation results for plutonium metal water systems are shown in fig.2. The calculation showed that the minimum critical mass of Plutonium in the system is 0.57 which is in good agreement with earlier calculation performed using the SCALE system using 218 multigroup cross section library [14].

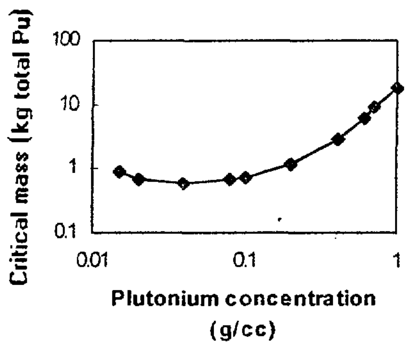

Figure2 : Critical mass of water reflected spheres of plutonium metal water mixtures

As the plutonium waste is usually stored in nitric acid solution , the criticality calculations were performed for plutonium nitrate solution with different plutonium concentration. The results are shown in fig .3. In this case the minimum critical mass is calculated to be .62kg of plutonium.

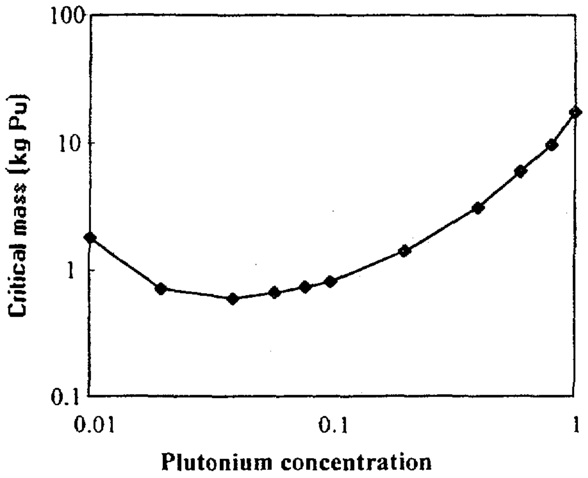

Figure 3 : Critical mass of water reflected spheres of plutonium nitrate solutions.

The effect of the presence of higher plutonium isotopes in the solution on criticality is shown in fig.4. In this figure the presence of Pu240 in the solution were varied between 0 to 15%. As it is shown from this figure a marked variation in the critical radii were found as the pu240 concentration increases as this particular plutonium isotope has a large neutron absorption cross section.

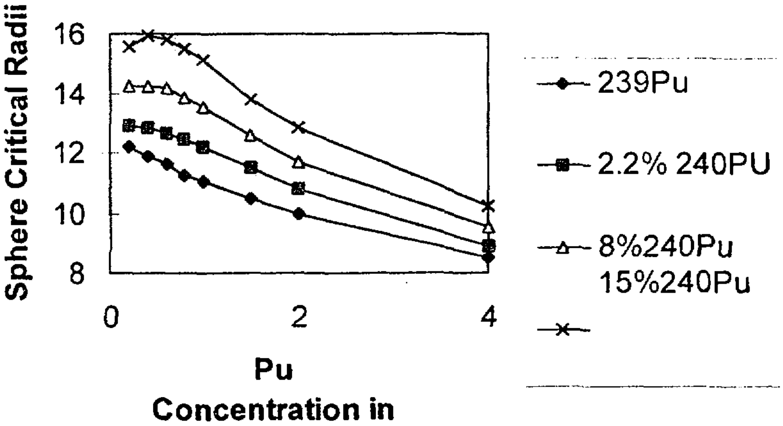

Figure4: Calculated critical radii of reflected spheres of Pu-water mixtures with Pu 240 up to 15 wt%.

The effect of nitric acid concentration on criticality is shown in fig.5 .This figure represents the variation of the critical mass for hydrogen to plutonium atomic ratio ranging from 100 up to 1000 and nitrogen to plutonium atomic ratio ranging from 0 up to 70.

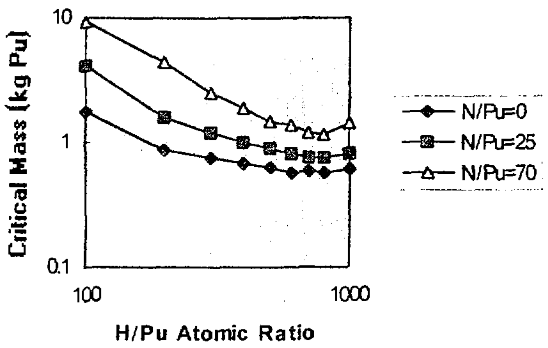

Figure 5: Critical masses of water reflected spheres of plutonium metal nitric acid mixtures

The results of plutonium system calculations agree within <2% with earlier results by Chamles [17] and by Claton et.el. [18].

Tank Installation

Highly active liquid waste is generally stored in a large tanks whose geometry's are not safe against criticality, because fissile materials exist in the nitric acid solution only as depleted uranium (<1% U-235) or in insignificant concentrations (< 300mg Pu per liter; < 1% of the originally reprocessed inventory of uranium, < 0.3% of the originally reprocessed inventory of uranium). Since the concentrations are below critical data, there are no criticality problems when the distribution of the fissile materials is homogeneous. But often there is a phase of solid material in homogeneous dispersion which contains plutonium (oxide) compounds in low concentration. This phase may form a sedimentary layer on the bottom of the tank and thus causing criticality hazard. In a recent German study [15], the influence of non-horizontal tank installation of the waste tank and of transuranium and transplutonium nuclides on reactivity of sediments containing plutonium .

Typically the tank is a lying cylinder with an outer diameter of 400 cm and a length of 562 cm. The thickness of the stainless steel wall is 1.2 cm. The computation model makes the following assumptions:

The uranium nitrate solution has a concentration of 7.4g/l (1%U235)
The chemical compound of the fissile particles in this solution is unknown
All fissile materials except U-235 precipitate on the bottom of the tank
Plutonium 239 mass in the solution is 11 kg, present as oxide. A water reflector of 30 cm thickness surrounds the cylinder.

In the German study the Monte Carlo code MORSE-K were used. In the present work the three dimensional diffusion code TRITON was used.

The problem of the tank inclination is treated as variation of non uniform precipitate material on the tank bottom. Both the present and the German results are given in fig.

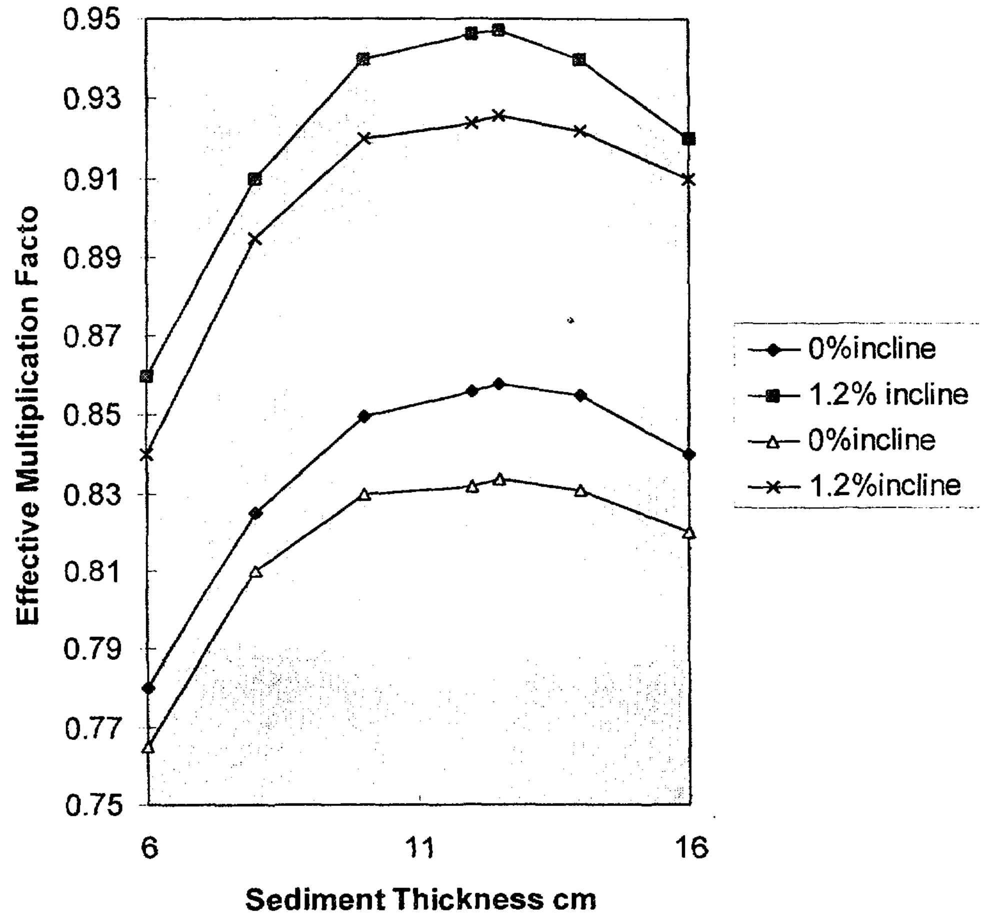

Figure 6: Effective multiplication factor as function of plutonium sedimentation thickness and tank inclination

The figure shows reasonable agreement with the German results. For inclined tank geometry the results worse but still good .

The presence of neutron poisons

Many of the wastes from processing fissile materials contain metals that serve as neutron poisons. It would be advantageous to the criticality evaluation of these waste to demonstrate that the poisons remain with the fissile materials and to demonstrate an always safe poisons-to-fissile ratio. Demonstrating that the materials may stay is the job of the chemists while calculating an always safe ratio is one of the objects of this paper. In earlier work by Williamson et. el.[16] safe ratio for iron manganese, and chromium oxides to different sub-critical arrangements of uranium wastes have been calculated using a Monte Carlo Code

HRXN with the Hansen-Roach 16 group cross section library . In Williamson work multiplication factors were computed, and safe ratios were defined such that the neutron multiplication values (k values) were < 0.95.
This safe ratio were found to be very sensitive to metal cross-section data in the intermediate energy range and the processing methods that are used. In the present work this safe ratio problem is studied using more elaborate cross section library and a sophisticated criticality code.
In the present work a detailed neutron cross section library based on ENDFB VI [17] was used. The library which contains 171 neutron energy group were used in the ANISN calculation.
Table 1 gives comparison for the safe ratios of different metals to uranium using the present cross section library and earlier data. The safe ratio is defined to be the ratio that gives a multiplication factor $< .95$.

Table 1 : Safe weight ratios for different metals in uranium assemblies.

Element	Earlier calculated ratio	Present calculated ratio
Fe : sup2 sup 3 sup 5 U	77:1	68:1
Mn : sup 2 sup 3 sup 5 U	30:1	34:1
Cr: sup 2 sup 3 sup 5 U	52:1	47:1

SUMMARY AND CONCLUSIONS

Criticality safety is the major concern in radioactive waste. In the present work some the NCNSRC codes are used for parameteric study for selected cases containing uranium or plutonium as fissile material. The one dimensional Sn code ANISN was found quite suitable to study problems of one dimensional nature. The minimum critical masses for reflected uranium water spheres at different u235 enrichement
,for plutonium water spheres and for plutonium nitrate solutions were determined.
All the results were found to be in good agreement with earlier results. The effect of nitric acid concentration and the presence of the plutonium isotope 240 were shown

The problem such as storage tank inclination had to be treated in three dimensional. The diffusion simulation of such problem led to reasonable results however this has to be treated using Monte Carlo technique to allow for accurate representation of the problem. The problem of the presence of metal poisons were found to be very sensitive to the cross section used. A detailed cross section set allowing for detailed description of the neutron resonance and epithermal ranges were used. The present calculation showed different results than earlier results [16]. The present result are considered to be more accurate because off the detailed update cross section library use.

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