Basis for criticality incident detection coverage for a typical MTR-type fuel elements fabrication plant

Cruzate, J.A.

Presentado en: International Conference on Nuclear Criticality Safety ICNC2015.
Charlotte, EE. UU., 13 al 17 de setiembre de 2015

BASIS FOR CRITICALITY INCIDENT DETECTION COVERAGE FOR A TYPICAL MTR-TYPE FUEL ELEMENTS FABRICATION PLANT

Cruzate, J.A.[1]

Nuclear Regulatory Authority
Argentina

ABSTRACT

The bases for the design, and nuclear criticality evaluation, of a typical facility for the manufacturing of fuel elements for material testing reactor (MTR) are given. A general-purpose process is presented, which starts with metallic uranium hexafluoride, and ends with the assembling of the final fuel element. Well-known international guides were used for establishing the main process parameters, including some important geometric dimensions.

The feasible design basis accidents were identified by analysing the defined representative process. The more probable accidents were found to happen during the chemical stage of the process. The criticality of the accidents were analysed, and the total number of fissions for these accidents were estimated using a simplified formula based on an improved heat energy method. As the assumed accidents take place in moderated media, the radiation spectra for neutrons and photons arising from thermal fission of ^{235}U were estimated in order to assess the doses received by workers. The whole facility was simulated, with the estimated primary radiation source placed at assumed accidents locations, and a dose mapping was made covering the entire facility. All the calculations were made by using the MCNP6.1 Monte Carlo code, adopting cross sections from the ENDF/B-VII.1 nuclear data.

The obtained dose mapping was used to define the composition and dimensions of walls and doors within the facility. On the other hand, according to ISO 7753 International Standard, a criticality accident alarm system (CAAS) is not required in areas where the maximum foreseeable accidental dose in free air will not exceed 0.12 Gy. Then, the obtained mapping may be used as an aid for determining the zones of the facility to be covered by CAAS, for stating the evacuation zone boundary and the evacuation routes, and to design the emergency plan. Moreover, this methodology may be adapted for deciding the feasible locations for criticality alarms.

Keywords: Criticality evaluations, Criticality accidents, dose mapping, MCNP6.1, ENDF/B-VII.1

1. INTRODUCTION

Nuclear facilities plants, where fissionable material is processed, handled, stored or used in any physical or chemical form, give rise to the risk of a criticality accident that may be lethal to nearby personnel. In addition, a criticality accident could bring out the release of nuclear material to the environment with the subsequent exposure of members of the public. Consequently, for these facilities nuclear criticality safety must be assured in normal operations, anticipated operational occurrences, and during and after design basis accidents. This does not apply to facilities where criticality is intended by design, as for example nuclear reactors and critical facilities. This also does not apply to severe or beyond design basis accidents, but in these cases countermeasures must be taken into account in order to mitigate the consequences of the accident. With this aim, a criticality safety analysis must be accomplished in order to

[1] jcruzate@arn.gob.ar

identify all possible criticality hazards, and its radiological consequences. On the grounds of this analysis, a criticality safety evaluation must be carried out to determine the parameters which assure nuclear criticality safety. The basic parameters that assure that a system will be subcritical under specified conditions may be drawn from international standards [1]. These parameters may used as a starting point of the criticality evaluation.

Nuclear facilities, in particular those related to fuel cycle, can be categorised in facilities where a criticality accident may be not credible, and facilities where a criticality accident may be credible. Good examples of the last ones are enrichment, fuel fabrication, spent fuel storage, reprocessing and waste treatment facilities. For facilities in the second group, ISO 7753 states that a criticality accident alarm system (CAAS) is not required in areas where the maximum foreseeable accidental dose in free air will not exceed 0.12 Gy [2].

This work is focused on the process to manufacture fuel elements for material testing reactor (MTR). The starting material for MTR fuel fabrication is UF_6 in solid state. The UF_6 is first heated over 80°C in order to get it in gaseous state and then is added to demineralised water, and so a uranyl fluoride (UO_2F_2) solution results. Later, ammonium hydroxide is added to the uranyl fluoride (UF) solution, producing a suspension of ammonium diuranate ($(NH4)_2U_2O_7$). Subsequently the ammonium diuranate (ADU) is filtrated, washed with ammonia solution and dried with alcohol. This ADU is calcined at 800°C in an oxidant atmosphere to form a U_3O_8 agglomerate, which is then milled and sieved. Subsequently this powder is heated up to 1400°C, and milled and sieved again in order to obtain a high density U_3O_8 powder. This powder, mixed with aluminium oxide, is compacted and framed with aluminium elements. These frames are rolled to obtain the fuel plates, which are finally arranged to form the final fuel element.

In this work the bases for the design, and nuclear criticality evaluation, of a typical MTR-type fuel element fabrication plant are presented. In addition, the feasible criticality accidents have been identified and their source terms were calculated. On the grounds of the determined possible source terms a dose mapping has been established for the postulated accidents. The obtained dose mapping was used to define the composition and dimensions of walls and doors within the facility. Furthermore, the obtained mapping may be used for determining the necessity of CAAS, and the zones of the facility to be covered by this. In addition, this mapping may be also used as an aid for stating the evacuation zone boundary and the evacuation routes, and to design the emergency plan. Moreover, this methodology may be adapted for deciding the feasible locations for the criticality alarms.

2. BASIS FOR THE DESIGN OF A TYPICAL PLANT, AND ASSOCIATED PROCESSES, FOR MTR-TYPE FUEL ELEMENT FABRICATION

The typical plant presented in this work is intended to produce generic fuel elements for MTR reactors, which use uranium enriched up to 20% in weight of ^{235}U. A general layout for this plant is presented in Figure 1. The plant has been conceived to produce fuel elements starting from solid UF_6. The only raw material, other than solid UF_6, that enter to the plant is aluminium powder, in addition to aluminium frame and covers. All the proposed activities require specific spaces, with constructive characteristics defined by the type of processes that take place in them. The present layout is intended to be generic, and so these specific characteristics were not taken into account. In this stage only process parameters will be determined. The walls, as well as floor and roof, are made of concrete, but its thickness will be determined in a subsequent stage, in the course of the evaluation of the doses generated during a criticality accident; the same applies to the thickness and composition of the doors which interconnect the different sections of the facility. In regards to fissile material handling, the plant is divided into the six sections shown at Figure 1. In addition, there is a chemical laboratory, not shown in the layout, where the necessary chemical measurements related to process are accomplished.

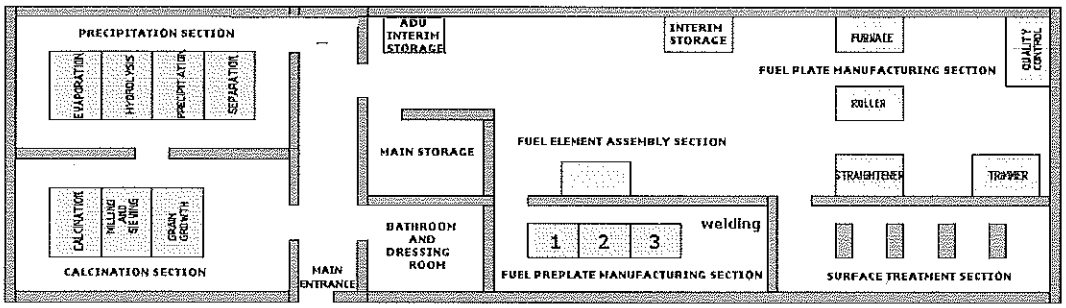

Figure 1. Layout of a typical plant for MTR-type fuel elements fabrication.

As far as fissile material handling is concerned, the precipitation, calcination, and fuel preplate manufacturing sections are the areas with the highest risk of contamination, associated to powder manipulation. Special care is therefore necessary regarding closing and ventilation. In addition, a special paint must be used for the floor and the walls, to make eventual decontamination easier. These three sections comprise the controlled area. The rest of the sections have less strict requirements and construction is therefore conventional and hence they, in addition to chemical laboratory, comprise the supervised area. All fissile material store rooms are inside the plant. Raw materials (other than UF_6), powders, scrap, and finished fuel elements are stored in the main storage. On the other hand, material in process is stored in the corresponding interim storage.

In order to assure the intrinsic subcriticality of the equipments and devices involved in the different stages of the process, it is helpful to take into account the subcritical parameters given by International Guides. In particular, the GUIDE DE CRITICITÉ issued by CEA [1] gives the following values, for uranium with 20% in weight enrichment:

a) Solutions and homogeneous salts:

Critical values: mass = 5.6 kg total uranium, volume = 12.9 l, diameter = 21 cm, thickness = 9 cm

Safe values: mass = 2.4 kg total uranium, volume = 9.6 l, diameter = 17.8 cm, thickness = 6.7 cm

b) Metal/water homogeneous mixtures:

Critical values: mass = 5.6 kg total uranium, volume = 10.5 l, diameter = 18 cm, thickness = 7 cm

Safe values: mass = 2.4 kg total uranium, volume = 7.9 l, diameter = 15 cm, thickness = 5.2 cm

c) Concentration: Critical value = 60 g/l, safe value = 51 g/l (expressed in total uranium).

Subcriticality may be assured if the mass involved in each section of the plant is limited by a batch system, and one batch is taken to be the safe mass, i.e. 2.4 kg of total uranium. The nominal production capacity will be 100 fuel elements per year, with a total uranium content of 2160 g each. A detailed description of each section, and related operations, is presented in the following.

2.1. Precipitation section: UF_6 to ADU conversion (wet process)

Here ammonium diuranate is obtained from solid UF_6. UF_6 reacts with air moisture forming a white fog which consist of hydrofluoric acid (HF) and uranyl fluoride (UO_2F_2). Both gases are toxic and corrosive, the last also being radioactive. In order to prevent spreading in case of UF_6 release, all the equipments in this sector are located into one glove box which is below

atmospheric pressure. The released gases are extracted through scrubbers spry towers, and highly efficient particle air (HEPA) filters. Furthermore, during hydrolysis and precipitation operations, liquids containing fissile material may leak from tanks, piping, pumps, and so on. In order to avoid criticality in these cases a stainless steel tray, 5.2 cm in height (safe thickness for metal/water homogeneous mixtures), is located in the bottom of the glove box. The process in this section is divided into four stages: vaporisation, hydrolysis, precipitation and separation.

2.1.1. Vaporisation

The UF_6 used in the process is contained in a cylinder model 5A, which contains no more than 24.95 kg of UF_6, which is equivalent to 16.85 kg of total uranium. UF_6 is solid at room temperature, therefore heat must be added in order to vaporise it before hydrolysis. The cylinder and pipes are then heated to about 80-100°C at normal atmospheric pressure.

2.1.2. Hydrolysis

Gaseous UF_6 is transferred to hydrolysation tank, where it reacts with demineralised water, according to the following reaction

$$UF_6(g) + 2\ H_2O \rightarrow UO_2F_2(aq) + 4\ HF(aq)$$

The hydrolysis tank is a closed right cylinder made of stainless steel. If it is assumed that all the uranium mass in the batch (2400 g) enters to the hydrolysis tank, this results in 47.1 litres of solution with 51 g/l of total uranium (safe concentration). If the diameter of the tank is assumed to be 17.8 cm (safe diameter for solutions), then the height of tank would be 189.1 cm. Such a tank would be too high for a practical process, so the following dimensions are proposed: diameter 35 cm and height 57 cm (54.8 litres in volume). In conclusion, during hydrolysis 2400 g of total uranium in the form of UF_6 are added to 47 litres of demineralised water, producing an aqueous solution with 51 g/l of total uranium, in the proposed tank. In this way, the hydrolysis tank is no safe by geometry, so mass transferred to it must be controlled by continuous weighing of UF_6 cylinder; in addition, uranium concentration in solution must be checked also. The chemical reaction occurs at room temperature, but the reaction itself increase the solution temperature. In consequence temperature and pressure inside the hydrolyser tank must be controlled permanently. In order to ensure homogeneity during hydrolyses, solution must be recirculated during operation. In order to prevent neutronic interaction, hydrolyser tank may be covered with a cadmium sheet, of about 2 mm in thickness. The produced solution is corrosive, hence this tank must be internally covered with polypropylene, polyethylene, or some other appropriate material.

2.1.3. Precipitation

Once the concentration of fissile solution at the hydrolysis tank has been verified to be equal to or less than 51 g/l, the solution can be transferred to the precipitation tank (or precipitator). When the solution transfer is complete, an ammonia solution (ammonium hydroxide) is added in order to produce $(NH4)_2U_2O$ (ammonium diuranate, or ADU) crystals, according to the following reaction

$$UO_2F_2 + 7\ NH_4OH + 4\ HF \rightarrow 1/2\ (NH_4)_2U_2O_7 + 6\ NH_4F + 11/2\ H_2O$$

The precipitator is proposed to be a closed right cylinder made of stainless steel with an elastomeric internal cover, and with the following dimensions: diameter 40 cm and height 56 cm (70.4 litres in volume). The precipitation tank is no safe by geometry, so uranium concentration in solution must continuously checked. In order to prevent neutronic interaction, this tank may be covered with a cadmium sheet, which thickness can be about 2 mm. ADU crystals are kept in suspension by means of recirculation, in order to prevent falling of the solids to the bottom of the tank. During process temperature and acidity (pH) of solution must be

permanently controlled, as well as ammonium hydroxide addition rate. In the final stage, an ADU slurry is precipitated to the bottom of the tank.

2.1.4. Separation

The precipitated ADU slurry, produced in the previous stage, is transferred from the bottom of precipitation tank to a filtration tank. This is a right cylinder, 40 cm in diameter and 56 cm in height, with a corresponding volume of 70.4 l, made of stainless steel. During this operation ADU crystals are separated from the liquid, typically by means of centrifuge or filter press. The centrifuge uses rotational forces to separate ADU crystals from the solution. On the other hand, filter press make use of mechanical force to push the solution through a porous material, withdrawing the solid particles behind it. After completing the filtering, the ADU precipitate is washed with ammonia solution, and then dried with alcohol. This ADU cake contains fluorine, which stoichiometry varies according to the specific process. The liquid drained is stored into two storage cylinders of safe diameter (17 cm) and 155 cm in height. Finally, the dried ADU is collected in leak-proof containers which are then stored in the ADU interim storage, from where it will be transferred to calcination section. These containers have a height of 5.2 cm (safe thickness for metal/water homogeneous mixtures), and its surfaces are separated by 50 cm or more.

2.2. Calcination section: U_3O_8 powder fabrication

The ADU cake obtained in the precipitation sector must be calcined, in an oxidant atmosphere, in order to obtain the U_3O_8 uranium oxide powder used for the fabrication of the MTR fuel element. In this sector dusty materials are handled, hence the operations are carried out in into three interconnected glove boxes, which are under depression, and the air extracted flows through HEPA filters. The ADU cake is loaded in alumina (Al_2O_3) open vessels, put into an electrical furnace, and heated in air up to 800°C.

During calcination NH_4F is released as well, so the internal surface of the furnace must be lined with a protective coat, typically inconel or similar alloys. In addition, the released gases must be scrubbed in a water spray column. U_3O_8 produced during calcination is a low density agglomerate. This agglomerate is then milled, and so bigger particles are broken, and smaller particles are agglomerated, in order to homogenise the sizes of calcined particles. Afterwards it is sieved in order to separate the smaller particles. In order to obtain the high density U_3O_8 powder required for fuel fabrication, the material is calcinated again, but this time the temperature is 1400°C. Subsequently the material is treated in a mortar, and sieved in order to separate the smaller particles. Afterwards, the powder is repeatedly milled and sieved up to obtain the required grain size distribution according to process specifications (typically particle size less than 90 μm). The final powder produced in the calcination section is stored in the facility's main storage. The storage unit is a polyethylene recipient, diameter 10 cm (lesser than the safe diameter for metal/water homogeneous mixtures) and height 10 cm, where 2400 g of total uranium (one batch) are stored. The separation between units surfaces will be 50 cm or more.

2.3. Fuel preplate manufacturing section

In this section, the fuel core compact is manufactured starting from a mixture of U_3O_8 and aluminium powders. Subsequently aluminium frame and covers are added, and the resulting fuel preplate is assembled and welded. In this sector dusty materials are handled, thus the operations are made within three interconnected glove boxes. These glove boxes are under depression, and the air extracted flows through HEPA filters. The total mass of the fuel core compact is 182 g, with following proposed dimensions and composition

dimension: $6.7 \times 6.07 \times 0.89$ cm^3 composition: 70% U_3O_8 + 30% Al

mass: 21.6 g of $^{235}U \rightarrow$ 108 g of total uranium$\rightarrow$ 127.4 g of U_3O_8, and 54.6 g Al

In the first stage the U_3O_8 powder, contained in the storage unit (containing one batch or 2400 g of total uranium) is transferred from main storage and entered in glove box 1. Afterwards the corresponding quantity of aluminium powder is also transferred. Within box 1 both powders are weighed and mixed, and the obtained powder is dosed and dropped into 20 appropriate independent vessels (right cylinders of 100 cm^3 in volume each), previously arranged in a metal frame. The frame with the 20 vessels is transferred from glove box 1 to glove box 2 where, one at the time, the vessel contents are dropped into an appropriate matrix, and the powder is pressed at a suitable pressure by means of a hydraulic press. The compacted core is then positioned in a suitable grid and the empty vessel is returned to its frame. Once that all the cores were compacted, the frame with the empty vessels is transferred back to box 1. The grid with the compacted cores is transferred to glove box 3, and later the aluminium frame and covers are entered. Afterwards all the elements are assembled together in order to shape the preplate. Finally the assembled preplate is taken out of glove boxes. Once out of glove boxes the assembled preplate, one at the time, are welded and then positioned in a frame carrier. When the twenty preplates that made the batch are positioned in the frame carrier, this frame is transferred to the fuel plate manufacturing section or to the interim storage.

2.4. Fuel plate manufacturing section

In this section, the fuel plate element is obtained from the preplate produced in the previous section. Taking into account the usual fuel elements utilised at MTR reactors, the following generic dimensions are proposed for the plate that will make up the produced fuel elements in the typical fabrication plant: fuel meat 70×6.43×0.08 cm^3, and external fuel plate 76×7.43×0.18 cm^3. Each fuel plate will consist of one fuel meat framed with some aluminium elements. The finished fuel element will consist of an assembly of 20 plates. The preplate is first rolled in order to produce the metallurgical bonding of its components. Two operations are carried out: hot rolling and cold rolling. In the hot rolling the preplates are loaded into a furnace, heated up to a predetermined temperature and then, one at the time, rolled. The cold rolling is repeated many times up to obtaining the final process-specified plate thickness. The rolled plates, one at the time, are subjected to quality control, x-rays inspection, trimming and straightening, in order to obtain the final specified dimensions. Afterwards they are transferred to the interim storage before to be subjected to the surface treatment. The plates that makes up the batch (twenty in total) are positioned in an adequate frame carrier.

2.5. Surface treatment section

In this sector the fuel plates are subjected to several chemical treatments in order to accomplish its final cleaning before the fuel element assembling. Two types of chemical reactions are used: acid and alkaline. First the plates are immersed in a sodium hydroxide (NaOH) solution and washed with water. Afterwards the plates are immersed in a nitric acid (NO_3H) solution and washed with water once again. Later the plates are positioned in an adequate frame carrier and transferred back to the interim storage.

2.6. Fuel element assembly section

Here the fuel elements components are assembled in order to make up the MTR fuel element. This fuel element consist of twenty fuel plates, framed by means of a certain number of structural metallic elements. The finished fuel element is subject to quality control, and transferred to the main storage.

3. BASIS FOR THE NUCLEAR CRITICALITY EVALUATION OF THE TYPICAL PLANT

As stated previously, nuclear criticality safety must be assured in normal operations, anticipated operational occurrences and during and after design basis accidents. As shown in the previous section, the process and equipment have been designed in order to meet the limiting values given by Reference [2]. Basically, the plant has been designed using mass, geometry, volume, and concentration control, and/or combinations of them. Mass and geometry controls are of capital importance, so they were used as far as possible. When dealing with solutions sometimes this is no possible, and hence concentration must be controlled by engineered and/or administrative measures. Now the basis for the criticality evaluation of the plant is presented. By design sections will be separated by at least 50 cm, so that they may be assumed to be neutronically isolated from each other [1]. For normal operation and for anticipated operational occurrence, two reflection conditions are possible to be assumed: minimal reflection, i.e. the only neutron reflector is the container itself and/or the stainless steel piping; and nominal reflection, i.e. 25 mm of water reflection which compensate the water in a cooling jacket that normally surrounds the equipments. As usual anticipated operational occurrence double batch may be assumed. The more usual design basis accident is a total flooding of the facility. In this case the nuclear material and water, under optimal moderation, are supposed to be reflected by an effectively infinite thickness of water.

During normal operation 2400 g of total uranium enter to the hydrolysis tank to react with 47 litres of demineralised water, resulting in a solution with 51 g/l in uranium concentration. The density for this aqueous solution of uranyl fluoride was calculated according to published experimental fitting [3], and resulted to be 1.059 g/cm^3 with a corresponding solution volume of 47.7 litres. As a usual anticipated operational occurrence double batch is assumed, that is to say it is supposed that 4800 g of total uranium enters at hydrolysis tank, to react with same quantity of water. In this case a solution with 102 g/l of total uranium is obtained, with a density of 1.116 g/cm^3 and a resulting volume of 48.5 litres.

After completion of hydrolysis, the produced solution is transferred to the precipitation tank. There an ammonia solution is added to the transferred one, and ADU is produced. The quantity of ammonia depends on the specific process carried out in each plant, so that in order to be general and for the sake of conservatism optimal moderation has to be considered. As a usual anticipated operational occurrence double batch is assumed again, as in the case of hydrolyses. As stated before ADU crystals are kept in suspension by means of recirculation, in order to prevent falling of the solids to the bottom of the tank. A failure in recirculation would produce an early ADU precipitation, so that it has been assumed as another feasible anticipated operational occurrence. In order to be conservative a sphere has been supposed for the shape of precipitated ADU, with a variable radius proportional to the precipitated mass. For this kind of solution there is no experimental formulae for predicting density, so in all cases density has been calculated by assuming no interaction among the different chemical substances present at solution. Concerning separation, the same considerations taken into account for precipitation applies. Non-fissile elements, mainly nitrogen, dissolved in solutions involved in the process are neutron absorbers and hence in order to be conservative they are not considered in the criticality calculations, and are replaced by water instead.

For the sake of conservatism, the nuclear material is supposed to take a spherical shape for nuclear criticality evaluation of the operations carried out within calcination section. In this section, nuclear material is present as ADU, or U_3O_8 according to the case. During normal operation 2400 g of total uranium (a batch), equivalent to 3148.80 g of ADU or to 2831.52 g of U_3O_8, are processed. The radius of the sphere results to be 5.32 cm for ADU (assuming 5 g/cm^3 for the bulk density) and 5.52 cm for U_3O_8 (assuming 4 g/cm^3 for the bulk density). During calcination itself, the ADU is loaded in the furnace and heated up to 800°C or up to 1400°C according to the case, but in order to be conservative ambient temperature is assumed for this

operation. As stated before, as usual anticipated operational occurrence double batch is assumed. Concerning design basis accident, a total flooding of the facility is supposed. In this case, a sphere containing ADU powder, or U_3O_8 powder according to the case, homogeneously mixed with water under optimal moderation condition, is supposed to be reflected by an effectively infinite thickness of water. In practice, at least 30 cm of water may be assumed as effectively infinite thickness.

In the fuel preplate manufacturing section, U_3O_8 and aluminium powders are mixed, homogenised and pressed to produce the core compact which will turn into the fuel meat. For this purpose, the initial 2400 g of total uranium are apportioned among 20 independent vessels. These vessels are usually cylinders placed in suitable fixed grids, but for the criticality evaluation they may be supposed to be spheres arranged in such a way that optimal moderation is achieved. In this way subcriticality is assured for normal operation followed by total flooding of the facility. Then the preplates are welded and assembled. In this case, preplates are supposed to be arranged in a grid with separation determined by the assumption of optimal moderation. Two usual anticipated operational occurrences may be assumed as double batch: 20 cores with double load of uranium each, or 40 cores with normal load of uranium each. This applies to core compacts fabrication as well as to cores during preplate assembling. As usual total flood is assumed for design basis accident.

During hot rolling ambient temperature is assumed for the sake of conservatism. Several grids are used for locating the plates: one for the furnace, other for the storage, and another for intermediate operations. For the evaluation, in every case plates are supposed to be arranged in the same grid, with separation determined by the assumption of optimal moderation. As in the previous section, two usual anticipated operational occurrences may be assumed as double batch: 20 plates containing cores with double load of uranium each, or 40 plates containing cores with normal load of uranium each. As usual total flood is assumed for design basis accident.

During chemical treatment the plates are immersed in several pans with different chemicals, in order to accomplish the final cleaning of their surfaces. The same considerations assumed at the previous section are adopted for the nuclear criticality evaluation. Finally in the fuel element assembly section the final fuel element is mounted by assembling the 20 plates with the rest of the structural parts. Fuel elements are assembled one by one, but as an anticipated operational occurrence the presence of another fuel may be supposed. In addition, separation distance between plates and between fuel elements may calculated by assuming optimal moderation. As usual total flood is assumed for design basis accident.

4. ANALYSIS OF POSSIBLE CRITICALITY ACCIDENTS IN THE TYPICAL PLANT

As stated previously, nuclear facilities can be categorised in facilities where a criticality accident may be not credible, and facilities where a criticality accident may be credible. For facilities in the second group, ISO 7753 states that a criticality accident alarm system (CAAS) is not required in areas where the maximum foreseeable accidental dose in free air will not exceed 0.12 Gy [2]. In the typical facility presented in this work solutions containing compounds with uranium enriched up to 20% in weight of ^{235}U are handled. In this plant, all operations are designed to take place by a batch system and they follow international guides which are based on well stated subcritical limits [1]. In spite of these, lessons learned from process criticality accidents occurred in the past [4] show that errors in management, operations, communications, procedures, fissile material accountability and accumulation, and operator knowledge, in addition to equipment malfunction, unanticipated movement of solutions, and a few others, may lead to criticality accidents.

As a first step, it is necessary to assess the consequences of all the credible criticality accidents which could take place in the plant on the basis of the facility design (design basis accidents). Subsequently, it is possible to prepare the emergency response plan for the facility. For this

assessment it is essential to estimate the number of fissions (also commonly named "fission yield") for the postulated criticality accidents. It must be taken into account that this estimated number of fissions has an upper limit of 2 10^{19} for events outside reactor cores [2]. In case of solutions, this estimation may be done by using a simplified method based on the "heat energy formula" [5],[6]. This formula assume that the number of fissions corresponds to the addition of the energy required to bring the solution to the boiling state and, if solution starts to boil, the energy required to evaporate a given quantity of mass [5] so that solution become subcritical again. For each postulated accident, the initial conditions giving rise to criticality must be determined. In addition, in order to estimate the mass of solution evaporated, the critical height must calculated. In this work these calculations were made by means of MCNP6.1 Monte Carlo code [7], using ENDF/B-VII.1 nuclear data library [8]. In all cases, for the sake of conservatism, 180 MeV were considered to be delivered during ^{235}U thermal fission.

Analysing the operations carried out in the typical plant, it was concluded that a criticality accident is credible in hydrolysis and precipitation tanks. As plausible contingency previous to criticality accident in the hydrolyser, it was supposed that all the UF_6 contained in the 5A cylinder was transferred to it. In addition, it was assumed that in the next step the tank was filled with water. In this way the hydrolyser resulted to be occupied with 16.85 kg of total uranium, contained at 54.8 litres (i.e. the total volume of hydrolysis tank) of solution with a corresponding concentration of 307.33 g/l of total uranium. As consequence of the hypothesised criticality excursion this solution was supposed to boil with subsequent evaporation of water, until that the solution became subcritical again. The final fission yield calculated in this case was 1.8260 10^{18}. On the other hand, for the hypothesised accident in the precipitator it was assumed that, as in the case of hydrolysis, it was occupied with 16.85 kg of total uranium. In this case, the final outcome was 70.4 litres of solution (i.e. the total volume of precipitation tank) with a corresponding concentration of 239.5 g/l of total uranium, in addition to 2481.18 g of ammonia. After criticality excursion solution was supposed to boil, with subsequent evaporation of both water and ammonia, until subcriticality of solution. The final fission yield calculated for this supposed accident resulted to be 4.0735 10^{18}.

The fission yields obtained for the assumed accidents are used to assess the corresponding dose mapping of the facility. For the given accidents, two scenarios are possible. On the one hand only one accident may be supposed to happen. In this case, the accident that produce the worst doses may be assumed to occur alone. On the other hand, both accidents may be supposed to occur at the same time. To be conservative the second alternative will be adopted, and the corresponding dose mapping will be calculated.

5. ANALYSIS OF POSSIBLE DOSES IN THE TYPICAL PLANT DURING A CRITICALITY ACCIDENT

In the previous section the credible criticality accidents, which could take place in the plant, were identified on the basis of the facility design, and the fission yields arising from these accidents were estimated. The next step, previous to criticality accident consequences assessment, is the estimation of doses produced by the radiation generated during the assumed criticality accidents. The assumed accidents take place in moderated media, and the only fission nuclide present is ^{235}U. In consequence, the radiation spectra for neutrons and photons arising from thermal fission of ^{235}U is needed in order to estimate the doses received by workers. The resulting prompt spectra for ^{235}U thermal fission, for both neutrons and photons, were calculated by using MCNP6.1 [7] adopting cross section from ENDF/B-VII.1 nuclear data library [8]. As a result, it was obtained that 2.4188 neutrons and 7.1714 photons are produced per fission, with spectra shown at Figure 2.

As stated before, according to ISO 7753 a criticality accident alarm system (CAAS) is not required in areas where the maximum foreseeable accidental dose in free air will not exceed 0.12 Gy [2]. Therefore these areas must be located under the assumption that a criticality

accident would occur in the facility. In the previous sections two different accidents were considered as feasible: one occurring in the hydroliser, and another one occurring in the precipitator. These accidents will be assumed to occur at the same time in order to maximise the doses delivered during a hypothesized criticality accident. The primary radiation source will be assumed to be that calculated previously.

The whole facility was simulated by using MCNP 6.1 [7]. A criticality accident was supposed to occur as described in the previous paragraph and the corresponding dose mapping was made. The composition for the structural materials throughout the facility were taken from reference [9], and the cross sections used were taken from ENDF/B-VII.1 nuclear data library [8]. The floor, roof and walls were assumed to be made of concrete with composition given by NIST [9]. The doors that interconnect the different sections of the facility were taken to composed of an internal layer of borated polyethylene (4.7 weight per cent of boron) and an external layer of lead. The floor and roof thickness were assumed to have a typical value of 10 cm. On the other hand, the dose evaluation was used for obtaining adequate dimensions for walls and doors. To this aim, several thickness values were tried in an iterative way, and the resulting dose mappings were analysed. The final wall thickness adopted was 60 cm. Concerning the doors, they were taken to have 9.5 cm of borated polyethylene and 5 mm of lead. The dose mapping for the finally adopted facility dimensions is shown in Figure 3.

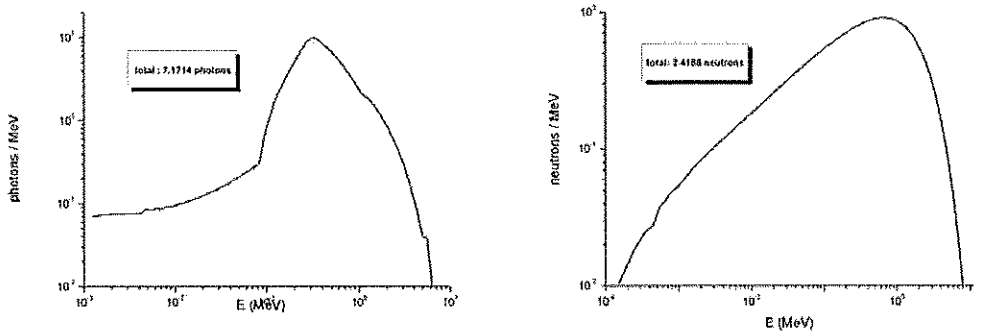

Figure 2: Calculated spectra for photons (left) and neutrons (right) arising from ^{235}U thermal fission.

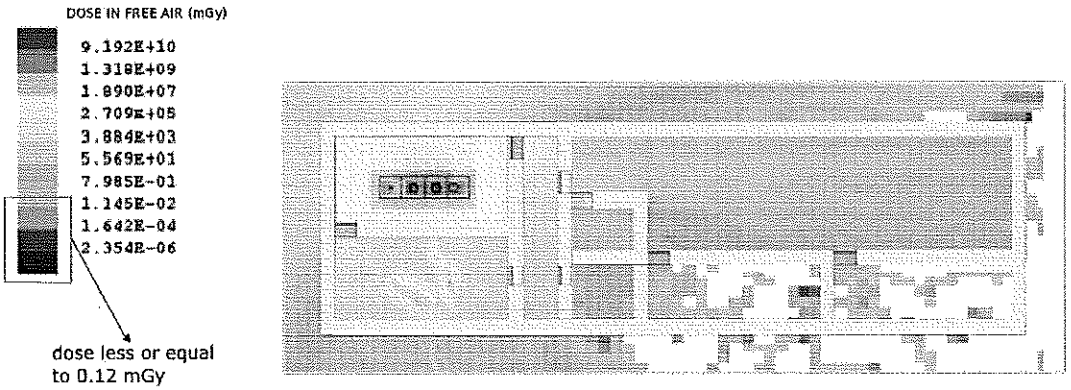

Figure 3: Distribution of dose in free air throughout the facility.

An analysis of the dose mapping shows that the dose in free air falls below 0.12 Gy only in the blue and white zones of Figure 3. So, according to ISO 7753, a criticality accident alarm system

(CAAS) is not required in that zones, but the rest of the facility must be covered with a CAAS. Criticality alarm system shall be designed to detect promptly the minimum accident of concern. For this purpose, in typical unshielded process areas, the minimum accident of concern may be assumed to deliver an absorbed neutron and gamma dose in free air of 0,2 Gy at a distance of 2 m from the reacting material within 60 s [2]. The number and locations of alarms will be defined taking into account specific facility layout, particular associated processes, alarm technical specifications and suitability of feasible locations. The present layout and processes are intended to be generic, so this item is beyond the scope of this work, but the presented methodology may be applied to any specific facility for deciding the alarms locations. In addition, the obtained mapping may be used as an aid for stating the evacuation zone boundary and the evacuation routes, and to design the emergency plan.

6. CONCLUSIONS

The bases for the design, and nuclear criticality evaluation, of a typical facility for the manufacturing of fuel elements for material testing reactor (MTR) were given. In addition, the feasible design basis accidents were identified by analysing the defined representative process. Then the criticality of the accidents were analysed, and the total number of fissions for these accidents were estimated. Moreover, the primary radiation source for these accidents was estimated also.

The whole facility was simulated by using MCNP6.1 Monte Carlo code, with the estimated primary radiation source placed at assumed accidents locations. Subsequently a dose mapping, covering the entire facility, was made under the assumption of the postulated criticality accidents occurrence. The obtained dose mapping was used to define the composition and dimensions of walls and doors throughout the facility.

In addition it was shown that, following ISO 7753 International Standard, the presented methodology may be applied in any specific facility for determining the zones to be covered by CAAS, and as an aid for deciding the more adequate locations for the alarms. Moreover, this methodology may be used as an aid for stating the evacuation zone boundary and the evacuation routes, and to design the emergency plan.

REFERENCES

1. Commissariat a l'Énergie Atomique, *Rapport R 311 4: GUIDE DE CRITICITÉ.*
2. International Organisation for Standardization, *ISO 7753: Nuclear Energy – Performance and Testing Requirements for Critical Detection and Alarm Systems,* First Edition 1987-08-01.
3. J.S. Johnson and K.A. Kraus, "Density and Refractive Index of Uranyl Fluoride Solutions", *J.Am.Chem.Soc.*, **75**, 4594 (1953).
4. Los Álamos national Laboratory, "LA-13638: A Review of Criticality Accidents 2000 Revision".
5. M. DULUC, "New improvement in simplified methods of estimating the number of fissions during a criticality accident in solution," *Proc. Topl. Mtg. Nuclear Criticality Safety Division (NCSD 2009)*, Richland, Washington, USA, September 13-17, (2009).
6. International Organisation for Standardization, *ISO/FDIS 16117:2013(E) Nuclear criticality safety — Estimation of the number of fissions of a postulated criticality accident*, (2013).
7. T. Goorley et ál., "Initial MCNP 6 Release Overview", LA-UR-11-07082, Los Álamos National Laboratory.

8. M.B. Chadwick et ál., "ENDF/B-VII.1 Nuclear Data for Science and Technology: Cross Sections, Covariances, Fission Product Yields and Decay Data," LA-UR 11-05121, Los Álamos National Laboratory.
9. R.J. McConn Jr et ál., "PIET-43741-TM963 PNNL-15870 Rev. 1 Compendium of Material Composition Data for Radiation Transport Modeling," March 4, 2011.