STUDYING THE PHENOMENOLOGY OF CRITICALITY ACCIDENTS

We cannot completely exclude the possibility of a criticality accident, in which the quantity of fissile material present exceeds the critical mass in spite of all the preventive measures. What happens then and what are the risks? Research provides answers to these questions, improving our knowledge and our ability to model accidents so that we can limit their consequences for people, for the environment and for the nuclear facilities themselves.

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Criticality accident dosimetry at Valduc (Côte-d'Or), sited next to the Silène test reactor (right, surrounded by orange shielding). Some of the sensors are attached to dummies, simulating the operators.

Since 1945 there have been about sixty criticality accidents reported world-wide, mostly in the USA and former Soviet Union. Two thirds occurred in research facilities (involving **critical assemblies** and reactors), the rest were in fuel cycle facilities. They caused 19 deaths in total, but have not resulted in significant radioactive releases into the environment (see table 1). In France, the preventive measures taken have meant that no incident resulting in acute exposure to an operator has occurred in the fuel cycle, but situations have arisen which *could have* led to an accident.

date and location	duration number of irradiated persons	number of fissions body exposure (Gy = gray; Sv = sievert)	circumstances, sequence of events and causes
1 13/12/1978 Siberian Chemical Combine (Russia)	1 peak 1 person strongly irradiated and amputated + 7 people irradiated	$3 \cdot 10^{15}$ fissions 2.5 Gy 0.05 to 0.6 Gy	Too many plutonium ingots (4 ingots, 10.7 kg Pu) introduced into a container. Stopped by ejecting or removing the ingot
2 13/11/1965 Electrostal Fuel Fabrication Plant (Russia)	1 peak	$8 \cdot 10^{15}$ fissions	Accumulation of UO_2 powder (70 kg) enriched to 6.5%, representing 4.6 kg ^{235}U, in the water tank of a vacuum system pump, following a breach in the filters. Stopped by dispersing the oxide
3 16/10/1959 Idaho (USA)	20 minutes	First peak: $1 \cdot 10^{17}$ fissions Total: $4 \cdot 10^{19}$ fissions	U (91%) - 30.9 kg of ^{235}U in 200 l at a concentration of 155 g/l and overflowing into a 19 m^3 tank containing 600 l of effluent. Stopped by evaporating 400 l of the solution and crystallizing out the nitrate
4 07/04/1962 Hanford (USA)	37 hours 3 people irradiated	First peak: $1 \cdot 10^{16}$ fissions Total: $8 \cdot 10^{17}$ fissions 1.1 Sv, 0.43 Sv, 0.19 Sv	45 l of solution at a concentration of 28.7 g/l, representing 1.29 kg ^{239}Pu; stopped by evaporation
5 15/02/1971 Kurtchatov Institute (Russia)	50 power peaks 2 people irradiated on the feet	$2 \cdot 10^{19}$ fissions 15 Sv	Critical model of 20% enriched uranium fuel assemblies in water used for the subcritical approach, with a beryllium reflector and absorbent elements; the tank was filled while the neutron source was not in place and the control rods were lowered; stopped by evaporating water and stopping the filling pump

Table 1. Characteristics of some of the most significant criticality accidents so far world-wide in fissile media of various physical and chemical forms (metal in case 1, powders in case 2, aqueous solutions (of uranium and plutonium) in cases 3 and 4 and critical fuel rod assemblies surrounded by water in case 5). These examples also illustrate that the criticality accident may last just for the moment of the pulse (case 1) or for several tens of hours (case 4) and may produce widely varying amounts of energy ($3 \cdot 10^{15}$ fissions for accident 1 and $4 \cdot 10^{19}$ fissions for accident 3).

Two reactor accidents occurred at Saclay during the nineteen sixties, but without causing any serious irradiation of personnel.

By studying the phenomenology of criticality accidents that might occur in different configurations and using appropriate **calculation code** to model these accidents, we aim to describe the different scenarios that might arise in a given experimental or industrial process. Such studies have several objectives: to assess the risks of irradiation for operators in the event of an accident, to determine the detection possibilities depending on the kinetics of the accident and the performance and location of the detection devices, to evaluate the long term behavior of the critical configuration, to assess the possible consequences in terms of radioactive release or irradiation in the environment, and to prepare for action aimed at bringing the system back to a **subcritical** state.

As early as 1967, the IPSN (Institute for Protection and Nuclear Safety) began work in its criticality laboratory at Valduc (Côte-d'Or) on experiments which reproduced criticality accidents by **divergence** of a **fissile** solution of uranyl nitrate.

The general phenomenology of a criticality accident

A criticality accident results from an uncontrolled **chain fission reaction** being triggered when the quantities of nuclear material present (uranium or plutonium) accidentally exceed a certain threshold called the **critical mass**. In **neutronics** terms, as soon as the critical state is exceeded, the chain reaction becomes divergent and rises exponentially with a period which depends on the initial **reactivity** of the system. The result is a rapid growth in the number of fissions produced within the fissile medium, called a **criticality excursion**. This phenomenon (see *The risk of criticality and its prevention in nuclear plants and laboratories and during transportation*) results in the release of energy, mostly in the form of heat, together with an intense emission of neutron and gamma **radiation** and the release of fission gas. The heating of the fissile medium generally results in the appearance of feedback effect mechanisms which reduce reactivity until the system is rendered subcritical, even if only temporarily (see chapter II). The usual result is therefore a power peak (figure 1). After this first peak, bubbles of **radiolysis** gas or steam migrate to the surface, the resulting anti-reactivity effect disappears and the power excursion starts again. It is this appearance and release of bubbles from the system which causes the power oscillation generally observed in a criticality accident.

Four main parameters determine the way an accidental criticality excursion proceeds: the physical and chemical nature of the divergent fissile medium, the reactivity of the system (an expression of the supercriticality level), the initial spontaneous neutron source (non-irradiated enriched uranium, irradiated uranium containing plutonium or just plutonium on its own) and the neutronics feedback effects. There are three types of feedback effects: effects linked to heating of the fissile material, either nuclear temperature effects (Doppler effects and spectral variation effects: see box 1, *Control and operation of pressurized water reactors*) or expansion effects (density and volume effects), and the void effect (bubbles of radiolysis gas in the case of a solution, steam, etc.). The environment of the facility also has

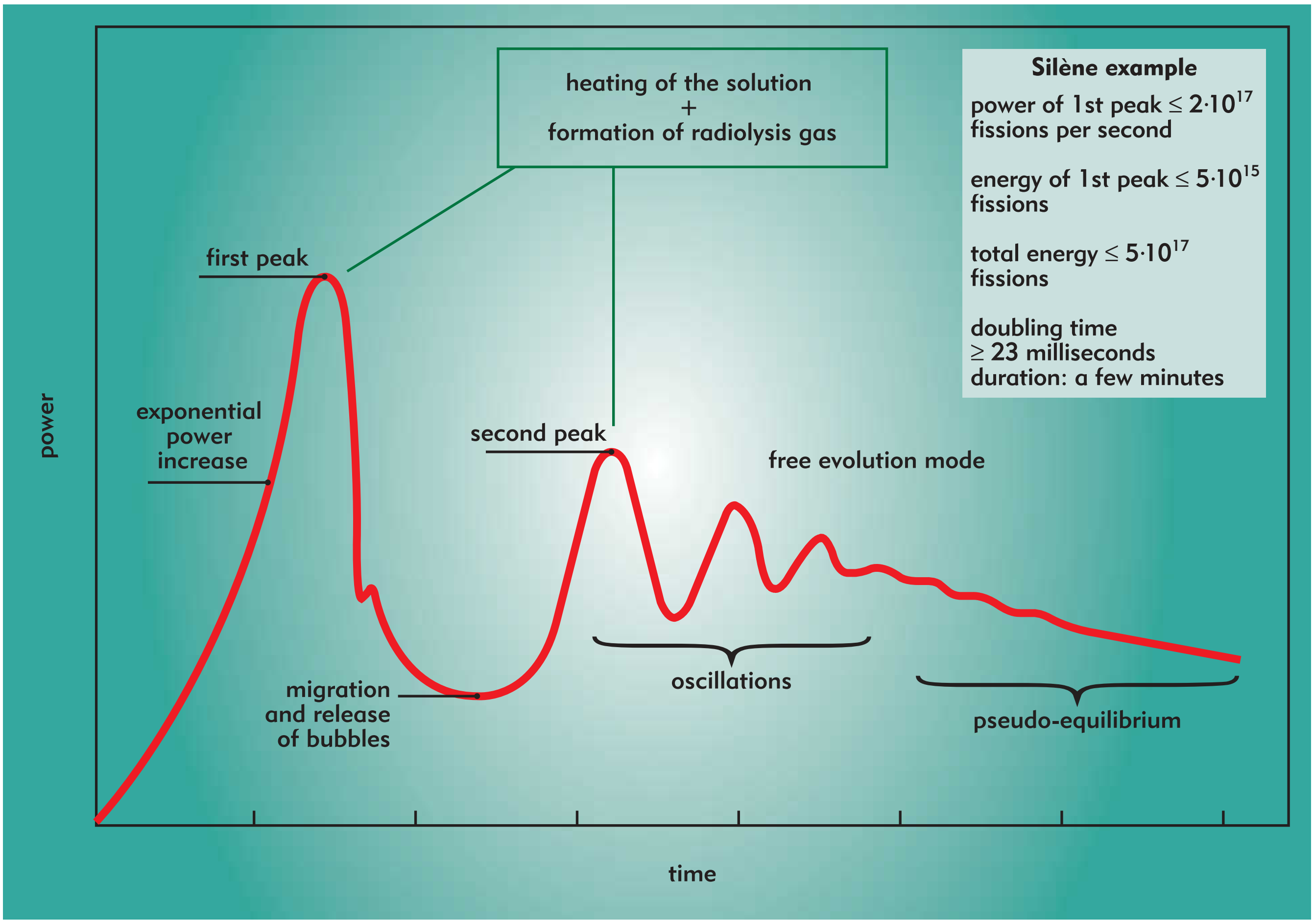


Figure 1. Power level variation during a criticality accident in an aqueous fissile medium.

an effect (thermal exchanges with the outside, confinement of the divergent system, etc.).

The experiments performed in the Silène reactor at Valduc confirmed that combining the phenomena described above with the initial conditions of the accident may result in three types of behavior (figure 2). In the first case, the divergent system returns permanently to the subcritical state as a result of some modification to the configuration (such as mixing, ejection or dispersal of material, or a change in geometry). In the second case, the system is temporarily made subcritical by the heating of the fissile material: the divergent reaction is then restarted after a time interval of varying duration depending on the thermal exchanges with the surrounding medium. In a third case, following an initial high reactivity, the system reaches the boiling point of the medium: the rise of the power then depends on whether the medium is over-**moderated** or under-moderated. As the liquid boils and the fissile solution becomes more concentrated, the reactivity of the system may either increase or decrease. The behavior during the post-accident phase of the divergent system differs depending on whether it is a *closed* system, in which the vapor may recondense and return to the solution, or an *open* system, in which case the vaporization or ejection of the solution will allow it to return to subcriticality.

This description corresponds to typical situations but is certainly not exhaustive. Each accident may have its own special characteristics, as witnessed by those that have occurred so far and especially the most recent, the 1999 Tokaï Mura accident in Japan, during which the post-accident phase was changed by a refrigeration system around the tank

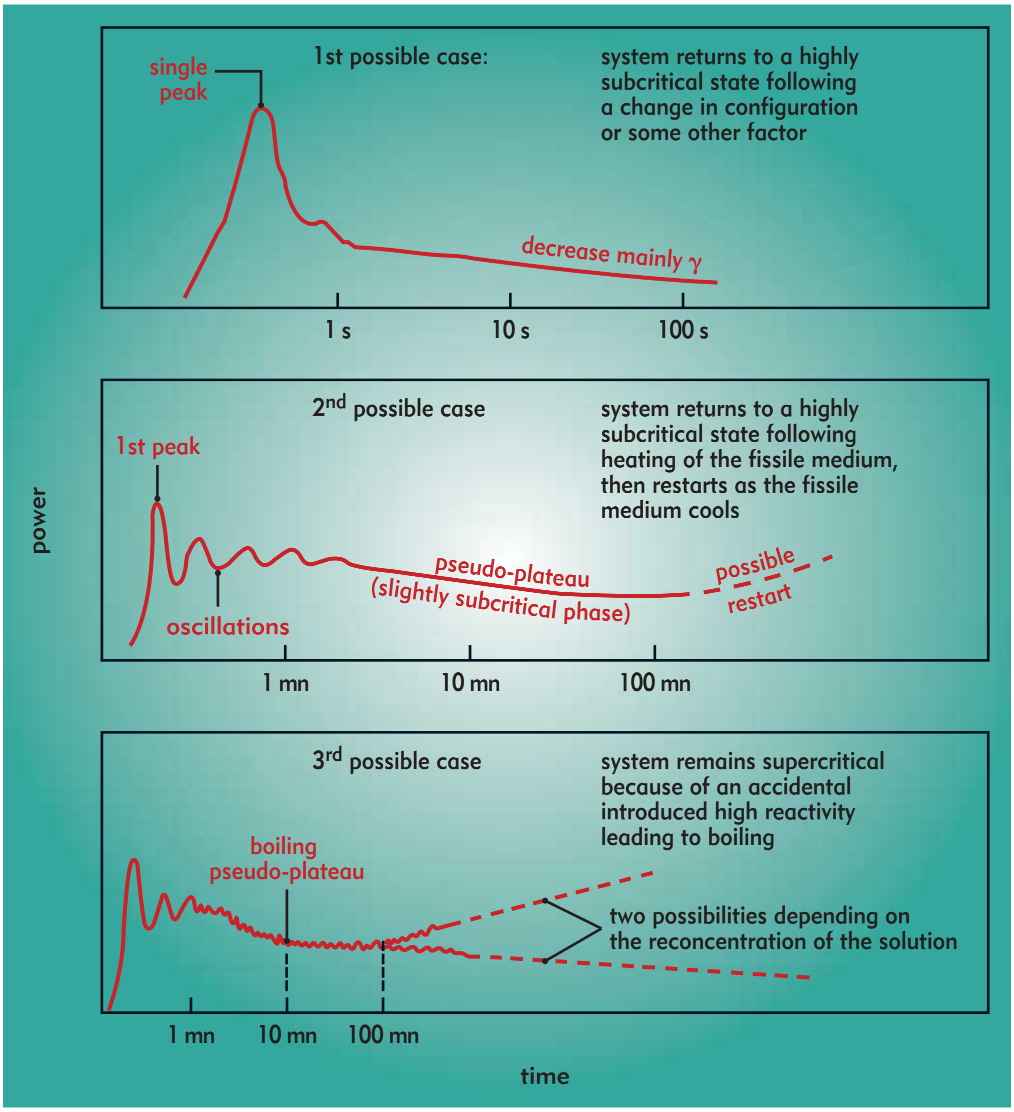
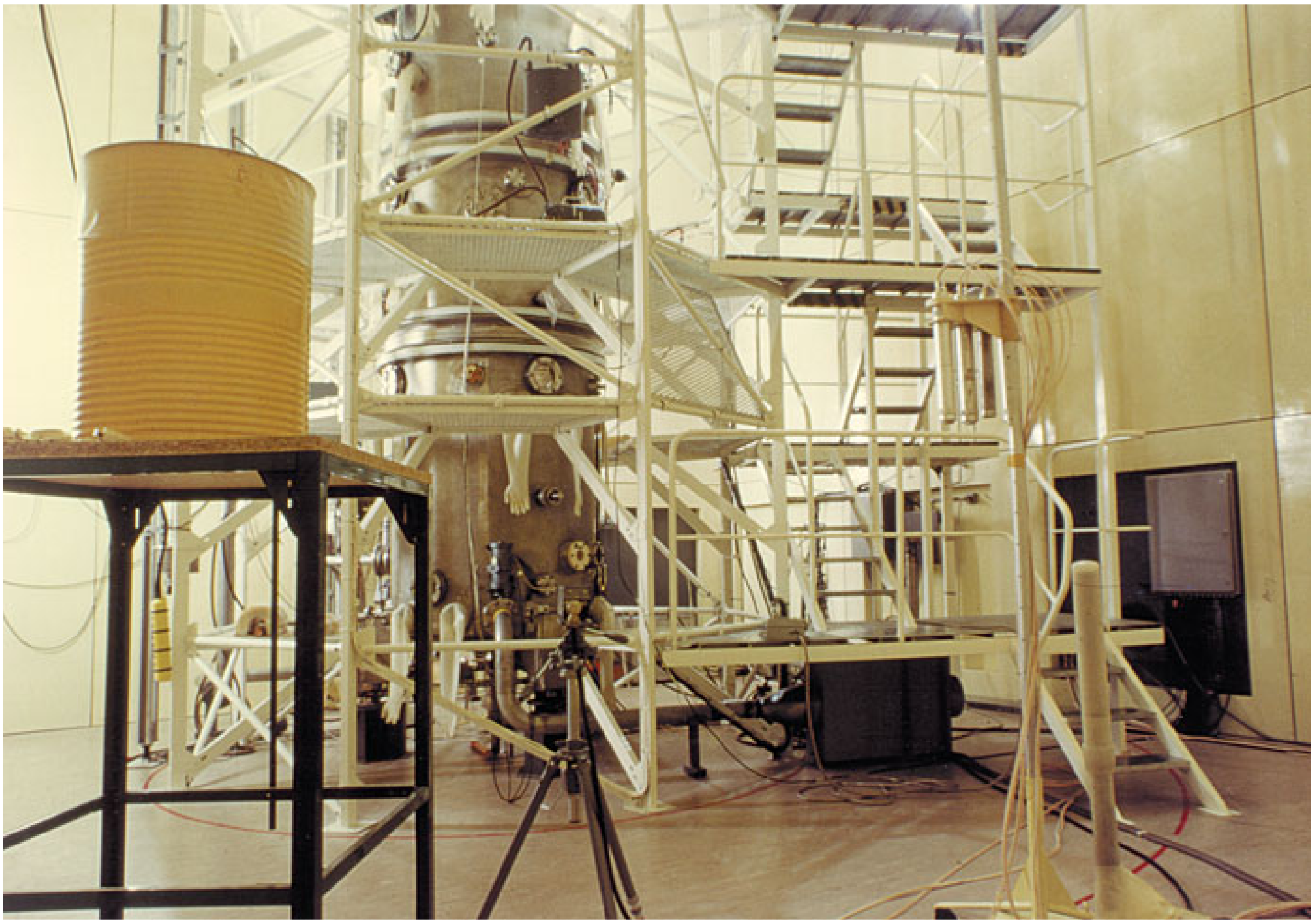


Figure 2. Post-accident phases of a criticality accident.



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General view of the Valduc Crac facility, where experiments on the radiological consequences of criticality accidents began in 1967.

containing the divergent system (see *The Tokaï Mura criticality accident*).

A criticality accident is also accompanied by the emission of neutron and gamma radiation and a release of gaseous radioactive **fission products** and aerosols. The experimental programs conducted at Valduc have enabled us to evaluate the associated risks of irradiation and contamination and to highlight the need to detect the accident, to monitor its progress during the post-accident phase and to prepare measures which will stop the process.

Using Crac and Silène to find out more about accidents in aqueous fissile media

France was the first country to start a research program in this domain, where detailed information on the accidents that have occurred world-wide are relatively recent. About 70 experiments reproducing criticality accidents in aqueous fissile media were carried out at the Valduc criticality laboratory in the Crac facility between 1967 an 1972 (the French acronym Crac stands for "radiological consequences of criticality accidents"). Research was continued from 1974 onwards in the Silène reactor.

In the Crac experiments, the reactivity was generally introduced by continuously pumping a solution of uranyl nitrate into cylindrical tanks 0.3 to 0.8 m in external diameter. The pump was capable of delivering up to 1,800 l/h. In the Silène facility, which comprises an annular tank 0.36 m wide, reactivity is added by removing a control rod from the core.

The parameters used in these experimental representations of accident situations were a uranium concentration in the range 20 to 340 g/l, a potential reactivity of less than 10 \$(1) in a homogenous system, reactivity "ramps" and "steps" of up to 2 \$/s and 3 \$ respectively and a variable initial neutron source.

(1) Dollar (\$) is the reactivity value corresponding to the **"prompt"** critical state.

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Experimental lessons on the physics of criticality accidents

The lessons drawn from experiments on criticality accidents may be summed up under five main points:

Initial power peak and its associated effects

Carrying out tests on homogenous media, we have been able to explore a domain characterized by a reactivity "step" ranging from 35 to 2,350 **pcm** (3 \$), a reactivity "ramp" of up to 2 \$/s, a power rise period T_e ranging from 0.9 ms (Crac test) to 4 minutes (Silène test) and a maximum power in the range 10^{12} to $3{\cdot}10^{19}$ fissions/s. The maximum values of the total energy in the initial power peak have been observed for the largest volumes ($3{\cdot}10^{17}$ fissions).

Four observations are particularly important

The first is the appearance of a pressure wave for rapid kinetics ($T_e <$ 10 ms), also causing a sound effect. The second observation, for these same kinetics, is the ejection of solution out of the experiment tank when the tank has no cover. The third event is the appearance of a blue flash caused by the **Čerenkov effect**, concomitant with the appearance of the power peaks, hence the notion of a flash often associated with a criticality accident. The fourth element is the major influence that the initial source of neutrons has on the initial peak, with the probability of triggering a power excursion being a function of the population of neutrons present in the medium.

Thus for solutions of plutonium, in which an intrinsic neutron source exists because of spontaneous fissions and the (α, n) reactions, the insertion of reactivity will be more like a "ramp", whereas for uranium solutions with a low neutron source, the accident may be more rapid and correspond to a reactivity "step".

Heat energy recovered

By comparing the total number of fissions (determined by radiochemical analysis of the formed fission products) with the thermal balance deduced from the heating of the fissile solution, we can determine the fraction of energy released which is actually recovered in the form of heat. The value measured in Crac and Silène tests is of the order of $1.45{\cdot}10^{11}$ fissions/cal, which represents about 180 MeV recovered in the form of heat for 200 MeV emitted by the fission.

Formation of radiolysis gas and related effects

After a large number of experiments, it has been shown that radiolysis gas forms at a rate corresponding to about $1.1{\cdot}10^{-13}$ cm^3/fission, making about 110 liters of gas for 10^{18} fissions. The threshold at which these gases appear is estimated at $1.5{\cdot}10^{15}$ fissions per liter of solution in the media studied.

Solution boiling

For power excursions resulting from a high reactivity (several \$ or a few thousand pcm), it has been observed in Crac and Silène experiments that the solution reaches boiling point (about 102 °C) for an energy of about $1.1{\cdot}10^{16}$ fissions per liter. These values are valid for a power excursion of a few minutes and a system which is not subject to forced cooling.

Relationship between energy and solution volume

An empirical relationship which expresses the growth in the total number of fissions N_f as a function of the solution volume (in liters) and of time (seconds) has been established based on the most representative Crac and Silène experiments in homogenous media:

$$N_f(t) = \frac{t}{3.55{\cdot}10^{-15} + 6.38{\cdot}10^{-17} \times t} \times V$$

This may be considered as giving a "boundary" value for the number of fissions over a time lasting a few minutes until the solution boils. The post-boiling phase has to be considered separately depending on the accident scenario in question. The relationship shows in particular the important influence that the volume of the fissile solution has on the energy likely to be produced.

Detailed physical modeling of phenomena based on the Crac and Silène experiments has led to the development of the Critex calculation code.

Physics of the accident: from experiment to modeling

The general phenomenology of the progress of a criticality excursion in an aqueous fissile medium has been described earlier. The experiments gave us results and allowed us to draw lessons in five main areas (see box 1).

A brief analysis of criticality accidents reported to date illustrates the extreme diversity of the accident situations (different fissile media, complex configurations, and a diverse range of causes) as well as that of the effects observed (their power, energy, and duration). This indicates the complexity of the computer models that need to be developed to account for these situations.

It should first be emphasized that the energy produced in a criticality accident (often expressed as a number of fissions) is much lower than that which would result from a reactor accident (in the case of the Chernobyl accident, the number of fissions has been estimated at between 6 and $8 \cdot 10^{21}$ fissions). A criticality accident of 10^{18} fissions releases about thirty megajoules of energy (1 joule = $3.1 \cdot 10^{10}$ fissions) and represents a "consumption" of uranium or plutonium of less than a milligram. An examination of the accidents shows that the energy may vary from 10^{15} fissions to $4 \cdot 10^{19}$ fissions for fuel cycle facilities, with the power of the initial peak reaching up to 10^{21} fissions/s for a very short time. The duration of a criticality accident may vary from just a single pulse lasting about a millisecond to a situation that persists for tens of hours.

The diversity of these effects is directly related to the parameters which influence the phenomenology of the accident. Different accident models have thus been developed which distinguish four main categories of medium: liquid, powder, metal, fuel rods and water. The common architecture of the corresponding calculation code is shown in figure 3. The currently developed codes (called "Critex" for aqueous media, "Powder" for powders and "Chateau" for immersed fuel rods) can be used to estimate the variations in power, energy and temperature of the medium as a function of time over a limited period (the first minutes of the accident). Based on their results, we can estimate the possible consequences for humans and the environment: the risks of irradiation and resulting contamination from emitted radiation and from the radionuclides released.

Risks of exposure: doses measured around Crac and Silène

Criticality accidents are accompanied by an intense emission of neutron and gamma radiation of variable duration, ranging from a few seconds to several hours depending on the phenomenology of the **power excursion**. The initial radiation field is a complex field of neutrons and gamma radiation of energy up to ten or so MeV. The contribution each makes to the total radioactive dose varies

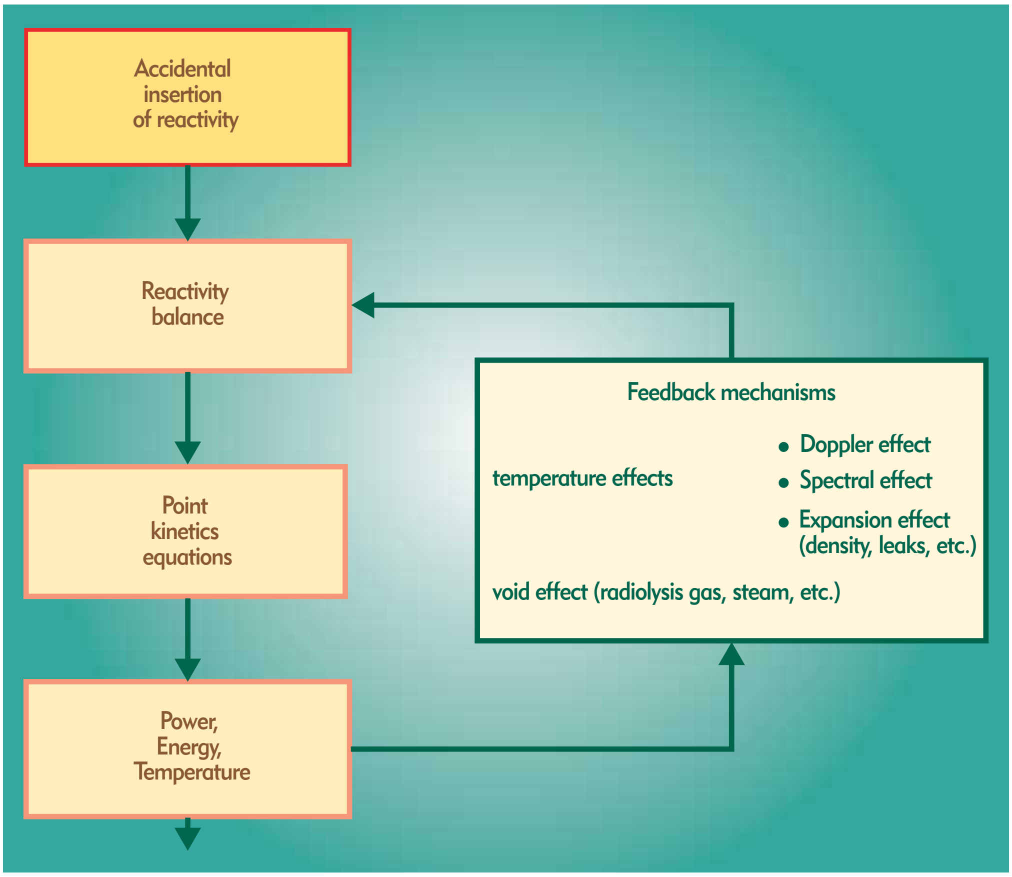


Figure 3. General architecture of the criticality accident computer software.

greatly depending on the nature of the fissile material (metal, powder, liquid, etc.), the dimensions and the composition of the divergent system and its environment. Thus the ratio of the neutron **dose** to the gamma dose may vary from 10 for a metal source to 0.2 for a strongly moderated ring medium. As we move further from the source, the radiation field is reduced in energy and its intensity drops very rapidly, varying approximately with the inverse square of the distance for the first few meters. Beyond this distance, the propagation laws are more complex, due to effects linked to the ground and the atmosphere.

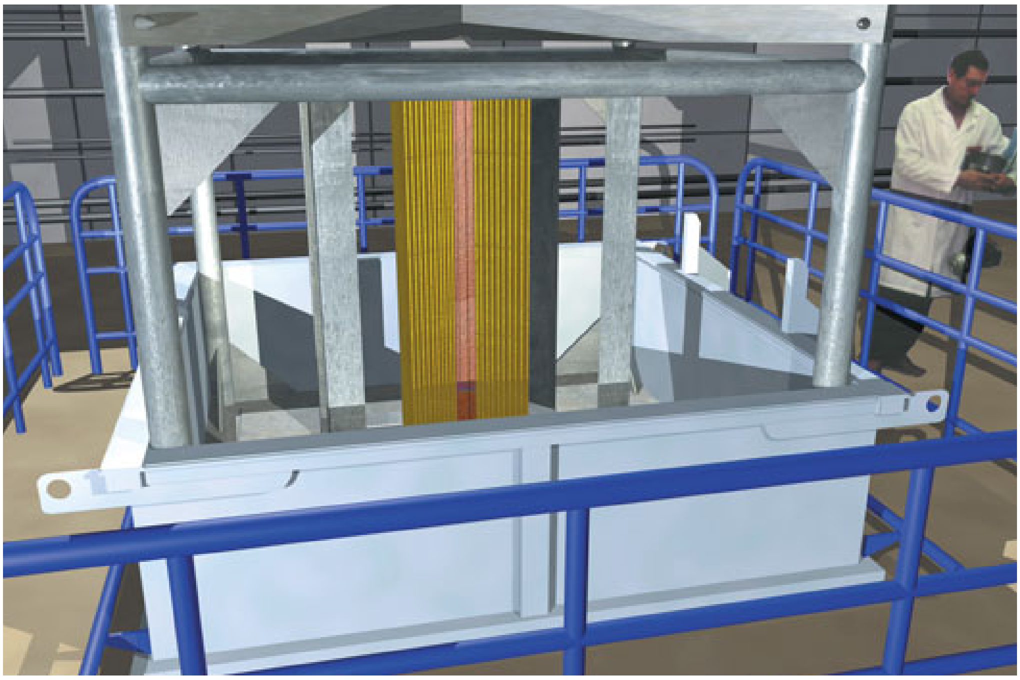
The dosimetric results obtained from Crac and Silène must be considered as representative of the dose that personnel may be exposed to when a criticality accident occurs in a uranyl nitrate solution, with the divergent system situated in a concrete room. For sources with very different configurations (variable concentration and cylinder diameter) there is no proportional relationship between the number of fissions and the dose emitted because the **leakage** radiation depends on the source characteristics. The maximum dose value observed in the Crac and Silène tests is $5.8 \cdot 10^2$ **grays** (Gy) at 1 meter from the axis of the source for 10^{18} fissions with a cylinder of 0.3 m diameter and a concentration of 80 g/l. As an indication, the doses produced in the initial power peak on Silène for 10^{17} fissions at 1 meter from a "core" of 40 liters of solution of uranyl nitrate are 20 Gy for the neutron dose (tissue Kerma)(2) and 25 Gy for the gamma dose.

In a criticality accident, there is therefore no direct proportional relationship between the number of fissions and the dose emitted for different sources. The nature of the fissile medium, its dimensions and its geometry play an essential role in the assessment of the exposure risks. These values do show that the exposure risk is a major risk in the event of any criticality accident and that the associated doses may be fatal for personnel working in the immediate environment of the equipment concerned.

Detecting criticality accidents

The aim of a criticality detection system is to trigger an alarm as quickly as possible so that personnel can be evacuated as soon as the accidental excursion begins, thus limiting the exposure

(2) The Kerma (*Kinetic energy released in materials*) is the sum of initial kinetic energies of all the charged particles released by uncharged ionizing radiation (neutrons and photons) in a sample of material divided by the mass of this sample. It is expressed in grays or a submultiple of grays.



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Artist's impression of the core of the "Appareillage B" facility at Valduc, used for subcritical experiments to validate criticality safety calculation codes.


EURYSIS

The component parts of the Edac criticality detection and alarm system.

from Crac and Silène, in 1976 the CEA designed a criticality detection and alarm system called Edac based on a monitoring unit with at least three criticality sensors connected to it. The criticality alarm is only given if at least two of the three sensors send an alarm signal to the main unit. This signal is only generated if two conditions are met for the sensors: a predetermined dose has been exceeded, generally set to 25 µGy, and a **dose rate** threshold set to about 10 mGy/h has also been exceeded. The system is based on measurement of the total radiation dose using two scintillators(3) which are sensitive to neutron and gamma radiation respectively. Tests carried out in Silène showed that the system can be used to detect all accident types: power excursions with rapid kinetics and those with slow kinetics (from 100 µs to several minutes for the period of the first power peak). They also showed that the detectors give a dose response proportional to the total dose in a mixed field of neutron and gamma radiation and that they remain reliable even after being subjected to the high doses delivered by the initial peak, so that the development of the accident can be monitored after the alarm has been triggered.

Because the Edac system can record and monitor the progress of the accident using criticality sensors, in particular using a console placed outside the evacuation zone, its contribution may be essential for managing the post-accident situation and any remedial action.

No comparison with releases from a hypothetical reactor accident

An experimental program has been carried out at Silène to determine the release rate of fission products emitted during a criticality accident in an aqueous fissile medium, with conditions going up to boiling point of the solution to maximize this release. The study focused on elements with a radiological impact on humans and the environment. The products concerned can be classified according to their physical and chemical properties into three categories (noble gases, aerosols and isotopes of iodine, bromine and ruthenium). The program of experiments allowed us to draw various lessons (see box 2).

Table 2 shows the quantities of fission products released from the solution along with the consequences of these releases under wind at 500 meters in normal diffusion conditions with a 5 m/s wind for several "cooling times" of the fission products at the moment of release. These values show that the risk of exposure for the population following a criticality accident are low and that there is no comparison with the potential consequences of a reactor accident.

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(3) Sensitive parts of a particle detection apparatus based on the property that some materials have of emitting light under the effect of radiation.

radionuclide family	maximum activities released (Bq) and doses (mSv) for 10^{18} fissions (about 0.4 mg of fissioned uranium) as a function of the cooling time		
	10 seconds	1 hour	10 hours
noble gases, halogens (excluding iodines) and aerosols (Bq)	$3.0 \cdot 10^{14}$	$2.5 \cdot 10^{13}$	$9.0 \cdot 10^{11}$
iodines (Bq)	$1.7 \cdot 10^{12}$	$1.8 \cdot 10^{12}$	$2.6 \cdot 10^{11}$
inhalation at 500 m in DN5 (doses engaged at the thyroid and in the lungs) (mSv)	0.12	0.20	0.12
direct irradiation by the plume at 500 m in DN5 (mSv)	0.50	0.13	0.004

Table 2. Quantities of fission product released in a criticality accident and the consequences of these releases under wind at 500 m in normal diffusion conditions with a 5 m/s wind (conditions referred to as DN5) for different "cooling times" of these products at the moment of release.

quences of a reactor accident.

Improved risk evaluation

Since 1967, multidisciplinary teams of physicists, dosimetry specialists and radiobiologists have acquired unique skills in dealing with criticality accidents thanks to the experiments carried out in Silène. The research has yielded improved knowledge of the physics of the accident, detection, dosimetry and the effects of emitted neutron and gamma radiation and of the release of radionuclides into the environment. These results contribute to an improved evaluation of the risks associated with a criticality accident as well as the implementation of intervention measures and crisis management.

More generally, the feedback from accidents that have occurred world-wide reinforced by the French experimental results bring us three main lessons. First, the energy released in a criticality accident is generally limited, since the maximum accident known in fuel cycle facilities involved $4 \cdot 10^{19}$ fissions, less than a hundredth of the energy released in the Chernobyl accident. The consequences for the environment are also slight, with gas releases being very


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Testing Edac accident detection system sensors around the core of the Silène reactor.

limited compared to those that might be caused by a reactor accident. On the other hand, the risks of irradiation for personnel working near to the equipment concerned are high and may lead to lethal doses.

The Tokaï Mura accident also showed that the consequences may be of another dimension altogether in terms of media coverage and the acceptability of the nuclear risk. It is therefore important to be able to detect *a priori* any possible incident situation, in spite of all the criticality risk prevention precautions already taken, and to provide the means necessary to stop neutronically the accident process, in addition to the intervention measures already planned. ●

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Release of fission products: the Silène results

The main results of the experimental program conducted using the Silène reactor to determine fission product release rates during a criticality accident in an aqueous fissile medium concern three product families:

Noble gases: (xenon and krypton): the release rates are practically 100% for gases at times greater than a minute. They vary between 10% and 50% for times ranging from a few seconds up to one minute, and are of the order of 10% for very short times (less than 2 seconds).

Aerosols: at boiling point a particle size of around 0.1 µm is observed while at a lower temperature it may be of the order of $5 \cdot 10^{-3}$ µm. Trapping these aerosols on filters may induce a significant local risk of irradiation.

Iodine, bromine and ruthenium isotopes: for the iodine isotopes, the importance of the chemistry of the medium has to be emphasized (acidity and initial abundance of iodine by weight, for example when dissolving spent fuel). In the Silène experiments, the maximum release rates observed for an acidity of around 2N were well below 1% when the solution was boiling. A release rate of 10% could be measured at the level of the core in the case of very low solution acidity and a high initial iodine abundance in the solution. For bromine and ruthenium isotopes, the other volatile products, the maximum emission rates were estimated at 20% and 1% respectively.