

Fission, Critical Mass and Safety – a Historical Review

Geoff Meggitt
2 Dingle Bank Close
Lymm, Cheshire
WA13 0QR
England

Abstract

From the discovery of fission the notion of a chain reaction in a critical mass releasing massive amounts of energy haunted physicists. The possibility of a bomb or a reactor prompted much of the early work on determining a critical mass but the need to avoid an accidental critical excursion during processing or transport of fissile material drove much that took place subsequently.

Because of the variety of possible situations that might arise, it took some time to develop adequate theoretical tools for criticality safety and the early assessments were based on direct experiment. Some extension of these experiments to closely similar situations proved possible but it was not until the 1960s that theoretical methods (and computers to run them) developed enough for them to become reliable assessment tools. Validating such theoretical methods remained a concern but, by the end of the century, they formed the backbone of criticality safety assessment.

The paper traces the evolution of these methods, principally in the UK and USA, and summarises some related work concerned with the nature of criticality accidents and their radiological consequences. It also indicates how the results have been communicated and used in ensuring nuclear safety.

Introduction

The discovery of fission and the possibility of a chain reaction based on it were exploited more quickly than most scientific discoveries and certainly with more terrible results. The key to this was the estimation of the critical mass needed for a chain reaction – and particularly the critical mass for a bomb. Most of the physical principles were established quickly in the wartime environment through the development of the weapon and through the design of the reactors that were to produce plutonium. This paper spends some time looking at the bomb development to see how the understanding of the critical mass developed through experimental and theoretical investigations. However, the main interest here is in safety when the key question is: how close to a critical configuration are we? The evolution of the methods of criticality safety that have been developed to answer this question is the central topic of this paper

Given the diversity of situations in experimental facilities, reactor management systems, industrial-scale reprocessing plants and fissile material transport, the techniques from the early days proved inaccurate and their results demanded great caution in application. The history since has been one of improving the assessment techniques to give greater confidence and remove excessive conservatism in design. Since the 1960s much of this effort has gone into developing and validating computer codes and the histories of the two principal Monte Carlo codes (KENO and MONK) with wide international use have been traced in some detail. Of course, Monte Carlo codes have emerged from other nations –notably France - and alternative deterministic computational techniques have been used by many. Some of these are outlined but, given the complex and dynamic nature of the field, it would have been impossible to do justice to all of them without the paper becoming a long and elaborate list.

Safety in criticality arises not just from improved assessment but from better understanding of what might cause an accident and how big that accident might be. These lead to principles for design and operation of facilities with fissile materials. While the principal aim is to trace the evolution of assessment techniques, accident consequences and management principles are briefly mentioned for completeness.

Criticality Principles

When a uranium nucleus fissions, on average, 2.4 neutrons are emitted with an average energy of 2MeV. At this energy each neutron has three possible fates: it may collide with a nucleus and lose energy by inelastic scattering, it may be absorbed and lost or it may cause another fission.

If we first consider natural uranium, with approximately 0.7% U-235, then the most likely fate, happening on about 80% of occasions, is inelastic scattering by a U-238 nucleus. Of the remaining 20% of neutrons almost all will cause fission in U-238 with just 0.3% will causing fission in U-235. The fissions will not produce enough neutrons to sustain a chain reaction so we should look at the scattered 80% . These will lose a substantial fraction of their energy and further inelastic collisions are still the most likely fate but, because the neutron energy is now below the threshold for fission of U-238, U-238 fission will not happen. However, because U-235 is more susceptible to fission at this energy and in spite of its low content, U-235 fission becomes more important with about 15% neutrons causing its fission. This is still not a high enough proportion to cause a chain reaction and the dominant effect is that the neutrons continue to lose energy by collisions. This continues until the energy falls to about 1keV when a new process becomes important. This is resonance capture when the neutrons are most likely to be absorbed by the U-238 nucleus and therefore to be removed completely

from the system. A few neutrons escape capture, and they do reach an energy region where U-235 fission is much more likely than any other process. However, in the U-238 – dominated natural uranium, their number is so small that a chain reaction cannot be sustained.

If the uranium is enriched and the U-235 content increased to 50%, the picture changes dramatically because at energies between 0.3 and 2MeV the probability of U-235 fission becomes about equal to that of non-elastic scattering. Since an average of about 2.4 neutrons are released on each fission, there is now the possibility of a chain reaction. This is fast fission and is the basis for the fission bomb and the fast reactor.

An alternative approach route to a chain reaction is to get the energy of the neutrons below the U-238 resonance capture region before too many are lost and this depends on the presence of a moderator. A moderator is a material composed of lighter elements such as hydrogen or carbon with low neutron absorption characteristics. When neutrons collide with nuclei of these lighter elements they lose a high proportion of energy in each collision and their energy is quickly reduced through the resonance region. In fact the energy falls until it is comparable with the thermal energy of the nuclei of the moderator. In this region the probability of fission of U-235 is at its highest and, provided the moderator is well-chosen, a chain reaction becomes possible even with natural uranium. This is the basic process of the thermal reactor and makes possible a chain reaction with little or no enrichment.

These factors are summarised for a chain reaction in a thermal reactor by the Four Factor Formula to give k_{∞} , the infinite multiplication factor:

$$k_{\infty} = \epsilon p f \eta$$

where

ϵ is the fraction of fission neutrons slowing down below 1MeV(the fast fission factor)

p is the resonance escape probability (the fraction of these that escape resonance capture)

f is the fraction of neutrons absorbed in fissile material rather than elsewhere (the thermal utilisation)

and η is the number of neutrons produced per neutron absorbed in the fissile material

If a chain reaction is to take place in an infinite medium then $k_{\infty} \geq 1$.

In practice media are not infinite and there is another way for neutrons to be lost from the potential chain reaction; they simply escape from the surface of material. If the material is spherical then the larger the radius, the less likely it is that neutrons will escape before they go on and cause a fission. The radius of the sphere just large enough to allow the chain reaction to progress in a particular material is its critical radius; its mass is the critical mass of the sphere.

Perhaps the most important parameter in criticality is the effective multiplication factor k_{eff} (called k effective):

$$k_{\text{eff}} = k_{\infty} \times \text{probability of neutrons not escaping from the system}$$

When $k_{\text{eff}}=1$ the non-infinite material will just be critical and sustain a chain reaction. Because of the different probabilities of neutron escape different geometric shapes of the same material will have different critical masses. The critical mass will also depend on the surroundings because these may act as a reflector and direct escaping neutrons back into the fissile material. They may also introduce a moderating effect. These factors will reduce the

amount of fissile material needed to make a critical mass. So, for example, a sphere of U-235 in water will have a lower critical mass than one in air.

So there are two distinct aspects to assessing whether criticality can occur in uranium or any other fissile material such as plutonium-239:

- Whether the material is intrinsically able to support a chain reaction
- Whether there is enough of it, given its environment, to limit neutron escape and actually support one

The first of these is not generally difficult to deal with: given the data some simple arithmetic will suffice to give an answer. The second presents a much greater challenge and this is the central theoretical problem of criticality safety.

There is one further complication that is important for reactor control and criticality accident evolution: the time spectrum of fission neutron emission. While most fission neutrons are emitted very promptly, a small fraction are released up to a minute later. Although this delayed fraction is small it is important in reactor control because, with careful design, it gives time for mechanical systems to control the chain reaction and the reactor power. It is also significant in criticality accidents. If a system is critical with just the prompt neutrons (prompt critical) the excursion evolves extremely quickly and is exceptionally violent. If the system is not made critical by the prompt neutrons but is once the delayed neutrons arrive (delayed critical) then, while the energy released may still be enormous, it will happen over a longer period.

Nuclear Fission and the Chain Reaction

Nuclear fission was seen by Enrico Fermi in the mid-1930s but mistaken for the creation of transuranic elements (1) and was actually discovered by Hahn and Strassmann in late 1938 (2,3,4). It was explained by Lise Meitner and Otto Frisch shortly after (5) and by the middle of February 1939 it was confirmed by experiments in Copenhagen, by Frederic Joliot's team in Paris and in four US laboratories. However, even before this, another possibility was being discussed: Fermi's suggestion that neutrons might be emitted when fission took place leading to a possible chain reaction.

The possibility of a nuclear chain reaction had been conceived in London by Leo Szilard in 1934 and he had filed a patent (6) for it. The basis of this was not fission but that when beryllium was bombarded with neutrons, two neutrons were released for every one absorbed. In the patent he made some calculations of the critical thickness of a shell of beryllium using diffusion theory and the limited amount of information then available. His experiments on this chain reaction failed but once made aware of fission he recognised the possibility for a *fission* chain reaction provided enough neutrons were emitted on splitting. By early March 1939 he had shown that neutrons were indeed released and it was soon clear that there were, on average, around 3 per fission. This was encouraging but there was at the same time one problem. Bohr had suggested on theoretical grounds that U-235 was the likely candidate for fission and by March this was confirmed experimentally in the USA. This wonder isotope formed only 0.7% of uranium.

Moderation had been discovered by Fermi in 1934. At the same time he found that slow neutrons were much more likely to be captured by atoms than faster ones. By 1939 the Joliot team in Paris realised that heavy water, because of its low neutron absorption compared with normal water, could be used as a moderator for a reactor (leading to the smuggling of most of the French team and its stocks of heavy water to England). Also in 1939, Szilard and Fermi showed that it was possible to achieve neutron multiplication with a uranium lattice in very

pure graphite. While these developments were to prove crucial to the development of nuclear reactors, the early efforts at understanding the nuclear chain reaction related to unmoderated systems.

The first published estimates of the critical mass necessary before a chain reaction could be sustained came from Joliot's Paris group where, in the summer of 1939, Francis Perrin estimated (7) that this corresponded to a ball of uranium oxide about 3m in diameter with a mass of about forty tonnes. The calculation was not dissimilar in principle to those developed later, and used still for reactor calculations, based on diffusion theory. However, it was not supported by accurate data for cross-sections. The calculations showed that the critical mass could be reduced to about 12 tonnes if the sphere were surrounded by an element that did not absorb neutrons. A more general transport theory treatment of the critical mass question, which removed the condition that the mean free path between collisions was much less than the diameter of the sphere, essential for diffusion theory, was quickly developed by Rudolf Peierls in Birmingham (8) and although he gave no estimates of critical mass, his techniques were later to have momentous consequences. For the moment the critical mass of uranium was estimated to be of the order tons and Otto Frisch in early 1940 (9)(10) thought that, even with uranium enriched by ten times, a chain reaction would not be fast enough to cause an explosion. He concluded: "our progressing knowledge of the fission process has tended to dissipate these fears (*of fission bombs*) and there are now a number of strong arguments to the effect that the construction of such a super-bomb would be, if not impossible, then at least prohibitively expensive".

In February 1940 the prospect of nuclear weapons changed dramatically when Frisch and Peierls calculated the critical mass of pure U-235 using the Peierls method. They guessed the fission cross-section (rather too large a value in fact) and obtained the astonishing result that the critical mass was only about 600 gm. Peierls then calculated the staggering power of the explosion that might result. They put their thinking in the celebrated Frisch-Peierls Memorandum leading to the setting up of the so-called MAUD committee to review its implications.. By March 1941 there was finally a reliable estimate for the fission cross-section for U-235 and, from the work it had sponsored, the MAUD committee was able to conclude in Part 1 of its July 1941 report that "the scheme for a uranium bomb is practicable and likely to lead to decisive results in the war". They estimated that a bomb containing 25lb of U-235 would have the destructive effects of 1800 tons of TNT and would release large quantities of radioactive material making bombed areas dangerous to life for a long period. They recommended collaboration on experimental work with the USA and thought that the U-235 separation plant that would be needed might be located there. In Part 2 of the report they considered the use of nuclear energy for power and considered it promising but not worth considering until after the war. However, because of the weapon potential of Pu-239 (found in the USA mid-1940), they considered that some work should continue on reactors.

Criticality and the Bomb

In the USA nuclear work had progressed on a number of areas relevant to the bomb but this was "in the spirit of scientific curiosity". By mid-1941 this was beginning to change anyway but the MAUD report brought a sense of urgency that had been lacking. Even so, the USA was probably more interested in the energy possibilities than uranium bombs until 7 December 1941, when Japanese planes attacked Pearl Harbour and war was declared. The initiative then passed quickly to the US and Britain's main contribution for the duration of the war was in supplying individuals to the American project. The French scientists who had made much of the original running were to be effectively side-lined as they were exiled first in England and then in Canada.

It was understood by the end of 1941 that there were two practical routes to fissile material for a bomb: (1) the separation of the isotope U-235 from natural uranium and (2) the production of plutonium in a reactor and its separation from other fission products. The research work necessary to realise them was took place across the US (with much of the reactor work at the Metallurgical Laboratory in Chicago), and the most memorable milestone was probably Fermi's achievement of the first man-made criticality at Stagg's Field, Chicago, in December 1942.

However, the most significant event was probably the creation of an organisation in September 1942 which was capable of meeting the unprecedented scientific and engineering challenges and delivering a weapon: the Manhattan Project. It concentrated the bomb development team as Project Y at Los Alamos, New Mexico and delivered two vast factory sites to produce the materials. The plutonium production reactors and the associated separation plants were built at Hanford in the north west USA and three separate isotope separation plants to produce enriched uranium and a plutonium separation plant were constructed at Clinton (later Oak Ridge), Tennessee. The project depended on criticality: for the weapon to work, for the reactors to produce plutonium and to avoid accidental criticality excursions in the experimental and production facilities. The early progress in criticality physics came from the weapon work at Los Alamos

Manhattan Project Theory

In a series of five lectures between 5 and 14 April 1943, Robert Serber summarised (11) what was then known of all aspects of the bomb. He reviewed the available nuclear data and gave an analysis, based on diffusion theory, of the critical size of a naked sphere of fissile material. He calculated that the critical radius would be given by

$$R_c^2 = \pi^2 D \tau / (v-1)$$

Where D is the diffusion coefficient, τ the mean time between fissions and v (**see note below**¹) the number of neutrons produced per fission. For U-235 metal the calculation gave a critical radius of 13.5 cm and a critical mass of 200 kg.

Serber recognised that the simple diffusion theory did not apply because the mean free path at 5 cm was comparable with the dimensions of the system. He quoted the result of a "more exact diffusion theory" as leading to a critical mass of 60kg. He later indicated that this difference was largely accounted for by better accounting for the effects at the sphere boundary.

In the practical weapon the fissile core was to be surrounded by a thick uranium metal shell (the tamper) which would reflect some of the escaping neutrons back into the core and also hold the core together for the briefest of times while the chain reaction built up. With a uranium tamper "best available calculations" gave R_c as 6 cm with a critical mass of 15 kg for uranium. Serber estimated the critical mass of a tampered sphere of plutonium as 5 kg.

Diffusion theory continued to form the basis for estimating the weapon critical mass throughout the project but it was extended in the most complex ways to account for the fact of a spectrum of neutron energies, the role of inelastic scattering and the slowing down process, and the anisotropic nature of the collisions. The need to handle a distribution of neutron energies led to the introduction in late 1943 of a multi-group approach in which neutrons were treated as having energies in three or four discrete bands. Also, since one model of the weapon involved a core of uranium hydride, the complexities introduced by hydrogen scattering had to be addressed. Of course, this was all complicated by the fact that the weapon was blowing itself apart while the chain reaction built up.

¹ Editorial note: this and the same symbol in the equation should be a Greek "nu"

The calculations for carbon moderated reactors were somewhat more straightforward because Fermi's age theory, based on diffusion and continuous slowing-down, could be used. However the experimental Water Boiler facility demanded a three energy group approach because of the large discrete losses of energy that occurred in neutron collisions with hydrogen nuclei. Diffusion theory was also used in plant criticality calculations and an example can be found in a report written by Edward Teller with Oak Ridge and Chicago staff (12).

Diffusion theory was not just useful for the prediction of critical mass. It could also extend the value of experiments, a procedure best explained through the concept of "buckling". The "material buckling" (B_m^2) depends only upon the properties of the nuclear material under consideration and is defined as:

$$B_m^2 = (k_\infty - 1) / M^2$$

Where k_∞ is the infinite multiplication factor in the material and M^2 is the mean square distance from the point where a neutron is released in fission to the point where it is absorbed in an infinite medium. M is known as the Migration Length; M^2 as the Migration Area. The material buckling is therefore a measure of the net production rate of neutrons combined with an indication of how far they spread from their origins.

From the diffusion calculation comes a factor that depends only on the geometry of the fissile assembly: the "geometric buckling" (B_g^2). For a bare sphere of radius R , for example:

$$B_g^2 = (\pi / (R + \delta))^2$$

Where δ is a small correction factor, the extrapolation length, which is usually determined experimentally

Diffusion theory says that, when the two bucklings are equal, the sphere becomes critical i.e. when

$$B_m^2 = B_g^2$$

$$(k_\infty - 1) / M^2 = (\pi / (R + \delta))^2$$

A key factor is the ratio between M and R . If M is large compared with R then the neutrons must wander a large distance before causing another fission and this may take them out of the sphere before they do so.

The term "buckling" is a strange one. It derives (13) from the change in shape of the neutron flux profile in a material as it passes criticality. Buckling or Shape Conversion is a method still recommended for preliminary or check calculations and proved, as will be seen, a useful way to extend the applicability of experimental results.

For example, the geometric buckling of a cylinder is calculated as:

$$(2.405 / (r + \delta))^2 + (\pi / (h + 2\delta))^2$$

where h is the height and r is the radius. If these are the critical dimensions known from some experiment or calculation on the cylinder then the critical radius R of a sphere of the same material will be given by equating the two geometric bucklings.

$$(\pi / (R + \delta))^2 = (2.405 / (r + \delta))^2 + (\pi / (h + 2\delta))^2$$

The technique probably originates from Paxton (16).

These methods, developed before and during the Manhattan Project, when combined with criticality experimental data, provided the foundation for criticality safety in the early nuclear programme.(14)

Experiments

While calculations could be made, there was considerable uncertainty about their accuracy and they could deal with only relatively simple shapes. In the early years of criticality safety direct measurement was the basis for most decisions and a considerable body of data for many materials and configurations was built up.

The aim in most such experiments is to approach criticality closely enough to be sure where it is but to avoid it and the damage that might be caused by the associated energy release. The general procedure involves measuring, with a suitable neutron detector, the neutron multiplication (m) produced in the system from a fixed neutron source. At criticality m will become infinite so if $1/m$ is plotted as the system is brought incrementally towards criticality it will be found to tend towards zero. It is then possible, at least in principle, to extrapolate to zero and find the critical point.(13)

Some early experiments on enriched U-235 have been described by Reider (15). The Water Boiler was an assembly in which enriched (14.7%) uranyl nitrate solution was pumped into a sphere to investigate the approach to criticality. The Dragon Experiment, dating from early 1945, dropped a piece of fissile material through a hole in another piece to give a 1/100 second burst of prompt criticality. The "Drop Leaf" assembly was used just after the end of the war and allowed a reflector to be placed remotely around the enriched uranium. The Moveable Table, from 1946, could be used to move two pieces of material together to reach criticality. This is just a selection: there were many additional tests at Los Alamos for safety reasons including a number on the effects of flooding. Some of these experiments were directed solely at the bomb design but they began to give general information on criticality.

Arrays of fissile material

Arrays are important for storage and transport of fissile material and they present special problems because, while the individual units may be safely sub-critical, when they are put close together they may reach criticality. This is because when neutrons escape from one unit they may enter another and this interaction may just tip the balance of criticality.

The need to understand arrays meant that they were the subject of many experiments from the 1940s onwards at several US facilities (17). However, such experiments are expensive and some (relatively) simple methods were needed for evaluating proposed arrays and eliminating clearly unsafe ones. Even when computer codes became available such methods could absorb their results and make useful guides. Several methods, each with various formulations, were developed and they fall into two broad classes: those that seek correlations from experimental data and those that start from a more fundamental basis. The four methods in the first category are the surface density, NB_N^2 , density analogue and solid angle methods. The Interaction Parameter method is more fundamental.

The **Surface Density** method was originated by Paxton at Los Alamos in the 1950's and involves imagining the array of fissile material projected onto a surface, merging into a slab

and then flooding with water. In one formulation the array is regarded as safe, based on experimental results, if the surface density of the slab σ

$$\sigma = 0.54 \sigma_0 (1 - 1.37f)$$

where σ_0 is the surface density of a critical water-reflected infinite slab of the same material and

f = mass of array unit/critical mass of un-reflected sphere of same material

The **Limiting Surface Density** (NB_N^2) method, developed by JT Thomas at Oak Ridge in the late 1960s (18) can be seen as an extension of the Surface Density method that takes explicit account of the finite number of elements in a real array. It is based on the observation that the neutron non-leakage fraction from the whole array, which must be the same for different **critical** arrays of the same units, can be represented by the expression:

$$1/(1 + NB_N^2)$$

where N is the number of units and B_N^2 is a geometric buckling independent of the fissile material involved. The material composition is brought in through a simple empirical relation based on experiments and Monte Carlo calculations. Combining the geometrical and material factors defines the critical conditions.

The Density analogue method, proposed by Paxton in 1975, defines limits that are independent of the storage arrangement and is based on regularities in data from critical arrays. In one formulation it gives the sub-critical limit for storage arrays of any shape as N units as:

$$N = (2.1\sigma_0(1 - 1.37f)/m)^3 V^2$$

Where V is the volume occupied by a unit in the array, m is the mass of a unit and N is the total number of units.

The **Solid Angle** method was developed at Oak Ridge in the 1950's as a fairly quick method for evaluating interactions between individual fissile units. It depends on the observation that, with no absorption or reflection, the probability of a neutron emerging from one unit reaching another one depends on the solid angle subtended by the target unit at the originating one. The method, as described by Knief (19), is based on calculating the total solid angles subtended at the centre of one unit by all the other units. Then the solid angle (in steradians) for that unit must be less than or equal to:

$$\Omega_{\text{allowed}} = 9 - 10 k_{\text{eff}} \text{ where } k_{\text{eff}} \text{ is the effective multiplication factor for that unit}$$

This is then repeated for all the units and all of them must satisfy the condition. There are some restrictions on the method including that the k_{eff} of individual units must be less than 0.8 and that no solid angles may be greater than 6 steradians. With these restrictions the method is conservative – and some times very so.

The **Interaction Parameter** method embodies a more fundamental approach and (20) originated with AF Thomas at AWRE Aldermaston in the UK in the 1950s and was subsequently developed into a powerful tool. It starts by considering the neutron balance of the array and notes that the condition for criticality is:

$$F_i = \sum_j q_{ij} F_j \quad (i=1,2,3,\dots,n)$$

j

Where F_i is the number of neutrons leaving unit i per unit time and q_{ij} (the interaction parameter) is the number of neutrons leaving unit i as a result of one neutron leaving unit j . It is assumed that the interaction between i and j is not affected by the presence of the other units.

Then a sufficient condition for sub-criticality is that :

Maximum value $\sum_j q_{ij}$ for all i should be less than 1

The key to the method is then efficiently and conservatively estimating the q 's. This may come from direct experimental work on a single array element, measuring its response to a neutron source representing another unit. It may also be estimated from the geometry of the array and the surface multiplication factor of individual units. The method and some applications are described in great detail by Thomas and Abbey (20).

Later Experiments

The Oak Ridge Criticality Measurements Laboratory performed many experiments on U-235 and U-233 until its closure in 1975. Some of the early ones addressed criticality limits for particular facilities but others (including the array studies mentioned below) made a vital input to code validation. The Battelle (PNL) facility, the Plutonium Critical Mass Laboratory, was designed specifically for experiments on plutonium and has conducted experiments on thousands of different system of many kinds. (13)

The most comprehensive measurements have probably been made at the Los Alamos Critical Assemblies Laboratory. The laboratory has experimented with U-235, U-233 and Pu-239 in all physical forms. The facilities there (Godiva, Jezebel, Flattop, Comet, Sheba and others) provided basic data for design, codes and criticality alarm detectors(13) and continued to do so under Lawrence Livermore management.

Measurements like these answered the immediate problem but many were not taken near enough to criticality to give data supporting the computer calculations that were beginning to be developed in the early 1960's. Higher quality experiments were made at ORNL in 1963-4 on U(93.2) metal cylinders with various reflection and interspersed materials and these gave the first precise data for metal arrays. They were very important for establishing standards for storage as well as providing an input to computer codes. Good data for plutonium metal arrays came from the Lawrence Livermore series mentioned above.

Rather smaller critical measurement programmes occurred in the United Kingdom and France. The UK programme included high-enriched uranium and plutonium solution measurements at Dounreay in the 1950's and 1960's, and the SCAMP programme at Aldermaston in the 1970s on mixed plutonium-uranium. There were also numerous low energy reactor experiments performed at Harwell and Winfrith (after the DIMPLE reactor was moved there from Harwell) as part of data gathering for the UK reactor development programme and at the Atomic Weapons Research Establishment, Aldermaston for more basic research. The UK programme, which effectively ended in the 1980s, has been described by Simister and Clemson (21).

The French have had a much greater sustained commitment to criticality experimentation. Their first experiments (ALECTO) at Saclay were concerned with U-235, U-233 and Pu-239 nitrate solutions. They then opened the Station de Criticit  at Valduc in 1963 and conducted

experiments on high enrichment uranium and plutonium solutions including the effects of neutron poisons and interaction effects.

Computer Methods

As criticality data accumulated and computers became more powerful the emphasis changed from collecting more data to developing better models for criticality calculation – although these models had to be supported by benchmarking experiments.

Multi-group Diffusion Codes

With the increasing availability of more powerful computers it became practicable to carry out more refined diffusion calculations using multigroup diffusion theory - as had been done at Los Alamos for the weapon. The first recorded application (22) was by Ehrlich and Hurwitz in 1949 at Knoll's Atomic Power Laboratory using the IBM 604 Electronic Calculating Punch. Similar one-dimensional multigroup calculations were undertaken on IBM Card Programmed Calculators at many AEC labs in the early 1950s. However, at about this time stored-program computers were becoming available, notably the ORACLE at Oak Ridge and the MANIAC I at Los Alamos and these allowed the calculations to be made more quickly and permitted features like automatic searches for the most critical configuration of a system. With the beginning of the nuclear power industry, such codes were a major development in reactor design and, as computing power and fast access memory increased, more detailed nuclear data could be made available to them(22). However, they were designed for reactor situations with high moderation, low leakage and a minimum of materials with high absorption losses and these were the conditions where the diffusion approximation worked best. Criticality safety often involves low moderation, high leakage and the use of materials with high absorption cross-sections – just where the diffusion approach was least reliable. Alternative techniques were required.(14)

Monte Carlo Codes

The Monte Carlo approach tries to simulate an actual process that depends on chance with a computer programme that uses a random number generator to select the options for the process. It is applicable to fission because the interaction processes of neutrons with nuclei are random ones. It has been claimed (23) that Enrico Fermi used a hand version of the technique in the mid-1930s to help him in work on neutron diffusion but its serious application to physical processes had to await the digital computer. ENIAC, one of the first computers, had been constructed towards the end of the war for ballistics calculations and had been used for thermonuclear problems associated with the Super (the hydrogen bomb). It appears to have been John Von Neumann who suggested, in 1947, that ENIAC could be used for a Monte Carlo solution of the neutron transport problems associated with a bomb and it was agreed that this would be done once ENIAC had been moved 200 miles to its permanent base in Maryland. While this move was being made (and there were those who thought its 18,000 valves would never work again) Fermi devised a simple mechanical analogue device (24) (a sort of trolley later christened the *Fermiac*) that could be used to trace out the path of neutrons on a scale drawing of the fissile assembly. Externally-generated random numbers determined whether the neutron was fast or slow and the direction and distance travelled after each collision. It could account for boundary crossings and was in use for about two years.(23)

However, the future lay with computers and when ENIAC survived its move it was used successfully for the calculations on neutron transport, handling complex geometries and a

realistic neutron velocity spectrum. Other workers ran other problems and by mid-1949 there was enough evidence and interest for a conference in Los Angeles(23).

The principle of Monte Carlo is easy enough to understand but, in practice, there are many technical issues to address to get a practical code:

the representation of the cross-section data as a function of neutron energy

the cross-sections of relevance – particularly those for resonance capture - vary with neutron energy and this must somehow be factored into the code. Both point and group representations have been used. A significant development was the provision of cross-sections in six and sixteen groups for fast and intermediate energy neutrons by Hansen and Roach in 1961 (25). The detail of cross-section data libraries is beyond our scope (although their increasingly international character is touched upon later) but they are now available with such fine structure that the distinction between point and group data sometime becomes blurred.

the representation of the geometry

The geometry of systems can be quite complex – far from the simple spheres, cylinders and slabs. To be useful for criticality assessments it must be possible to construct the required geometry of the system in a way that is straightforward and readily checked.

the neutron tracking system

The simplest Monte Carlo routine would try to simulate directly the behaviour of each neutron: it would be born from a fission, collide a number of times and then finally be absorbed or escape from the system. In practice this is wasteful because the history of neutrons that leave the system, which contains valuable information, is lost. Rather than losing it, a more efficient method is to continue to track the neutrons and adjust the weight given to them in the calculation.

variance reduction

suitable biasing techniques allow the user to steer the calculation so that neutrons are guided towards regions of importance to the calculations. Such non-analogue strategies can reduce calculation times and/or statistical errors.

The first general Monte Carlo code (26) for neutron problems was the O5R code from Oak Ridge, designed to run on the ORACLE mainframe computer. Initially conceived as a reactor code, it used pseudo-point neutron cross-sections and could deal with a range of geometries. However, the complexity of the cross section data made it difficult to validate and the geometry was difficult to set for complicated arrangements. It was difficult to use with confidence for criticality calculations.(14)

These shortcomings led to the development of the KENO code at Oak Ridge beginning with Elliott Whitesides' work in 1963. He started with a small Monte Carlo testing programme and steadily expanded this to make a complete criticality assessment code with a geometry package. This has continued to be enhanced by Whitesides, his co-workers and successors and remains one of the world's principal assessment codes today.

By the end of the 1950's the civil criticality safety code development effort in the UK was concentrated around Ed Woodcock at United Kingdom Atomic Energy Authority (UKAEA) Risley. Woodcock had been drawn from the Meteorological Office for his expertise in

diffusion theory and was responsible with his team for the GEM (27)(28)(29)(30) code. This ran on the IBM 7090, first appeared in 1962 and was designed specifically for criticality applications. It had an unusual neutron tracking scheme and, while easy to use, its results (the British rather disliked the k_{eff} concept at this stage and expressed the code predictions in a more complex way) were rather difficult to interpret. However, it had the capability to perform calculations on assemblies of several materials made up of spheres, cylinders and regular parallelepipeds. This geometric capability could be refined with the so-called HOLE routine which, adapting an approach developed by John von Neumann (31) to handle the problem of voids in fissile material, introduced a notional collision that had no effect on the neutron. This reduced the problem of tracking the neutron across boundaries to the much simpler one of knowing the material in which the interaction, notional or otherwise, occurred. The cross-section data capability was high but, in practice, the data were simplified and approximate. (14).

The successor to GEM in the UK was MONK (32) originally developed in the 1970's. This, like KENO, has been updated and enhanced regularly since and is the first-choice code today in the UK. MONK originally used point data and this continues to be the preferred option for most users. However, group data was introduced in MONK-G in the mid-1970's and this was later consolidated as the better-known MONK-5W. Both KENO and MONK have become much more flexible codes with options on the nuclear data available. MONK offered options on the tracking regime used and included "superhistory tracking" in the 1990's. It retained the HOLE option for geometry specification to the end of the century.

Other Monte Carlo codes have been developed and used for criticality safety assessment. The principal competitor for KENO in the USA is MCNP, developed at Los Alamos and a direct descendent of the original Monte Carlo work there (Los Alamos claim that over 300 person-years have been invested in it). It is a much more general code than either KENO or MONK since it includes both neutron and photon transport and it has been widely used for physics problems related to radiation including dose calculations and radiation damage studies (22). For criticality work its comprehensive geometry package and variance reduction techniques are key features. It finds some use in the UK where its complete independence of MONK and KENO makes it of particular interest to the UK nuclear regulator. The French code MORET is also a well-validated and internationally-recognised multigroup Monte Carlo code developed by the Institut de Radioprotection et de Sûreté Nucléaire (IRSN) and is the approved Monte Carlo assessment code in France. As with TRIPOLI, a continuous energy Monte Carlo code from the French CEA, it has not, so far, established itself elsewhere.

The increase in computing power has changed the availability of Monte Carlo techniques immensely. Calculations that could be only be run on a mainframe could, by the 1990's, be performed to sufficient precision on stand-alone workstations and personal computers. The power and dominance of the Monte Carlo codes by the end of the century led to concerns among some regulators that no independent checking of their predictions was possible. Also, even though so powerful, the Monte Carlo method still requires considerable experience from the practitioner if reliable results are to be obtained efficiently. The main weakness derives from its statistical nature; Whitesides in 1971 pointed out a particular problem leading to underestimation of k_{eff} in loosely coupled systems (33) that remains a classic challenge.

Deterministic Computer Codes

An alternative to the Monte Carlo approach is to solve the transport equation that describe the generation, movement and capture of neutrons. This complex equation can be simplified by breaking the nuclear data down into energy groups – the multi-group approach used in both diffusion and Monte Carlo. The resulting equations can then be solved numerically on a spatial mesh in a finite difference approach. However, one of the most difficult things to

manage in such calculations is the directional anisotropy – the colliding neutron has a preference to continue in its original direction of travel. The flux at one point therefore depends in a very complex way on the flux at many others. One way to proceed in numerical computations is to divide the possible directions into a discrete number of groups and then to weight these to reflect the directional behaviour.

The method (often known as the S_n method) emerged from the nuclear weapons design programme at Los Alamos in the mid-1950s(14) and was recognised quickly as a more accurate tool than diffusion theory for most criticality situations. Initially as a one-dimensional tool it was able to handle spheres, infinite cylinders and planes but it was extended to two dimensions in the late 1950's. By the 1960's it was a standard computational tool for criticality computations with computers with the two-dimensional version giving a capability for some simple arrays.(13). The one-dimensional codes were DTF at Los Alamos and ANISN at Oak Ridge. The well-known two-dimensional codes DOT from Oak Ridge (later developed as DOT2B by General Electric) and TWOTRAN (Gulf General Atomic) appeared in the late 60's. Even then the methods were restricted to relatively simple geometries and even the two dimensional codes could not be fully exploited until computer power increased and became cheaper. However, they were a useful as a cross-check on other methods.

The Winfrith Improved Multigroup Scheme (WIMS) code began to be developed by the UKAEA at Winfrith in England in the 1960s as a deterministic thermal reactor physics code. Although primarily aimed at reactor calculations, some of its later versions – much enhanced by reactor research and development - found extensive use in the UK as a safety assessment tool when simplifying assumptions on geometry could be made and advantage taken of the lattice geometry. Such deterministic approaches have particular value in scoping calculations and sensitivity studies where they give results free of the uncertainty associated with Monte Carlo methods. Similarly, the development of the deterministic APOLLO code by the CEA in France began in the 1970s and continued to the end of the century. The entry of new deterministic codes such as Imperial College, London's EVENT in the last decade of the century seemed to mark a resurgence of interest in deterministic techniques, driven perhaps by cheaper and more powerful computing resources.

The US and UK codes (Monte Carlo and deterministic) mentioned above are now generally operated on a strictly commercial basis for the use of the current and latest editions. The not inconsiderable cost of this leads some smaller users to employ older versions of some of the codes which are available (free of charge to some non-commercial users) primarily through the OECD Nuclear Energy Agency (OECD/NEA) Databank in Paris (www.nea.fr).

Hybrid Methods and Code Suites

The Monte Carlo and deterministic approaches such as diffusion and discrete ordinates developed quite independently and indeed have been to some degree in competition. In the 1990s there was an interest in combining the best features of each to make a more efficient overall tool and provide a complete package for criticality assessment. Typically, these use the deterministic approach, perhaps on a simplified geometry, to estimate the flux distribution and then use this to ensure that the Monte Carlo neutrons are concentrated in regions of greatest importance.

Most of the major codes were, by the end of the century, incorporated into code suites which performed criticality calculations and other related ones such as shielding. Provision was made for easier and more reliable data management and, in some cases, the use of a hybrid methods. For example, the UK WIMS code (managed by the ANSWERS Service of Serco Assurance www.sercoassurance.com) has developed into a code suite built around MONK. Similarly the Oak Ridge SCALE suite is built around KENO and the French nuclear industry

began, in the mid-1990s, to construct an integrated suite of programs called CRISTAL based on the MORET/APOLLO/TRIPOLI combination.

Data and Validation of Computer Methods

The computer codes for criticality are complex and demand some considerable skill in use if they are to be accurate and efficient. They have allowed designers to decrease the conservatism demanded by the cruder methods of earlier years so their accuracy is crucial to safety and this depends on the nuclear data that is used. This is a complex topic rather beyond the scope of this paper but at least one trend is worth noting: the increasingly global nature of the databases used. In the early work it was necessary for individual national groups to develop their own data sources – although there was always much sharing of information(25) – and structure it for their particular requirements. Progressively this cooperative approach has become more international with general availability through the OECD/NEA Databank of US (ENDF) and other national nuclear data libraries. This cooperative approach culminated in an international project, sponsored by the OECD/NEA, to produce the Joint Evaluated File (JEF) data. This began in 1995 and the first evaluated data became available in 2002.

Although there has never been an accident as a result of a failing in a code, the need for validation against benchmark experiments has long been recognised. For example in the USA, although there had been comparisons by individual workers for many years, the requirement for validation was formalised in a Nuclear Standard in 1975 (34). This was reiterated in ANS 8.1 in 1983 and subsequent revisions.

An essential part of the validation process has been international benchmarking and the most significant of these is the International Criticality Safety Benchmark Evaluation Project (ICSBEP) that was initiated by OECD/NEA in 1992. The International Handbook of Evaluated Criticality Safety Benchmark Experiments, prepared under the project, contains over 400 evaluations of nearly 4000 critical or near critical configurations and is widely used around the world in code validation.

Handbooks, Standards and Safety Principles

The data from experiments and code calculations have been made available to practitioners through handbooks, where they were generally combined with guidance on criticality safety principles. The earliest guidance came with the AEC's Nuclear Safety Guide TID 7016 in 1957(16), the declassified LA-2063 from the previous year. This was not intended as a handbook but, in its 24 pages, it sets down the basic nuclear safety problem, some rules and data and then goes on to consider applications to processing plants.

It begins by pointing out the challenges. First, the nuclear data were not sufficient and the theoretical methods not well enough understood to calculate critical masses to better than 15 to 20 %. Reliance had to be placed on experimental measurements of critical mass and extension of these by theory. Second, the disposition of fissile material may not be well-known in a process, particularly in off-standard conditions. Third, administrative controls, based on operating rules, must be rigorously applied.

After reviewing the factors that contribute to criticality, the need for conservatism in design criteria is stated. The inability of instrumentation to warn, in practical situations, of the approach to criticality was noted. However, criticality accident detectors that warn that an excursion has occurred (already installed in many operations) were important. A short section considers the consequences of an accident.

The preference was for protection through safe geometry by limiting pipe and container diameters and keeping them far enough apart to avoid significant interaction. Where safe geometry was impracticable, protection could be through control of quantities or concentrations but this must require two or more simultaneous and independent contingencies before a chain reaction could occur. These key principles have survived to the present day.

Basic limits are given for individual units and then for arrays. These are handled with the two-group diffusion theory and a limit for the total solid angle subtended at any unit by all the others. This was set at 1 steradian. The revision in 1961(35) corrected a few mistakes and made comments on mishaps. It dealt with neutron poisons more completely and expanded on the solid angle method.

TID 7016 continued to develop and in the 1978 revision(19) (by now the document had reached nearly 130 pages and came from the Nuclear Regulatory Commission) there was mention of computer codes and reference to the 1975 ANSI standard N16.1(36), first adopted in 1964. The methods for control were reiterated as the double contingency principle, control through safe geometry and the use of neutron poisons and the sub-critical limit was introduced from the Standard. The surface density and density analogue methods were offered for arrays in addition to the solid angle approach.

The document thus evolved (the document says "matured")over the 20 years as a much broader data base became available. The dramatic development was the increased reliance on computer codes: "The advance of calculational capability has permitted validated calculations to extend and substitute for experimental data." However, the principles of protection remained much the same (37).

Two other US documents have found wide use. The important "Criticality Handbook" ARH-600 was published in 1968 (38). While it too expressed the principles of criticality safety, its significance was probably in the wide range of critical data drawn from experiment and code calculations (mainly GEM and KENO). Although intended for use at the Hanford site it found wide use around the world as a source of authoritative data that was, on the whole, less conservative than TID 7016. Similarly the Los Alamos report LA-10860 (39), published about the same time, presented a range of data that made it a useful reference document, after a 1986 update, to the end of the century.

In the UK the earliest principles and criteria were prepared by UKAEA and are associated with staff at the Authority Health and Safety Branch Safeguards Division. The Branch was formed, with Safeguards and Radiological Protection Divisions after the Windscale reactor accident in 1957 as the focus for safety within the organisation. The Safeguards Division was, as has already been noted, responsible for the development of criticality methods but it was also responsible for criticality inspection and it was from this part, led initially by Jack Chalmers, that the guidance came (40). The most significant early collection of data was probably the Manual of Criticality Data (41).

The Handbooks and Manuals were originally intended primarily for designers and as guides for safe operation and their roles in this and in safety assessment were overtaken by codes. However, they are still used by assessors for scoping calculations and cross checking

A particularly complex and important aspect of guidance and criteria has been the role of regulating authorities in criticality safety. Detailed description of this is beyond the scope of this paper. Suffice it to note that regulators around the world have recognised, within their different frameworks, the importance of criticality control and have themselves maintained a high degree of technical expertise while taking an active interest in the competence of criticality assessors. There is a general agreement on the broad structure of protection that flows from the early handbooks. For example, controls which are passive – such a safe-by-

geometry - are preferred to those which depend upon active engineering and these are better than those that rely on human management control.

Criticality safety has also not been divorced from the risk-based approach that dominated safety thinking, particularly in the UK, in the final decades of the century. This extended from the application of structured techniques such as HAZOP to identify criticality hazards to efforts to apply Probabilistic Risk Assessment methodologies in situations where this was appropriate. In the UK such an approach has been promoted by the Nuclear Installations Inspectorate by, for example, the setting of frequency targets for criticality events which may trigger the installation of Criticality Incident Detector (CID) systems. As a result of adopting such approaches, criticality safety has become more integrated into the overall radiological risk assessments.

Criticality accidents

Although this paper is focussed on the techniques used to establish how close a fissile system is to criticality it would not be appropriate to ignore the work that has been done on establishing the consequences of criticality accidents. This falls into two parts: understanding the accidents that have happened and devising methods to calculate the consequences of possible future accidental criticalities.

Past Accidents

The first fatal criticality accidents occurred at Los Alamos in 1945 and 1946 and both involved the same 6.2 kg sphere of plutonium – just below the critical mass in air. In the 1945 accident, on 21 August, the lone experimenter, Harry Daghlian, was adding tungsten carbide bricks as a reflector around the sphere. As he was about to place the last brick he realised from the response of neutron counter that it would make the assembly critical but, as he withdrew his hand, the brick slipped onto the assembly and which became critical. He managed to push off the final brick to bring the excursion to an end but received a dose of 510 rem and died 28 days later.

In the second accident a senior scientist, the group leader Louis Slotin, was using the sphere to demonstrate to a group of six people how criticality experiments were performed. This time the sphere was surrounded by two beryllium hemispheres as reflectors. The scientist had removed two safety spacers and was holding the top hemisphere away from the sphere with his thumb and a screwdriver. The screwdriver slipped and the hemisphere fell onto the sphere, its neutron-reflecting capability making the assembly critical. The demonstrator received some 2100 rem and died nine days later. The others received non-fatal doses between 360 and 37 rem. (42).

Both these accidents resulted in a chain reaction without the involvement of the delayed neutrons. This accounts for the speed with which they developed. This kind of excursion is referred to as prompt criticality.

A third, much later, accident in a Los Alamos plant on 30 December 1958 (42) illustrates how things can go wrong with solutions. More generally it shows the need to understand the physics and chemistry of systems if accidents are to be avoided. The plant was designed to recover plutonium from a variety of laboratory operations and, for reasons that are not clear, the residues from several process tanks were transferred into a single large tank. Here two layers formed. At the bottom was acid/water and on top of it a layer of organic solvent. Dissolved in the organic solvent was just over 3kg of plutonium. The arrangement was sub-critical until an operator turned on a stirrer when there was an excursion that gave him a

dose of over 10,000 rem that killed him 36 hours later. It seems that the stirring had made the organic layer thicker at the centre of the vessel and this, with the change in shape of the reflecting water layer, created a criticality excursion.

The systematic study of criticality accidents began with WR Stratton in 1960 (43) and the revisions and updates he made to produce report LA-3611 in 1967 (44). That report was revised by David R Smith in 1989 as NCT-04 and then by joint US and Russian Federation authors in 2000 as LA-13638(42). This most recent report lists 22 criticality accidents in process operations (with over half in the Russian Federation and 21 of them involving solutions) and another 38 in experiments. In the process accidents nine people died and a further three had limbs amputated. Most of the accidents occurring in experiments actually happened in facilities designed to study criticality. In the process accidents there was very little (if any) damage to equipment but there was of course disruption of activities. Altogether 12 people are known to have died in these experiments but members of the public were exposed in only one accident and this at a relatively low level.

Evolution of a Criticality Accident

The evolution of a criticality accident is complex and depends on the specific circumstances. However, the general pattern is that criticality excursion creates changes in the system that quench the excursion (at least temporarily) to bring the system back to sub-criticality. Experiments have been conducted with specialised reactor systems in the USA and France and these have shown several quenching mechanisms. In LA-3611 in 1967 (44) Stratton was able to refer to results in the mid to late 1950s from a number of reactor systems: Dragon, Borax, Godiva-I and Godiva-II, the Spert and Kewb reactors, Triga, Treat and Snaptran. He was then able to list the significant quenching mechanisms (thermal expansion, rise in neutron temperature, boiling, U-238 Doppler effect and radiolytic bubble formation) and indicate how these came into play in experiments with the reactors.

He used the energy model suggested by Klaus Fuchs (the atom spy) in 1946 (45) and developed by Hansen (46) to make some estimates of likely energy release profiles and his calculations showed some patterns useful in interpreting the accidents. The first key parameter was the size of reactivity step injected in the accident. Where this took the reactivity from delayed critical to significantly above prompt criticality there was a characteristic energy spike. The spike would be more energetic and shorter in unmoderated systems and be followed by a plateau in power generation lasting up to a second. The moderated systems showed a smaller spike and no plateau. When the excess reactivity injected is lower and just takes it to prompt criticality, neither unmoderated nor moderated systems showed the short spike and a pulse lasting a few seconds is predicted.

An indication of Stratton's strength, and perhaps of the maturity of the topic, is that, 33 years later, in the second revision (LA-13638) his calculations and wording about power excursions and quenching mechanisms could be repeated with little change. The additional data that could be produced related to the French CRAC studies undertaken at Valduc between 1967 and 1972 (and, incidentally, continued to the end of the century) that were specifically aimed at understanding criticality accidents in process solutions and these did not cause any changes.

One of the first calculations of criticality consequences, for an aqueous solution, may well have been that of Christy and Wheeler (this was the Wheeler who coined the term "buckling") in January 1943 (47). They imagined a solution of Pu-239 that evaporated until it just reached criticality in a closed vessel. The temperature rise that followed, they estimated, would reduce the density of the solution and increase the leakage of neutrons enough to drop the effective multiplication factor below 1. Fission energy generation would stop and the

solution would cool down enough for the process to repeat. This would go on until a steady state was reached. The system would be a potential self-regulating power plant. If the vessel were open, they thought that the solution would boil down until the solution was so concentrated that it was not critical. They estimated that the boiling away of 1 litre of solution 10m from an unshielded individual would give them an "integrated dosage of 44 roentgen units". A figure, in itself, "not dangerous".

The development of codes designed to predict the development and consequences of accidental criticality excursions was a natural extension of the early work. In the UK, Don Mather of UKAEA was responsible from the 1970s to the 1990s for codes that predicted transients in both solutions (CRITEX) and powders (POWDER). By the end of the century the FETCH code from Imperial College, London was a world leader.

Conclusion

Perhaps the most striking fact in the history of criticality assessment is that there seems never to have been an accident because of error in the assessment calculation itself. The accidents that have happened have arisen because either some unforeseen chemical or physical process arose accidentally or because humans have behaved rashly or in ignorance. The story of assessment itself started with the theoretical calculations that led to bombs and reactors. It matured into validated computer tools that now form the backbone of criticality assessment across the world. There are still problems to be solved but, by the end of the century, criticality assessment had become an essentially routine process. It remained however one which demanded a high level of skill, an appreciation of the limitations of the tools available and an intuitive understanding of the physical background to assessments.

I would like to thank the people who helped in assembling this review, notably Elliott Whitesides, Frank Abbey and Peter Thorne.

References

- 1 Fermi E, 1934, Possible production of atomic number higher than 92, *Nature*, 133, (898-899)
- 2 Hahn O and Strassmann F, 1938, *Naturwiss*, 26, (756)
- 3 Hahn O and Strassmann F, 1939, Über der Nachweis und das Verhalten der bei Bestrahlung des Urans mittels Neutronen entstehenden Erdalkalimetalle, *Naturwiss*, 27, (11-15)
- 4 Hahn O and Strassmann F, 1939, Nachweis der Entstehung activer Bariumisotope aus Uran und Thorium durch Neutronbestrahlung, *Naturwiss*, 27, (89-95)
- 5 Meitner L and Frisch O, 1939, Disintegration of uranium by neutrons: a new type of nuclear reaction, *Nature*, 143, (239-240)
- 6 Szilard L, 1934, Improvements in or relating to the transmutation of chemical elements, UK Patent Specification 630726
- 7 Perrin F, 1939, Calcul relatif aux conditions eventuelles de transmutation en chaine de l'uranium, *Comptes Rendus B* 1 May 1939, 208, (1394-1396)
- 8 Peierls R, 1939, Critical conditions in neutron multiplication, *Proc Camb Phil Soc*, 35, (610-615)
- 9 Frisch O R, 1939, Radioactivity and sub-atomic phenomena, *Ann Repts Prog Chem*, 36, (7-24)
- 10 Frisch O R and Peierls R, 1940, Radioactivity and sub-atomic phenomena, *Ann Repts Prog Chem*, 37, (7-22)
- 11 Serber R, 1992, *The Los Alamos Primer: the first lectures on how to build an atomic bomb*, Berkeley; Oxford: Univ Cal Press
- 12 Konopolis E, Metropolis N, Teller E and Woods L, 1943, USAEC CF-548
- 13 Knief R A, 1985, *Nuclear Criticality Safety: Theory and Practice*, La Grange Park, IL: ANS

- 14 Whitesides G E, Westfall R M and Hopper CM, 2003, Criticality Safety Methods, ORNL Seminar Gatlinburg,TE
- 15 Reider R, 1971, An Early History of Criticality Safety, LANL LA-4671
- 16 Callihan A D, Ozeroff W J, Paxton H C and Schuske C L, 1957,Nuclear Safety Guide ,USAEC TID-7016
- 17 Schuske C L and Paxton H C, 1976, History of fissile array measurements in the United States, Proc Am Nucl Soc,30,(101-137)
- 18 Thomas J T, 1966, Method for estimating critical conditions of large arrays of uranium, ORNL-3973 Vol1
- 19 Thomas JT (ed), 1978, Nuclear safety guide, TID-7016rev 2 NUREG/CR0095
- 20 Thomas A F and Abbey F, 1973, Calculational methods for interacting arrays of fissile material, Oxford:Pergamon Press
- 21 Simister D N and Clemson P D, 2003, A historical review of critical experiment research facilities in the United Kingdom, Nucl Sci Eng, 145,(64-71)
- 22 Greenspan H G, Kelber C N and Okrent D(eds), 1968,Computing Methods in Reactor Physics, New York: Gordon and Breach,
- 23 Metropolis N, 1987, The Beginning of the Monte Carlo Method, Los Alamos Science , Special Issue,(125-130)
- 24 UKAEA, 1966, Fermi invention rediscovered at LASL, Atom, October,(7-11)
- 25 Hansen G E and Roach W H, 1961, Six and sixteen group cross sections for fast and intermediate critical assemblies, LAMS-2543
- 26 Whitesides G E, 2004, Private communication,
- 27 Longworth T C, 1965, The GEM Monte Carlo code, IAEA Conference Proceedings: Criticality Control of Fissile Materials,
- 28 Woodcock E R, Murphy T, Hemmings P J and Longworth T C, 1965,Techniques used in the GEM code, USAEC ANL 7050
- 29 Hemmings P J, 1967, The GEM code, UKAEA AHSB(S)R105
- 30 Longworth T C , 1968,The GEM4 code,UKAEA AHSB(S)R146
- 31 ANSWERS Software Services, Hole geometry, MONK/REPORT/003
- 32 Moore J G, 1974, The general Monte Carlo code MONK, ANL-75-2 & NEA-CRP-L-118
- 33 Whitesides G E, 1971, A difficulty in computing the k-eff of the world, Trans Am Nucl Soc,14, (68)
- 34 ANS, 1975, Validation of calculational methods for nuclear criticality safety,ANS-8.1/N16.9-1975
- 35 US Atomic Energy Commission, 1961, Nuclear Safety Guide, TID-7016 Rev1
- 36 ANS, 1975, Criticality safety in operations with fissionable material,ANS-8.1/N16.1 -1975
- 37 ANS, 1983, Criticality safety in operations outside nuclear reactors, ANS-8.1-1083
- 38 Carter R D, Kiel G R, Ridgeway K R, 1968, Criticality Handbook, ARH-600, Atlantic Richfield Hanford Co
- 39 Paxton H C and Pruvost N L, 1986, Critical dimensions of systems containing U-235, PU-239 and U-233(Revision of 1967 report), LANL, LA-10860
- 40 Jordan G M, 1963, The identification of criteria for criticality assessment, AHSB(S)R56, UKAEA
- 41 Abbey F, 1967, Manual of Criticality Data,AHSB(S) Handbook 5, UKAEA
- 42 McLaughlin P M, Monahan S P, Pruvost N L, Frolov V F, Ryazanov B G and Sviridov V I, 2000,A Review of Criticality Accidents 2000 Revision, LANL LA-13638
- 43 Stratton W R, 1960, A review of criticality accidents, Prog Nuclear Energy Ser IV,3,
- 44 Stratton W R, 1967, A review of criticality accidents, LASL LA-3611
- 45 Fuchs K, 1946, Efficiency for very slow assembly, LASL LA-596
- 46 Hansen G E, 1952, Burst characteristics associated with the slow assembly of fissionable material, LASL LA-1441
- 47 Christy R F and Wheeler J A, 1943, Chain reaction of pure fissionable materials in solution, USAEC CP-400 (1 Jan 1943)