CRITICALITY CALCULATIONS FOR A TYPICAL NUCLEAR FUEL FABRICATION PLANT WITH LOW ENRICHED URANIUM

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ABSTRACT

The operations with the fissile materials such as U^{235} introduce the risk of a criticality accident that may be lethal to nearby personnel and can lead the facility to shutdown. Therefore, the prevention of a nuclear criticality accident should play a major role in the design of a nuclear facility. The objectives of criticality safety are to prevent a self-sustained nuclear chain reaction and to minimize the consequences. Sixty criticality accidents were occurred in the world. These are accidents divided into two categories, 22 accidents occurred in process facilities and 38 accidents occurred during critical experiments or operations with research reactor. About 21 criticality accidents including Japan Nuclear Fuel Conversion Co. (JCO) accident took place with fuel solution or slurry and only one accident occurred with metal fuel.

In this study the nuclear criticality calculations have been performed for a typical nuclear fuel fabrication plant producing nuclear fuel elements for nuclear research reactors with low enriched uranium up to 20 %. The calculations were performed for both normal and abnormal operation conditions. The effective multiplication factor (k_{eff}) during the nuclear fuel fabrication process (Uranium hexafluoride – Ammonium Diuranate conversion process) was determined. Several accident scenarios were postulated and the criticalities of these accidents were evaluated. The computer code MCNP-4B which based on Monte Carlo method was used to calculate neutron multiplication factor. The criticality calculations were performed for the cases of, change of moderator to fuel ratio, solution density and concentration of the solute in order to prevent or mitigate criticality accidents during the nuclear fuel fabrication process. The calculation results are analyzed and discussed.

1. Introduction

Nuclear facilities and activities containing fissile material or in which fissile material is handled are required to be managed in such a way as to ensure criticality safety in normal operation, anticipated operational occurrences and during and after design basis accidents. This requirement applies to large commercial facilities, such as nuclear facilities that deal with the supply of fresh fuel, with the management of spent fuel and with radioactive waste containing fissile nuclides, including the handling, processing, use, storage and disposal of such waste. This requirement also applies to research and development facilities and activities that use fissile material and to the transport of packages containing fissile materials.

The subcriticality of a system depends on many parameters relating to the fissile material, including its mass, concentration, geometry, volume, enrichment and density. Subcriticality is also affected by the presence of other materials, such as moderators, absorbers and reflectors. Subcriticality can be ensured through the control of an individual parameter or a combination of parameters, for example, by limiting mass or by limiting both mass and moderation. Such parameters can be controlled by engineered and/or administrative measures [1].

The objectives of criticality safety are to prevent a self-sustained nuclear chain reaction and to minimize the consequences of this if it were to occur. The criticality safety analysis should be used to identify hazards, both internal and external, and to determine the radiological consequences. The criticality safety calculations makes recommendations on how to ensure sub-criticality in systems involving fissile materials during normal operation, anticipated operational occurrences, and, in the case of accident conditions [2].
Fuel cycle facilities may be split into two groups: facilities where a criticality hazard is not credible, e.g. mining, milling and conversion of natural uranium facilities; and those where the criticality hazards may be credible e.g. enrichment, uranium and mixed oxide fuel fabrication, fresh fuel storage (and transportation), spent fuel storage (and transportation), reprocessing, waste treatment facilities and disposal facilities. Facilities in this second group are designed and operated in a manner that ensures subcriticality in all areas and in operational states and design basis accidents. The facilities are operated in a manner that ensures that excessive amounts of fissionable material do not accumulate above specified limits in vessels, transfer pipes, ventilation ducts, ancillary equipment and other parts of the facilities.
Sixty criticality accidents were occurred in the world. These are divided into two categories, 22 accidents those that occurred in process facilities, and 38 accidents those that occurred during critical experiments or operations with research reactor. Of the 22 criticality accidents to have occurred in fuel processing facilities reported in Ref. [3, 4].
Process facilities carrying out operations with fissile material avoid criticality accident through physical and administrative controls. These controls are intended to prevent critical or near-critical configurations from ever occurring in the facility.
Nuclear criticality safety is achieved by controlling one or more parameters after system within critical limits. The factors affecting the criticality are: (1) mass of material (uranium), (2) enrichment, (3) volume of the material, (4) shape, (5) size, (6) concentration of fissionable material in solutions, (7) moderation, (8) reflection, (9) interaction, and (10) poisons. The first five factors are very important for control purposes in design equipment's where the uranium enriched are handled. For design purpose however, the moderation and reflection are usually assumed as optimum for criticality [2].
The plant to produce nuclear fuel element for research reactors has been operated with enriched uranium up to 20% enrichment. In the design of equipment, the type of criticality calculation and control to be used must be considered. Once the control method is established, the process and equipment must be designed to meet the limiting values of the control method used. Basically, the plants may be designed using mass, concentration, volume, geometry control and combination.
In this study the nuclear criticality calculations have been performed for a typical nuclear fuel fabrication plant producing nuclear fuel elements for nuclear research reactors with low enriched uranium up to 20 %. The calculations were performed for both normal and abnormal operation conditions. The effective multiplication factor (k_{eff}) during the nuclear fuel fabrication process (Uranium hexafluoride – Ammonium Diuranate conversion process) was determined. Several accident scenarios were postulated and the criticalities of these accidents were evaluated. The computer code MCNP-4B which based on Monte Carlo method was used to calculate neutron multiplication factor. The criticality calculations were performed for the cases of, change of moderator to fuel ratio, solution density and concentration of the solute in order to prevent or mitigate criticality accidents during the nuclear fuel fabrication process. The calculation results are analyzed and discussed.

2. Factors Affecting Criticality Limits in Abnormal Conditions

The factors affecting the criticality limit can be summarized as,
- A change in intended shape or dimensions resulting from bulging, corrosion, or bursting of a container, or failure to meet specifications in fabrication.
- An increase in the mass of fissionable material in location as the result of operational error, improper labeling, equipment failure, or failure of analytical techniques.
- A change in ratio of moderator to fissionable material resulting from:
a- inaccuracies in instruments or chemical analyses

b- evaporating or displacing moderator,
c- precipitating fissionable material from solution,
d- diluting concentrated solutions with additional with additional moderator.
- A change in the fraction of the neutron population lost by absorption resulting from:
a- loss of solid absorber by corrosion or by leaching
b- loss of moderator
c- redistribution of absorber and fissionable material by precipitation of one but not the other from solution
d- redistribution of solid absorber within a matrix of moderator or solution by clumping.
- A change in the amount of neutron reflection resulting from:
a- an increase in reflector thickness by adding water
b- a change in reflector composition such as loss of absorber[1].

3. General Process Description

3.1 Wet Process (UF_6-ADU Conversion Process)

In the wet process or ammonium diuranate (ADU) process, the UF_6 with enriched Uranium is 20% is vaporized and transferred to reaction vessel, hydrolyzed with water, and neutralized with NH_4OH to form a slurry of ADU in an aqueous solution of ammonium fluoride and ammonium hydroxide. The ADU is recovered by centrifuging and then is clarified, dried, and calcined to form UO_2 or U_3O_8 powder. Figures (1,2) is a flow diagram and sheet for the conversion process to convert UF_6 to ADU.
The 4 steps of process are:
◙ Vaporization process – conversion of a UF_6 solid into a gaseous state by adding heat for UF_6 Cylinder.
◙ Hydrolysis process – a chemical process by which the oxygen or hydrogen in water combines with an element, or some element of a compound, to form a new compound.
◙ Precipitation – formation of finely divided solids in a chemical reaction.
◙ Separation – remove or separate solid particles ADU from the liquid effluent.

a- Vaporization process

The UF_6 (a solid at room temperature) with enriched Uranium 20% is vaporized by adding heat. UF_6 is a colorless, volatile crystal that sublimes (changes directly from solid to vapor phase) at atmospheric pressure and approximately 86 °C. Under pressure UF_6 will be in the liquid state. An electric blanket is used to heat the UF_6 cylinder.

b- Hydrolysis process

After vaporization, gaseous UF_6 transferred to the Hydrolyser tank to react with deimineralzed water to form uranyl fluoride (UO_2F_2) solution and hydrofluoric acid (HF) as in the equation below. The hydrolyser is a container made of stainless steel with polypropylene lining. It has an internal plastic (polypropylene or polyethylene) cylinder. The chemical process of the hydrolysis occurs in the hydrolyser tank according the reaction:

$$UF_6\,(vapor) + 2H_2O\,(liquid) \rightarrow UO_2F_2\,(solution) + 4HF\,(solution)$$

The hydrolyser tank (35 cm in diameter and 55 cm in height) contains a mass of demineralized water about 40 liters which allows hydrolyzing a maximum of 2.7 kg of UF_6 which provides a solution of UO_2F_2 with a uranium concentration no more of 50 gram/liter. The solution is corrosive, thus the hydrolyser tank must be internally covered with polypropylene. The reaction occurs at ambient temperature. The temperature is increase due to the chemical reaction is 11°C. The maximum working pressure is, according to experience is 2 bar. The temperature and pressure inside the hydrolyser tank is controlled permanently. When the reaction is finished, quantitative analyses should be conducted in the laboratory to determine the final concentration of solution UO_2F_2 precisely.
A criticality accident may be generated, due to an unanticipated event causing; all of the UF_6 is transferred to the hydrolyser. To avoid that the following actions must be applied,

1- continuous weighing of the cylinder

2- continuous measuring of the mass
3- measuring of Uranium concentration in the hydrolyzed solution.

The hydrolyser and liquors boxes are covered with cadmium to prevent neutronic interaction. A 2 mm thick Cadmium sheet, with a plastic cover to prevent corrosion, is placed inside of the plastic internal cylinder. The cadmium make the hydrolyser tank is subcritical.
There are two recirculation pneumatic pumps ensure solution homogeneity during hydrolyses. While one pump is in operation, the other is in stand by these pumps are also used for transferring UO_2F_2 solution to the precipitator tank.

c- Precipitation process

The uranyl fluoride solution UO_2F_2 is pumped to a vertical container called precipitator tank (40 cm in diameter and 60 cm in height), where ammonium hydroxide (NH_4OH) is added to produce ADU crystals. This tank is made of stainless steel with elastomeric cover. To prevent settling of the solids in the tank, the contents have to be kept in suspension by recirculation.

Addition of NH_3 to the uranyl fluoride solution causes the precipitation of uranyl fluoride to ammonium diuranate [$(NH_4)2U_2O_7$], according to the following reaction:

$$2\ UO_2F_2 + 8\ HF + 14\ NH_3 + 3\ H_2O \longrightarrow (NH_4)2U_2O_7 + 12\ NH_4F$$

Depending on the specific process used at a facility, ammonium diuranate may also be precipitated by adding NH_4OH to the hydrolysis solution as follows:

$$2\ UO_2F_2 + 6\ NH_4OH \longrightarrow (NH_4)_2U_2O_7 + 4\ NH_4F + 3\ H_2O$$

$$8\ HF + 8\ NH_4OH \longrightarrow 8\ NH_4F + 8\ H_2O$$

d- Separation process

The precipitated ADU slurry is pumped from the bottom of the precipitation tank to a filtration tank (with a diameter of 50 cm and a volume of 70 liter) to concentrate the crystals by separating out the liquids. The filter tank is made of stainless steel. This process is typically accomplished by means of a centrifuge or filter press. The centrifuge uses rotational forces to separate the solid particles from the liquid, while a filter press uses mechanical force to push the liquids through a porous medium, leaving the solid particles behind. The liquid drained is stored in two liquid storage cylinders placed horizontally, one slightly above the other. The diameter of these cylinders is about of 17 cm and the length of about 180 cm.

4. Criticality safety of UF_6-ADU conversion process

The wet part of the installation has a limit in the Uranium mass, which 2.7 kg per a lot. It was set considering the possibility of duplicating the mass without criticality risk. To consider there is criticality risk it is necessary to have, at least:
1- A uranium mass larger than the minimum critical mass, which 5.6 kg total uranium with enrichment 20%.
2- Enough quantity of moderator substance to obtain the optimum moderation degree.
The operation procedures indicate before the transferring the solution into the precipitator, the concentration of the UO_2F_2 solution which in hydrolyser must be controlled. Once the concentration has been verified which lower than the subcritical concentration, the solution can be transferred to the precipitator tank by opening a valve.
A mass larger than the operational limit may enter the precipitator tank due to; transference from the hydrolyser a solution with more than one Uranium lot. Overload in the hydrolyser may be due to bad operation in the control of the mass entered. It may therefore occur due to the following simultaneous failures in the balance and/or operation, and the blocking which stops the UF_6 cylinder heating and closes valves transference when the balance detects 2.7kg UF_6 have gone through. This situation may occur once the hydrolyser has a load larger than the limit. For that load to be transferred, the following failures must occur:
- in measuring the concentration which enables solution transference.

- in the criticality prevention system, which must detect overload in the hydrolyser, triggering an alarm in the radioprotection switchboard.
Addition of a lot of solution into the precipitator without unloading the previous lot.
For this addition to occur, the solution concentration should be larger than the operation concentration, since the volume of two solution lots would exceed the precipitator volume. The uranium does not accumulate in the precipitator because the solution would be vented out, spilling first in the glove box and then in the collecting trays adopting a subcritical geometry.
The solution inlet the precipitator from the hydrolyser due to administrative error, procedure violation which prevents working simultaneously with the hydrolyser and the precipitator full. Failure in the criticality prevention system, which must detect the work simultaneously in two excluding units, triggering an alarm at the radioprotection office. This event does not imply criticality risk as more than two lots are necessary to exceed the value of the minimum critical mass.
The solution inlet the precipitator tank from the filtering tank. This event may occur under the following conditions; procedure violation, during filtering there must be no material in the precipitator. The criticality prevention system, which detects work in two excluding units, must fail.
If filtering is performed under the mentioned conditions the filtering mesh must break without the operator noticing it, the ADU batch goes into the tanks and when the mother liquor recirculate the material is transferred to the precipitator. Not only is occurrence of this event highly improbable, but also with double load in the precipitator tank an accident with criticality risk does not occur.
For a uranium mass exceeding the operation value to be inside the filter the following should occur:
Filtering without removing the previous lot from the filter. This would occur due to; repeated violation of several operation procedures (hydrolysis, precipitation and filtering .Failure in the criticality prevention system, which must detect the presence of material in excluding operations.
This events highly improbable, does not imply criticality risk, since more than two
lots are necessary to exceed the value of the minimum critical mass.

5. Criticality Detection and Alarm Systems

Criticality Accident Alarm Systems (CASs) are important for the rapid evacuation and the reduction of the operators' exposure.

A criticality detection and alarm system should be provided to minimize the total dose received by personnel from a criticality accident and to initiate mitigating actions.

The criticality detection and alarm system should be based on the detection of neutrons and/or gamma radiation. Consequently, consideration should be given to the deployment of detectors that are sensitive to gamma radiation neutrons, or both [5].

In areas in which criticality alarm coverage is necessary, means should be provided to detect excessive radiation doses or dose rates and to signal an evacuation of personnel.

The alarm signal should meet the following criteria:

-It should be unique, i.e. it should be immediately recognizable to personnel as a criticality alarm;

-It should actuate as soon as the criticality accident is detected and continue even if the radiation level falls below the alarm point until manually reset;

Systems to manually reset the alarm signal, with limited access, should be provided outside areas that require evacuation;

-It should be audible in all areas to be evacuated;

-It should continue to alarm for a time sufficient to allow a complete evacuation;

-It should be supplemented with visual signals in areas with high background noise.

To prevent the criticality accident happens in the nuclear fuel fabrication plant must be follow:
1. to follow the instruction written in the criticality card control in all devices/equipment's.
2. Avoiding the flood, spraying other material in water groups, oil, wood etc.
3. Avoiding the additional of reflector materials such as (graphite, beryllium etc.)
4. Avoiding the placement/the use of unit of equipment in the wrong place.
5. to check periodically the loss of neutron absorber (if any).
6. To remember the favorable critical mass for each process of equipment.
7. To remember the change of fissile material density during process production.
8. Not to change the unit of equipment that cause unsafe geometrically.

In addition, the plant is fully equipped with 5 criticality detector in strategic places to give an early warning to employees whenever the postulated mass criticality accident occurs. The signal alarm will active when the radiation exposure exits the limit 10 mR/hour over background.

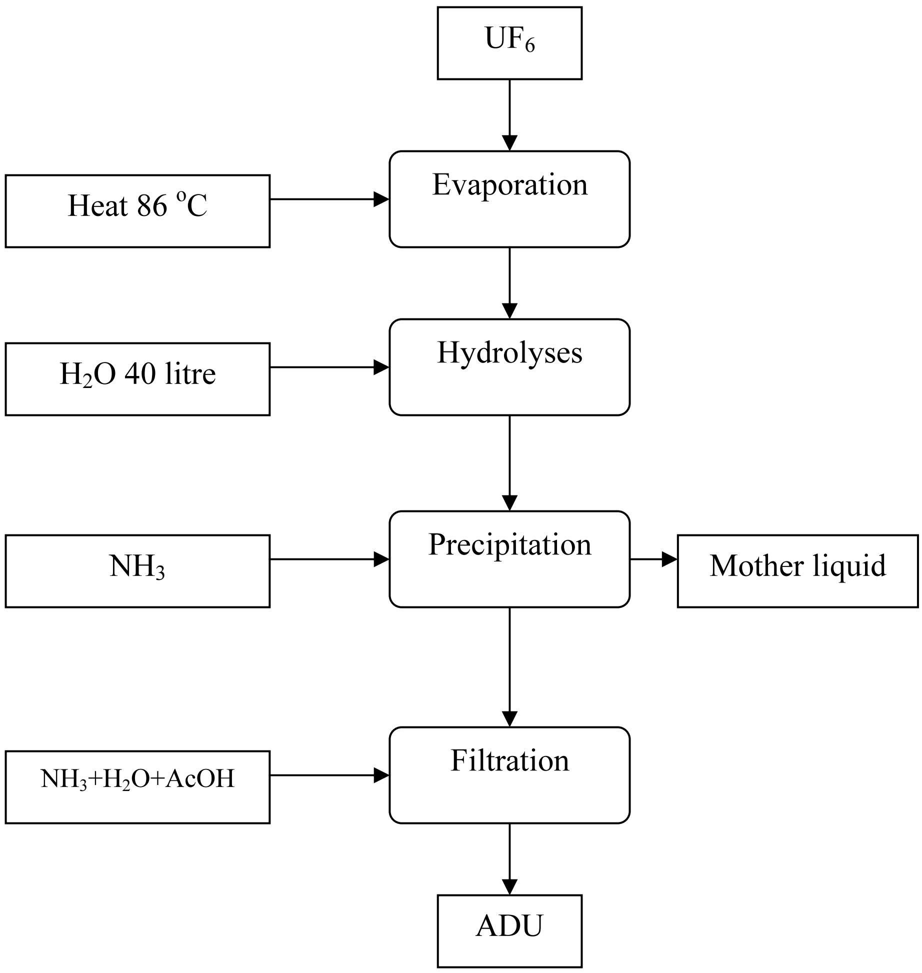

Fig 1. Flow diagram of UF_6 – ADU conversion process

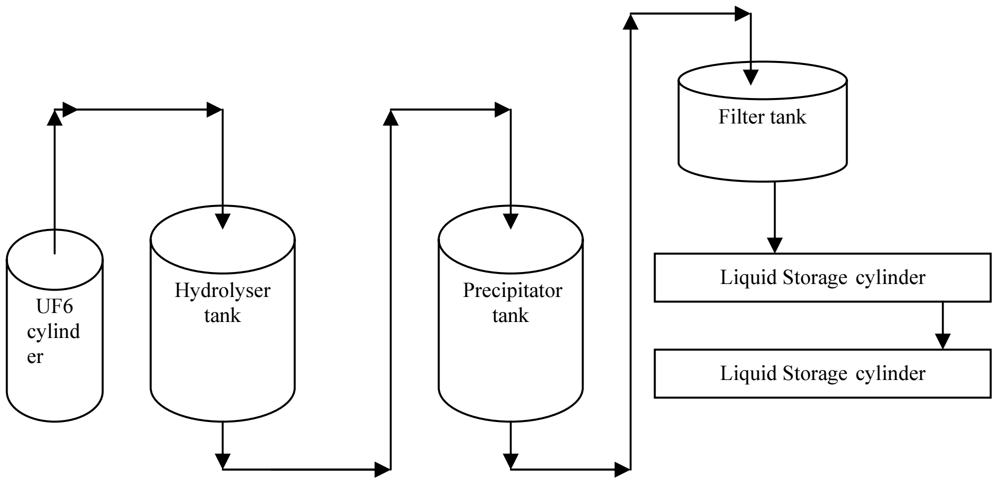

Fig 2. Process flow sheet UF_6-ADU Conversion Process

6. Calculation Methodology

MCNP is a general Monte Carlo N-Particle transport code. It has the capability of transporting neutrons, electrons and photons. In this work the transporting of neutrons is considered. These features make it possible for MCNP to simulate different types of fissile systems, e.g. a nuclear reactor or a nuclear fuel container, and obtain information such as k_{eff}. The neutron energy regime is from 10^{-11} MeV to 20 MeV, and the photon and electron energy regimes are from 1 keV to 1000 MeV. The capability to calculate k_{eff} eigenvalues for fissile systems is also a standard feature MCNP provides four types of boundary conditions that are accepted for performing criticality safety analyses. The default boundary condition is a vacuum where neutrons leaving enter a region of zero neutron importance [6].

7. Results and Discussion

7.1 Multiplication factor as a function of solution density in precipitator tank

Figure 3 and 4 shows the relation of the effective of multiplication factor as a function of solution density in the precipitator tank. This figure shows that an increase in the density assumed for the solution implies an increase in the system multiplication factor. It is therefore important to know the system behavior in case of a change in the solution density, under optimum moderation conditions.

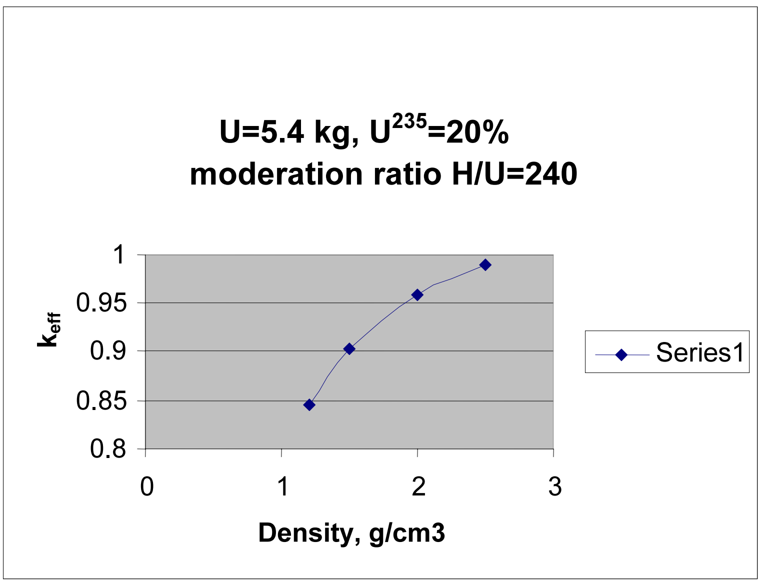

Fig 3. Multiplication factors as a function of the solution density in the precipitator tank.

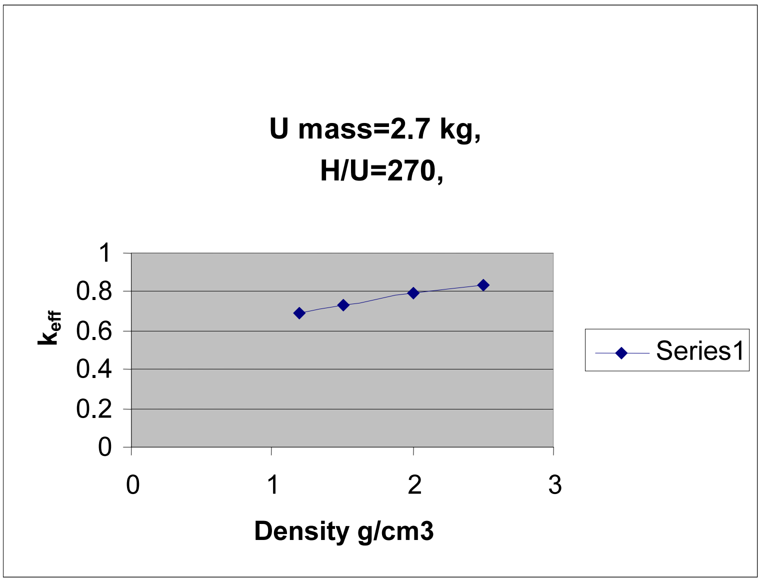

Fig 4. Multiplication factors as a function of solution density in the precipitator tank.

7.2 Multiplication factor as a function of the solution concentration and solution density in the hydrolyser tank

The effect of changing the solution concentration and density of UO_2F_2 on the effective of multiplication factor shows in figure 5 and 6. The system subcriticality in case of changes in solution concentration caused by variation of the starting quantity of water in the hydrolyser or variation of the UF_6 mass poured to hydrolyser tank.

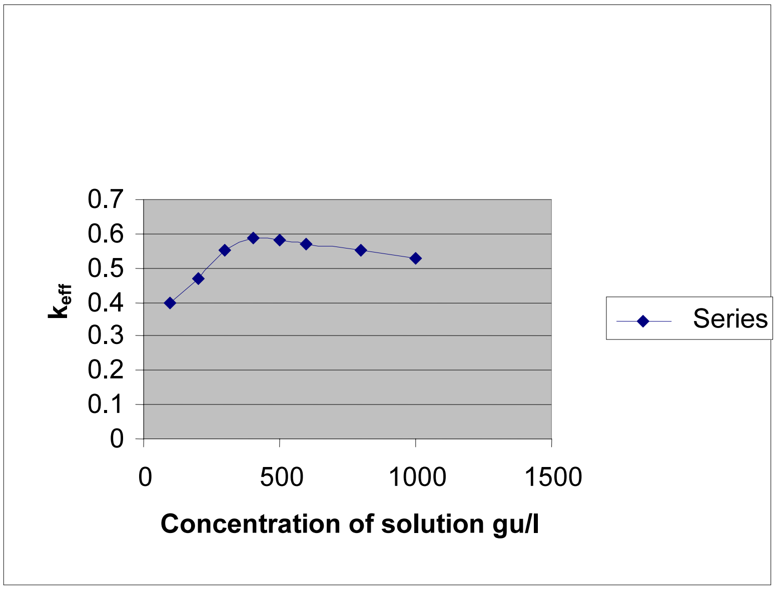

Fig. 5 Multiplication factor as a function of the solution concentration UO2F2 in the hydrolyser tank.

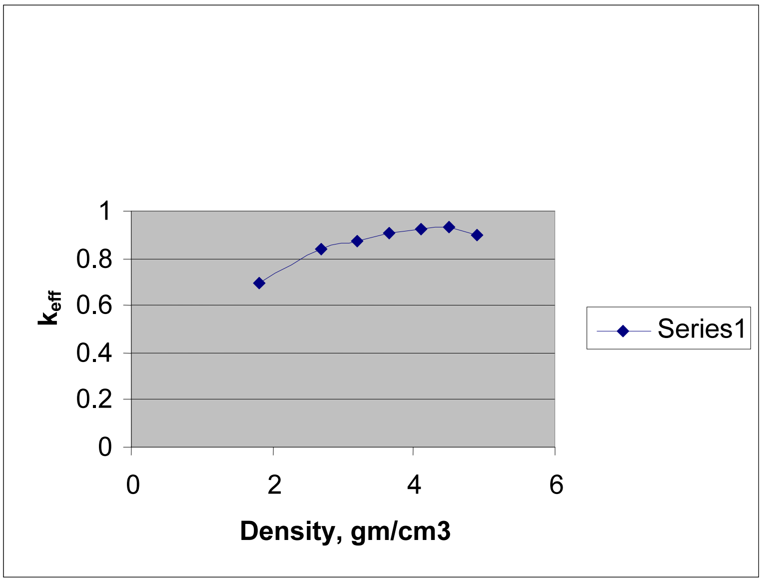

Fig 6. Multiplication factor as a function of the solution density in the hydrolyser tank.

7.3 Multiplication factor as a function of moderation ratio of ADU

Figure 7 and 8 shows the effective of multiplication factor as a function of moderation ratio of ADU. An increase the moderation ratio H/U increases the multiplication factor at certain moderation ratio and then will decrease.

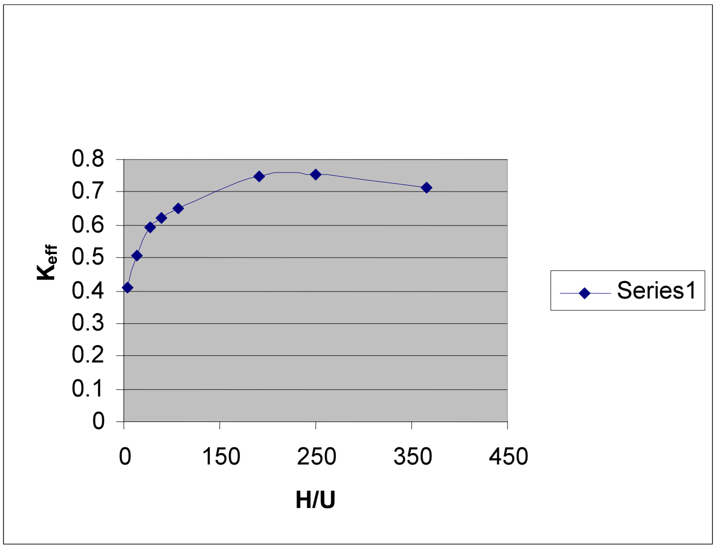

Fig 7. Multiplication factor as a function of the H/U for ADU, U=2.7 kg.

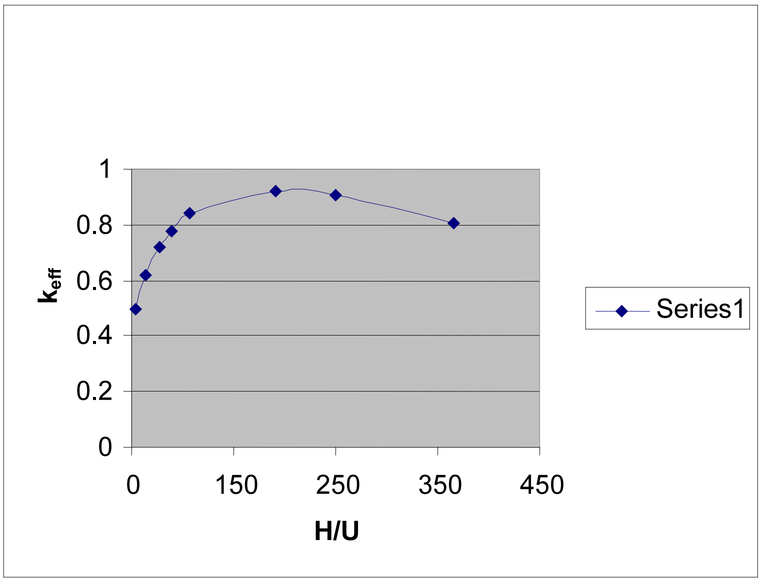

Fig 8. Multiplication factor as a function of the H/U for ADU, U=5.4 kg.

7.4 Multiplication factor as a function of moderation ratio in hydrolyser tank With cadmium sheet

We assume that all the masses of U=16.8 kg in the UF_6 cylinder transfer into the hydrolyser tank with hypotheses accident. Figure 9 shows the effective multiplication factor as a function of H/U with cadmium sheet inside the hydrolyser tank. The system will be subcritical in the case when the full UF_6 load is inadvertedly transferred from the UF_6 cylinder to the hydrolyser tank.

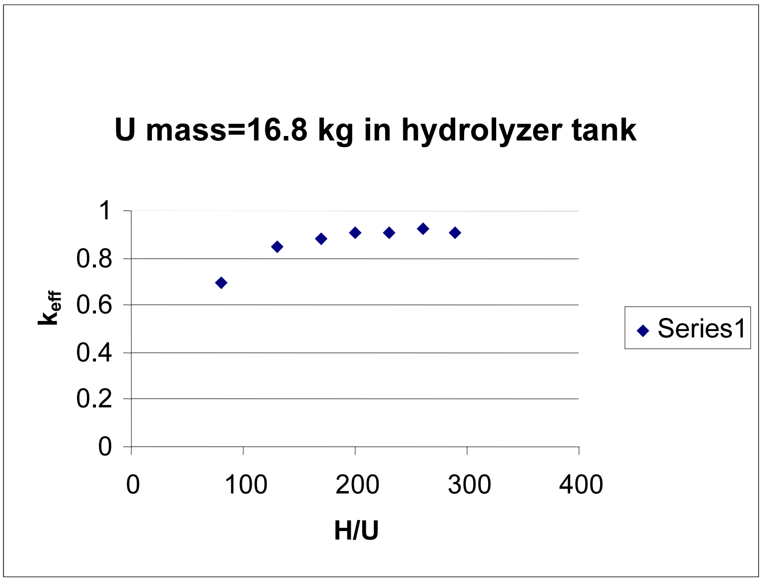

Fig 9. Multiplication factor as a function of the solution moderation degree H/U.

The safety of precipitation tank in case of criticality accident has been proved, assuming the uranium mass in process is effectively controlled and kept below 2.7 kg uranium per lot. Different events which cause a change in the moderation conditions for the solution produced in the precipitator tank have been analyzed , i.e. different starting water volumes in the hydrolyser tank, ADU precipitation to the vessel bottom, different volumes of ammonium solution added into the precipitator tank.

8. Conclusion

The criticality safety of a typical nuclear fuel fabrication plant is studied and analyzed with MCNP-4B code.

The multiplication factor was determined for UF_6-ADU conversion process for normal operation and abnormal conditions.

The results indicate for normal conditions the k_{eff} is subcritical values, while for abnormal conditions the multiplication factor may be critical.

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