CRITICALITY ACCIDENT STUDIES AND RESEARCH PERFORMED IN THE VALDUC CRITICALITY LABORATORY, FRANCE

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Abstract

In 1967, the IPSN (Institut de Protection et de Sûreté Nucléaire – Nuclear Protection and Safety Institute) started studies and research in France on criticality accidents, with the objective of improving knowledge and modelling of accidents in order to limit consequences to the public, the environment and installations.

The criticality accident is accompanied by an intense emission of neutronic and gamma radiation and releases of radioactive products in the form of gas and aerosols, generating irradiation and contamination risks. The main objectives of the studies carried out, particularly using the CRAC installation and the SILENE reactor at Valduc (France) were to model the physics of criticality accidents, to estimate the risks of irradiation and radioactive releases, to elaborate an accident detection system and to provide information for intervention plans.

This document summarizes the state of knowledge in the various fields mentioned above. The results of experiments carried out in the Valduc criticality laboratory are used internationally as reference data for the qualification of calculation codes and the assessment of the consequences of a criticality accident. The SILENE installation, that reproduces the various conditions encountered during a criticality accident, is also a unique international research tool for studies and training on those matters.

1. PURPOSES OF CRITICALITY ACCIDENT STUDIES

The study of criticality accidents that could occur in installations aims to the following objectives: evaluating exposure risks for operators, identifying possible means of detection, studying the long term behaviour of the critical configuration, evaluating the consequences of radioactive releases on the public and the environment and providing information that could help to prepare intervention plans and crisis management.

Up to the present day, about sixty accidents occurred throughout the world, two thirds of them in research installations and one third in fuel cycle installations, causing the death of about twenty operators [1, 2].

Criticality accident study programs were started in France in 1967 in order to improve knowledge about accidents. IPSN initiated experiments reproducing the criticality accident by divergence of a fissile solution of uranyl nitrate on the CRAC and SILENE installations.

2. GENERAL PHENOMENOLOGY OF A CRITICALITY ACCIDENT

The criticality accident is the result of an uncontrolled chain fission reaction being started when the quantities of nuclear materials (uranium or plutonium) present accidentally exceed a given limit called the ‘critical mass’.

As soon as the critical state is exceeded, the chain reaction increases exponentially within a time period that depends on the overall reactivity of the system. The result is a fast increase in the number of fissions that occur within the fissile medium. This phenomenon results in a release of energy mainly in the form of heat, accompanied by the intense emission of neutronic and gamma radiation and the release of fission gases. The increase in the temperature of the fissile medium usually causes the appearance of neutronic feedback mechanisms that will reduce the reactivity present until the system becomes sub-critical, even if only temporarily. The result is usually the appearance of a power peak. After this first peak, radiolysis gas or steam bubbles migrate to the surface such that the resulting antireactivity effect disappears and the power excursion restarts. This process by which bubbles are formed and then released outside the system causes the oscillating phenomenon usually observed during a criticality accident [Fig. 1].

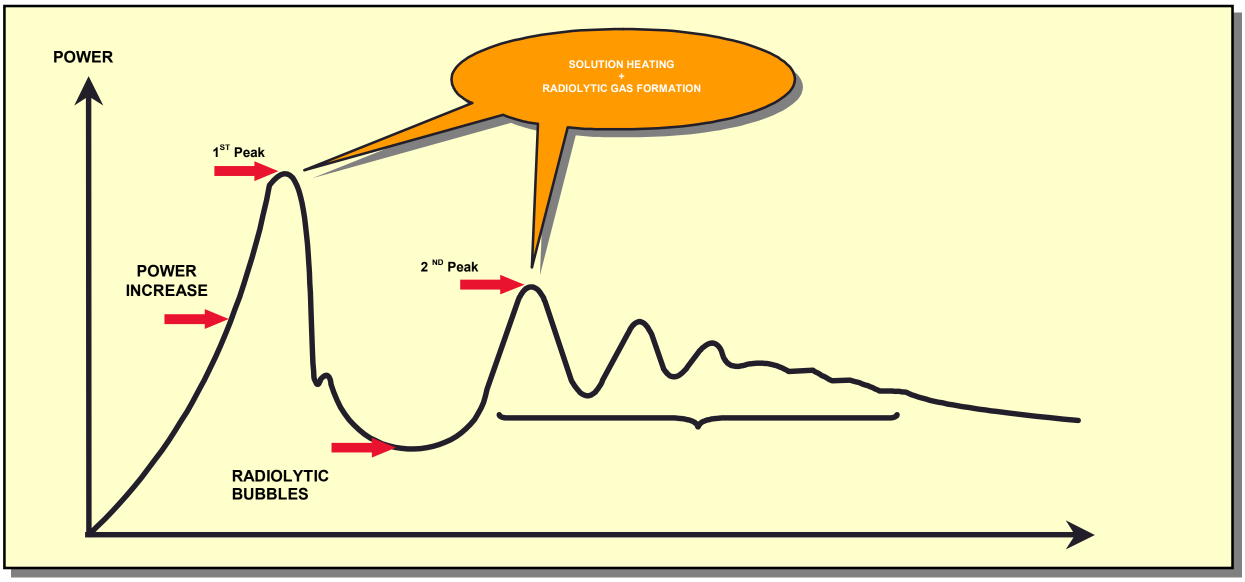


FIG. 1. Typical criticality accident in a fissile solution

Therefore globally, the behaviour of a criticality excursion is defined by the following main parameters:

- the physicochemical nature of the critical fissile medium;
- the initial neutronic source, depending on whether it is uranium or plutonium;
- the reactivity inserted in the system and the neutronic feedback mechanisms resulting from temperature increase, void effects (radiolysis gas and steam bubbles), and heat exchanges with the environement.

As confirmed by experiments performed in the SILENE reactor, the combination of the previous phenomena with the initial accident conditions can lead to three types of behaviour [Fig. 2]:

1. the critical system becomes permanently sub-critical by modifying the configuration (mixing, splashing or dispersion of material, modification of the geometry, etc.);
2. the system is made temporarily sub-critical by the increase in the temperature of the fissile material, and in this case the critical reaction will restart after a variable time interval that depends on heat exchanges with the environment;
3. following a large initial reactivity, the system reaches temperatures at which the medium boils and the variation in power then depends on whether the medium is under or over moderated. The behaviour of the critical system during the post-accident phase depends also on whether or not the system is confined.

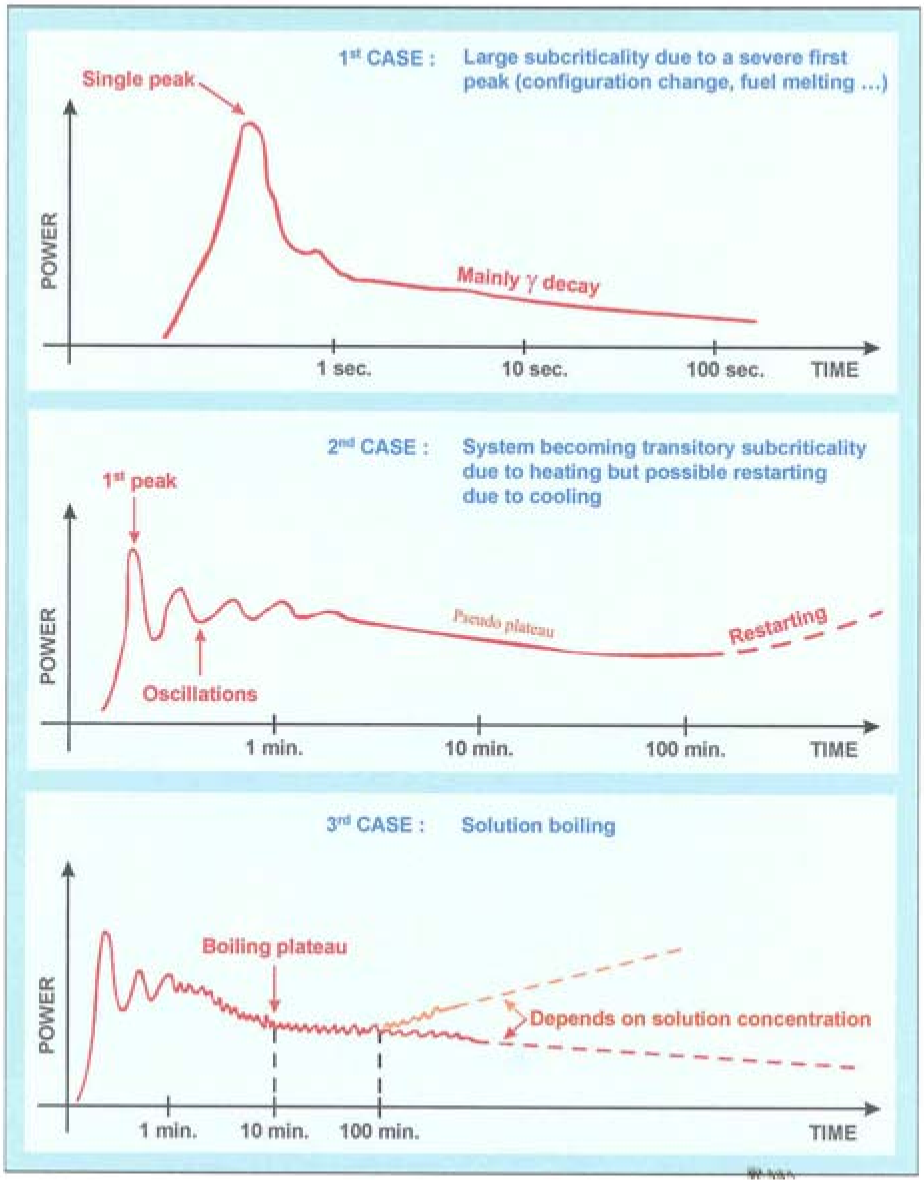


FIG. 2. Post-accident phases of a criticality accident

This description applies to typical situations but it is in no way exhaustive since every criticality accident can have unique circumstances, as is clearly demonstrated by looking at accidents that have actually occurred throughout the world and particularly the most recent accident in Tokai-Mura in which a tank cooling device modified the events during the post-accident phase.

3. ACQUIRED KNOWLEDGE ABOUT CRITICALITY ACCIDENTS PARTICULARLY ABOUT AQUEOUS MEDIA USING THE CRAC AND SILENE FACILITIES

About 70 experiments were performed on the CRAC (*Conséquences Radiologiques d'un Accident de Criticité* - Radiological Consequences of a Criticality Accident) installation in the Valduc Criticality Laboratory between 1967 and 1972, reproducing criticality accidents in an uranyl nitrate fissile medium [3, 4]. The studies carried out were continued on the SILENE reactor starting in 1974 and more than 2000 divergences have been carried out so far [5, 6].

Parameters varied within the following ranges in these experiments representative of accident situations: uranium concentration between 20 and 340 g/l, potential reactivity ρ less than 10 $ in a homogeneous system, the dollar $ being the value of the reactivity corresponding to the 'prompt' critical reactivity (also called β).

3.1. Results and practical information about accident physics

The results and acquired information can be summarized as follows:

First power peak and associated effects

- power period Te varying from 0.9 ms to 4 minutes,
- maximum power ranging from 10^{12} to $3x10^{19}$ fissions.s^{-1}.

The maximum values of the total energy of the first power peak were observed for the largest volumes ($3x10^{17}$ fissions for a volume of 230 liters). For fast transients ($\rho >> \beta$), the maximum first peak power $\dot{E}$ is varying with the reciprocal period ω as a function of $\omega^{1.8}$.

Some of the observations were :

- the appearance of a pressure wave for fast kinetics ($T_e < 10$ ms), also causing noise ;
- splashing of the solution under the fast transients if there is no lid on the vessel ;
- a blue light due to the CERENKOV effect concomitant with the power peaks.

Energy recovered under the thermal form

The fraction of energy that is actually retrieved in thermal form during the CRAC and SILENE tests corresponds to $1.4x10^{11}$ fissions.cal^{-1} (about 180 MeV) retrieved in the form of heat.

For power excursions subsequent to high reactivity (several \$), it was observed that the boiling of the solution was reached for an energy corresponding to about $1.1x10^{16}$ fissions per liter. These data are valid for a power excursion lasting for a few minutes and for a system without any forced cooling.

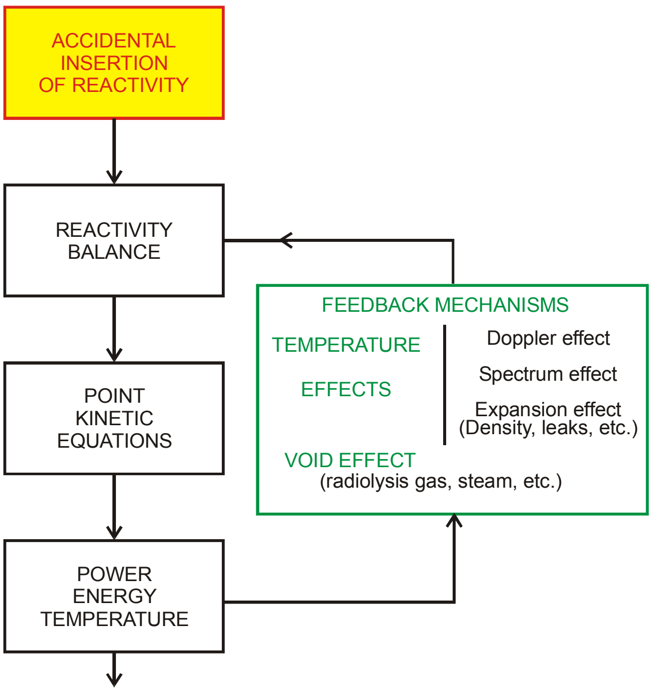
Formation of radiolysis gases

Many experiments have shown a rate of formation of radiolysis gases corresponding to $1.1x10^{-13}$ cm^3/fissions. Furthermore, the threshold at which these gases appear has been estimated at $1.5x10^{15}$ fissions per liter of solution [7].

3.2. Modelling of accident physics

An analysis of past criticality accidents illustrates the wide variety of situations encountered : media, configurations, causes and observed effects (power, energy, duration, etc.). The results show that the energy can vary from a few 10^{15} fissions to 4×10^{19} fissions for fuel cycle installations, and the power during the first peak can be as high as 10^{20} fissions.s^{-1} for a very short time. The duration can simply be a 'flash' of a few milliseconds, or it can continue for tens of hours.

The diversity of these effects is directly related to parameters that affect the accident phenomenology. This is why different accident models were developed, making a distinction between four main environment categories (liquid, powder, metal, fuel rods in water). The following diagram illustrates the common architecture of these calculation programs.



Accident calculation programs developed jointly with the UKAEA (CRITEX for aqueous media, POWDER for powders, CHATEAU for immersed fuel rods) can be used to estimate the variation of the power, energy and temperature of the medium during the early times of the accident [8].

3.3. Exposure risks associated with a criticality accident: CRAC and SILENE results

The contribution of neutrons and gamma rays to the total dose is very variable depending on the nature of the fissile material (metal, powder, liquid, etc.), the dimensions and compositions of the system, and its environment. As the distance from the source increases, the energy of the radiation field degrades and its intensity decreases approximately inversely as a function of the square of the distance over the first few meters. For longer distances, radiation propagation laws are more complex due to effects related to the ground and the atmosphere.

Dosimetry results obtained on the CRAC and SILENE installations [9] must be considered as being representative of the dose to which personnel could be exposed during a criticality accident in a uranyl nitrate solution. The number of fissions and the emitted dose are not proportional to each other for sources with very different configurations, since leakage radiation depends on the source characteristics. It is found that the dose/fission ratio is maximum for small sources with low concentrations.

The maximum value of the observed total dose during tests on the CRAC and SILENE installations is 5.8 x 10^2 Gy at 1m from the centerline of the source, for 10^{18} fissions for a 30 cm diameter cylinder with a concentration of 80 g/l.

For information, doses emitted during the first peak on the SILENE reactor for 10^{17} fissions at 1 m from the core (40 liters of uranyl nitrate solution) are as follows:

- Neutrons : Dose (KERMA tissus) $\approx$ 20 Gy $\Rightarrow$ Equivalent dose $\approx$ 300 Sv
- Gamma : Dose $\approx$ 25 Gy $\Rightarrow$ Equivalent dose $\approx$ 25 Sv.

Measured doses demonstrate that the risk of exposure is one of the major risks in a criticality accident and the resulting doses can be fatal for personnel working in the immediate vicinity of the equipment concerned.

3.4. Detection of criticality accidents

The purpose of a criticality accident detection system is to trigger an alarm as quickly as possible in order to trigger immediate evacuation of personnel at the beginning of a criticality accidental excursion and thus limit exposure risks.

In 1976, the CEA designed the E.D.A.C system (*Ensemble de Détection et d'Alarme de Criticité)* making use of information derived from the CRAC and SILENE experiments, based on a monitoring unit connected to at least three criticality detectors [10]. The criticality alarm is only triggered if at least two detectors send an alert signal to the monitoring unit. The detection system is based on measuring the total dose due to neutronic and gamma radiation by means of two scintillators, sensitive to these two types of radiation.

Tests carried out in the SILENE reactor demonstrated that the system can be used to detect all types of accidents, in other words power excursions with fast kinetics and with slow kinetics. The EDAC accident detection system can also record and monitor the evolution of the accident by means of criticality detectors, particularly through a remote console placed outside the evacuation area. Its contribution may be essential for management of the post-accident situation and intervention.

3.5. Estimate of releases of radioactive products during a criticality accident in solution

The SILENE installation was used for an experimental program to determine the rates of release of fission products (FP) emitted during a criticality accident in an aqueous fissile medium, the experimental conditions varying up to and including boiling of the solution to facilitate the release of fission products [11]. The main information derived from the SILENE fission products program is as follows :

- the release ratios of rare gases (Xe and Kr) are almost 100% for gases with half-lives of more than one minute. They vary between 10% and 50% for half-lives varying from a few seconds to a minute, and are of the order of 10% for very short half-lives;
- the maximum release ratios observed for iodine for acidity close to 2N, were very much less than 1% for a boiling solution, but about 10% for a low solution acidity and a high initial content by load of iodine in the solution;
- the maximum emission ratios for other volatile fission products are estimated at 20% for bromine and 1% for ruthenium.

For information, the maximum quantities of fission products released from the solution for 10^{18} fissions are 3 x 10^{14} Bq for rare gases and aerosols and about 1.8 x 10^{12} Bq for iodine.

3.6. Experiments and exercises carried out on the SILENE reactor for action management

The SILENE reactor is used to provide evaluation data necessary for intervention management following a criticality accident. The following themes are considered:

- estimate of the possible dose to a work team during the post-accident phase [9];
- dosimetry of the criticality accident ; SILENE is an international reference source and has already been used for the purposes of international exercises under the auspices of the AIEA and the CCE [12];
- radiation instrumentation test during the post-accident phase;
- fast checking of exposed personnel and dose estimate (sodium activity and dosimeter measurements, for example) for an appropriate therapeutic treatment.

4. CONCLUSIONS

The criticality accident studies carried out have improved knowledge in several fields : physics, detection, dosimetry and the release of radionuclides. These results must contribute to a better assessment of the risks of irradiation and contamination associated with a criticality accident and the application of action measures and provisions for crisis management. They emphasize the need to define well intervention plans and to be capable to stop the accident process.

More generally, operating experience with real accidents that have occurred throughout the world confirms that the energy released during a criticality accident is generally limited, but there are severe risks of irradiation for personnel working close to the equipment concerned and lethal doses are possible. The unfortunate Tokai-Mura accident also demonstrates that the scale of the consequences in terms of the media and acceptability of the nuclear risk may be completely different.

The SILENE reactor is a unique international research installation that can be used for training teams and to maintain the skills necessary for management of action to be taken following a criticality accident.

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