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NUCLEAR SAFETY IN MANUFACTURING PLANTS

By
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NUCLEAR SAFETY
IN MANUFACTURING PLANTS*

by

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November 3, 1959

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on April 30, 1959.

ABSTRACT

Loosely speaking, criticality control in the processing of fissionable material requires the assurance that there shall be less than one neutron available in a fission generation for every neutron produced in a previous generation. This poor neutron economy is achieved by design conditions which are essentially the reverse of those involved in reactor design. A critical reactor must contain at least the critical mass of the fissionable material in question. This mass is not single-valued but depends on many factors, one of these being neutron leakage.

A nuclearly safe design varies as the type of process; that is, whether it is a continuous or a batch process. The continuous process is often regulated through the geometric parameters of the system. It is often desirable to control the batch process by limiting the amount of fissionable material to some value below the minimum critical mass. Important adjuncts to nuclear safety in processes are the problems relating to storage and shipments, both within and out of the plant.

In all cases, several fundamental criteria are desirable.

- 1) Safety of a configuration should depend on the occurrence of at least two unlikely events.
- 2) Configurations should be nuclear safe even if totally flooded with water or, if this possibility can be excluded, account should be taken of other incidental reflectors.
- 3) In the case of mass control, the possibility of double-batching should be considered.
- 4) The intentional use of neutron poisons should not be necessary to achieve subcriticality in manufacturing processes. However, certain materials may be used to afford additional safety.

To assure that nuclear safety is established and maintained, three basic problems must be solved satisfactorily. Solution of the scientific problem

involves theoretical and experimental knowledge of the various criticality parameters. The second, or engineering problem, requires that the proposed process be analyzed in detail in the light of the scientific information. Finally, administrative controls are necessary to minimize either personnel or equipment failure.

INTRODUCTION

Over recent years, a sizable and increasing fraction of reactor fuel fabrications has been carried out in industrial plants not associated with the large Atomic Energy Commission installations. These plants operate either under government contract or the AEC licensing program. Fabrications performed under contract remain under AEC control and are obligated to conform to health and safety provisions which are specified in the particular contract. To obtain a license to fabricate fuel, one must first apply to the AEC, demonstrating a capacity for assuming the responsibility of handling the fissionable material, as well as that adequate health and safety measures can be and will be incorporated into the process. In both cases, health and safety provisions are needed to assure that unwanted criticality does not occur. This criticality control is called nuclear safety.

In general, the assurance of nuclear safety is a three-step problem.¹ First, there are the scientific considerations having to do with the measurement of basic nuclear properties, the subsequent calculation of critical parameters, and a comparison of these parameters with the results of critical experiments. The next step involves engineering, wherein one determines that the contemplated disposition and configuration of fissionable material in the design of a fabrication or process conforms to nuclearly safe values. This safety of design must not be alterable by off-standard conditions, such as human or equipment failure. The third step in assuring nuclear safety is administrative in nature in that controls must be set up or modified to offset possible equipment and personnel failure. Oftentimes this step also involves an independent check of the proposed process, for example, in examining a proposed contract or license application. However, the nuclear energy business has expanded so rapidly that there are not enough persons available to resolve adequately the numerous nuclear safety problems. This is especially true in the case of small plant or commercial-type fabrications, when the services of a full-time criticality engineer cannot be justified. Hence, the independent analysis can become very important.

The conditions sought in nuclear safety are quite opposite to those desired in nuclear reactor design. In the latter case, good neutron economy is essential, while when striving for nuclear safety the absorption of neutrons in nonfissionable materials and neutron leakage ought to be as high as reasonable. In the following pages are summarized the factors which influence neutron economy and which, therefore, are important in nuclear safety.

SHAPE

A shape having a low ratio of surface area to volume is optimal in reducing neutron leakage. Spheres, as well as near spherical shapes such as cubes or cylinders having equal diameters and heights, are the most reactive. Hence, arranging fissionable material as thin cylinders or slabs is desirable in assuring nuclear safety. This can be illustrated by referring to a parameter called buckling or B^2 .

The description and derivation of B^2 , which appears in many technical reports and books, is beyond the scope of this discussion. In particular, the reader is referred to the textbook, The Elements of Nuclear Reactor Theory² by S. Glasstone and M. Edlund. Briefly, the buckling is a measure of the curvature or "buckling" of the neutron flux as a function of position in a reactor core. The critical equation of a reactor system relates B^2 with the physical characteristics affecting neutron multiplication and diffusion of the materials combined to form the system. B^2 is actually a geometric quantity, its value depending upon the shape and size of the reactor. Hence, a reactor is critical when its shape and size results in a value for B^2 which is equivalent to that B^2 value inferred from the materials in the system.

Values of B^2 for some common shapes and the corresponding minimum critical volumes as reported by Glasstone and Edlund are shown in Table I. The letters in the Table refer simply to the dimensions of the shape in question. It should be noted that B has no specific label, though its value is calculable from the system's dimensions. The Table indicates that a spherical shape has the smallest minimum critical volume. Since neutron production in a critical system occurs throughout the volume and leakage depends on surface area, it is clear that a sphere will be most reactive for a given composition and such shapes should be avoided.

TABLE I

Dimensions and Bucklings for
Some Common Shapes

Geometry	Bucklings	Minimum Critical Volume
Infinite Slab	$(\frac{\pi}{a})^2$	—
Rectangular Parallelepiped	$(\frac{\pi}{a})^2 + (\frac{\pi}{b})^2 + (\frac{\pi}{c})^2$	$\frac{161}{B^3}$
Cylinder	$(\frac{2.405}{R})^2 + (\frac{\pi}{H})^2$	$\frac{148}{B^3}$
Sphere	$(\frac{\pi}{R})^2$	$\frac{130}{B^3}$

a = thickness b = width c = length
R = radius H = height

MODERATION

Cross sections for neutron captures which lead to fission increase greatly as the neutron energies decrease to the thermal range. This neutron thermalization takes place when fissionable atoms are mixed with certain materials called moderators. The most common moderating material found in a processing plant is ordinary water. The effect of neutron moderation is to decrease greatly the minimum critical mass for metallic U-235, about 22.8 kilograms, to one of about 0.8 kg for a homogeneously mixed U-235 water system. Both of these values refer to the fully water-reflected situations. Figure 1 is a curve obtained from an Oak Ridge report, Studies in Nuclear Safety,³ which illustrates the variation of the critical masses of U-235 as highly enriched uranium with the size of metallic pieces in water lattices. The uranium solution is the most reactive system having a minimum critical mass of less than 1 kg of U-235. Critical mass increases with the metal size till a value of 22.8 kg of U-235 is necessary for the reflected solid, metal sphere. Each of the intermediate points represents an optimum lattice, that is, one with a minimum mass.

Fig. 1 Minimum Critical Masses of Enriched Uranium
Water Lattices

REFLECTION

Surrounding fissionable material with almost any material will increase the system's reactivity by reducing neutron leakage. Just as the mixing of a moderator with fissionable material often increases reactivity; *i. e.*, results in a lower mass requirement for criticality, so the addition of a neutron reflector has a similar effect. In evaluating the safety of processes, it is customary to assume more severe operating conditions than one would normally expect. Hence, most processes are designed to be safe even in the unlikely event of flooding with water. Of course, flooding of a process area is possible, because of a water main or sprinkler system failure. A more important reason for the flooding assumption, however, is to account for the unknown neutron reflecting properties of incidental materials such as equipment, walls, and personnel. If a single unit or system containing fissionable material is subcritical or safe under flooding conditions, it would be safe during reasonably foreseeable abnormal conditions. Removing the reflector from between neighboring units, however, can increase the reactivity of an array by neutron interaction. The interaction problem is discussed later. Two examples to illustrate the effect of a reflector are noteworthy. First, Table II illustrates how the addition of a thick water reflector affects the critical mass (M_c) of metal spheres of U-235 and Pu. An unreflected sphere of highly enriched uranium has a critical mass of about 48 kg of U-235 which is reduced to 22.8 kg, the value mentioned earlier.³ Plutonium experiences a similar factor of two reduction in critical mass. The second example, which was inferred from some early Oak Ridge work with uranyl fluoride solutions by Beck, Callihan and others,⁴ is shown in Figure 2. The reactivity increase by neutron reflection is illustrated clearly here. For a 10-in. diameter aluminum-contained, homogeneous reactor, water reflection reduces the required critical U-235 mass by about 58% of the mass needed for a bare cylinder. The reduction achieved by adding a reflector for the stainless steel case is only about 51%. The curves indicate that in both cases adding reflectors becomes somewhat less important as diameter increases. This is explained by the fact that, for a given concentration of U-235, a large fraction of the neutrons escape a smaller diameter reactor and are available for reflection. As the reactor size increases, therefore, the critical masses with and without a reflector become more comparable.

TABLE II

Comparison of Critical Masses
of Plutonium and U-235 Metal Spheres

Reflector	M_c (Pu) (kg)	M_c (U-235) (kg)
None	16.2	48.0
Water	8.0	22.8

(Pu density 15.8 g/cc, U-235 density 18.8 g/cc)

NOTE:
 HfU^{235} ATOMIC RATIO=100 FOR
 VARIOUS REACTOR DIAMETERS

Fig. 2 Effect of Reactor Container Material and Diameter on Critical Mass

CONCENTRATION

The U-235 concentration in solution also has an important effect in criticality. In the previous curve (Figure 1), the reduction of critical mass with decreasing size of the highly enriched uranium metal units was shown. Hence, homogeneous or solution data can often be used to assure the nuclear safety of heterogeneous enriched systems. However, the variation of U-235 concentration in solutions also changes the criticality parameters significantly. This variation is attributable to fast neutron leakage and multiplication factor, k , which are competing effects. A fairly simple computation can illustrate this variation. First, the infinite multiplication factor, for a highly enriched uranium system, is

$$k_{\infty} = \eta f = \frac{\eta}{1 + \left(\frac{H}{U-235}\right) \left(\frac{\sigma_H}{\sigma_U}\right)},$$

where η is the average number of fission neutrons per thermal neutron capture produced in the uranium and f is the ratio of the thermal neutrons absorbed in the fuel to the total number absorbed. The σ 's are the microscopic absorption cross sections for the hydrogen of the water and the uranium. The oxygen in water can usually be neglected. H:U-235 is the ratio of hydrogen atoms to U-235 atoms. Obviously, as H:U-235 increases, k_{∞} decreases slowly.

Now the systems of practical interest are finite in size and account must be taken of neutron leakage. This can be done by considering the modified two neutron energy group treatment employed by J. A. Pond,⁵ namely

$$k_{\text{eff}} = \eta f U_t U_f,$$

where U_t and U_f are the thermal and fission neutron nonleakage probabilities, respectively, and k_{eff} is the effective neutron multiplication factor of the system in question; i. e., it is k_{∞} multiplied by the fraction of neutrons which do not escape or leak out. For a critical reactor, $k_{\text{eff}} = 1$, and the above equation can be rearranged to

$$\frac{H}{U-235} = \frac{\sigma_U}{\sigma_H} (\eta U_f U_t - 1) .$$

To maintain criticality while varying the H:U-235 ratio, obviously the nonleakage probabilities must be changed also. As indicated earlier, increasing H:U-235 at first improves neutron thermalization by the hydrogen atoms and neutron leakage decreases. Hence, a smaller critical mass is needed as H:U-235 increases through low ratio values. At some point, thermalization becomes relatively constant and the required critical mass reaches a minimum. At high H:U-235 ratios, neutron absorption by hydrogen becomes important and the critical mass increases. A further effect is noteworthy, namely that the increasing H:U-235 ratio or greater dispersion of atoms further increases the critical mass and, hence, the reactor volume increases very rapidly.

There exists an H:U-235 atom ratio above which criticality is impossible. Recall that

$$k_{\infty} = 1 = \frac{\eta}{1 + \left(\frac{H}{U-235}\right) \left(\frac{\sigma_H}{\sigma_U}\right)}$$

for criticality in an infinite system. Since $\eta = 2.08$ and $\frac{\sigma_H}{\sigma_U} = 0.00048$,

$$1 + 0.00048 \left(\frac{H}{U-235}\right) = 2.08$$

$$\therefore \frac{H}{U-235} \doteq 2250 .$$

Hence, uranium solutions having atom ratios above this value, or containing less than about 11.6 g U-235 per liter, cannot become critical.

The effect of the H:U-235 atom ratio can be illustrated further by referring to some data given by Beck in the work mentioned earlier.⁴ Table III shows the distribution of neutron absorption for two extreme concentrations. It should be noted that the fraction of absorptions by the hydrogen in the water increases by a very large factor with decreasing concentration. To summarize, then, Figure 3 illustrates how minimum

critical mass varies with H:U-235 ratio for an aluminum-contained reactor. The curves are the envelopes of experimental curves for reactors of several diameters and hence relate values for the most reactive cylinders for both reflected and unreflected aluminum and stainless steel contained reactors. Up to H:U-235 ratios of about 200, the 10-in. diameter cylinder was critical with the smallest critical mass when not reflected. At higher ratios, 15-in. diameter cylinders were more reactive. For reflected aluminum reactors, the minimum critical masses occur in smaller diameter cylinders than in the unreflected case at most H:U-235 ratios. This can be explained by the fact that neutron leakage decreases with increasing reactor size and reflectors become less important in large reactors. The minimum critical mass for all the solutions indicated is about 1 kg of U-235 at an H:U-235 of about 400.

For comparison's sake, note that the critical masses at H:U-235 equals 100 are 5 kg of U-235 in an unreflected cylindrical solution and only about 1.8 kg for the same solution, reflected. It should be noted further that the minimum mass occurs in an 8-in. diameter cylinder for the reflected case, but in a 10-in. diameter cylinder for the bare case. Now Figure 4 shows similar curves for stainless steel cylinders. Here, the critical mass for the unreflected solution of H:U-235 equals 100 is only about 4 kg of U-235 versus 5 kg for the unreflected aluminum case. Obviously, then, the container of a fissionable material can influence criticality greatly. Adding full, water reflection, however, results in a critical mass of about 2 kg, a value only slightly higher than the same reactor in aluminum. In the one unreflected case, stainless steel is an important reflector compared to aluminum. Adding a water reflector around the steel enhances the neutron poison effect of the steel, accounting for the slight difference in the reflected cases. Hence, it is important to account for neutron reflection when verifying the safety of a process.

TABLE III

Distribution of Neutron Absorption
in Enriched Uranium Solutions

Absorber	H:U-235 = 25	H:U-235 = 1000
U-235 fission	84.1 %	57.0 %
U-235 radiative capture	14.1	9.5
Hydrogen	1.2	33.3
Impurities	0.4	0.2

CRITICAL ALUMINUM REACTORS

Fig. 3 Variation of Critical Mass with H:U-235 Ratio in Aluminum Cylinders

CRITICAL STAINLESS STEEL REACTORS

Fig. 4 Variation of Critical Mass with H:U-235 Ratio in Stainless Steel Cylinders

NEUTRON POISONS

Reference was made to the poison effect of stainless steel in comparing the reflected aluminum and stainless steel critical reactors. In general, the use of neutron absorbers to assure that a system or process is nuclear safe is undesirable. To predict the behavior of neutron poisons easily, the poison should be intimately mixed with the fissionable material and it must always remain there. Except in certain cases, neutron poisons are not mixed in reactor fuels. When they are intermixed, the proportions must be precisely determined. To use poisons in heterogeneous systems is also undesirable. For example, wrapping an unreflected vessel with a poison can actually increase reactivity, as was seen in comparing Figure 3 with Figure 4. It should be realized that one cannot identify a poison material, for example cadmium metal, simply by looking at it and, therefore, the poison ought to be at least permanently fixed in the system in question. Further, it is notable that the effectiveness of any neutron absorber varies with neutron energy. Even if a poison is effective, say during complete flooding when any fission neutrons would become pretty well thermalized, it might not be as effective under partial flooding when neutron thermalization could be incomplete and large numbers of fairly energetic neutrons exist. To summarize, then, each proposed process should be examined on its own merits. However, the usual rule is not to employ neutron poisons to achieve nuclear safety, but rather additional safety.

HETEROGENEITY

Earlier, it was mentioned that the use of homogeneous or solution data for highly enriched uranium can be used conservatively to assure nuclear safety of such heterogeneous systems. Lately, however, much of the processing has involved slightly enriched uranium, in the 1 - 5 w/o of U-235 range. With slightly enriched uranium, a heterogeneous lattice can form a more reactive system than one which has the fuel in solution. The increasing fraction of the U-238 isotope with decreasing enrichment accounts for increased reactivity of a lumped system over a homogeneous one. The probability that neutrons being thermalized will be captured at intermediate energies increases with the amount of U-238 present in the system; i. e., intermediate captures increase in systems having low enrichments of uranium. Lumping the uranium to form a heterogeneous lattice decreases the chance for such parasitic captures. A second effect is the possible increase of fast neutron fissions with increasing uranium lump size and U-238 content.

To illustrate the enhanced reactivity of heterogeneous over homogeneous systems of slightly enriched uranium, we refer to some experiments and calculations, done principally by Ketzlach at Hanford.^{6,7,8,9} Table IV compares estimated minimum critical masses of some systems of interest. It should be noted that as the degree of enrichment increases, the critical masses for the two classes of systems seem to converge, as expected.

TABLE IV

Estimated Minimum Critical Masses
for Uranium of Various Low Enrichments

w/o U-235	Homogeneous (kg U)	Heterogeneous (kg U)
1.5	1940 ⁶	800 ⁷ (not minimum)
1.6	—	265 ⁸
3.0	228 ⁶	79 ⁷ 92.5 ⁹ (in UO ₂)
4.9	39	—
5.0	—	37 ⁹

DENSITY

In many processes, metallic uranium is handled in the form of an alloy with nonfissionable metal. This disperses the U-235 atoms and reduces the chance that a neutron will interact with a U-235 atom. Advantage of the reduced reactivity can sometimes be taken.

INTERACTION

Once individual units are determined to be sufficiently subcritical under foreseeable off-standard conditions, the possibility of neutron interaction among units must be considered. Obviously, subcritical units must be separated adequately, the amount of separation varying for different kinds of units. Essentially two kinds of interaction problems exist in manufacturing plants, those concerned with the fixed installation of subcritical units, as during storage or transportation, and those concerned with the movement of material from one process to another in the same plant. The latter interaction problems generally require both mechanical devices to insure separation and administrative controls to offset any decrease in safety introduced by the material movement. These controls are strongly dependent on individual plant practices.

Examples of nuclear safety in fixed installations are more closely related to the earlier considerations. Figure 5 shows a storage array of enriched uranium scrap in one-gallon, sealed pails mechanically arranged on 20-in. centers. Each pail is mass-limited and the resulting subcritical neutron multiplication factor is estimated using Pond's method mentioned earlier.⁵ Then, the possible interaction is estimated from solid angle considerations. Fortunately, permissible solid angles have been devised by Oak Ridge and other groups and appear in several reports, including the new Guide to Shipment of U-235 Enriched Materials.¹⁰ Generally, the mass of U-235 is limited to about 300 grams per pail to assure that each pail would be subcritical even if flooded. Under conditions of total flooding, the units would be effectively insulated from each other neutron-wide by the thick water layer. Hence, the more reactive situation of partial flooding ought to be the storage design basis.

Much experimental study of interaction has been done with massive fissionable metal. A result of these studies has been the Nuclear Safety Guide which recommends safe parameters to be used during storage

and shipping.¹ The Guide specifies unit separations for different classes of material. For example, one should never ship more than 20 kg of unmoderated, fully enriched uranium in any single package. This mass would be subcritical even if immersed in water. Actually, many plants prefer to ship only up to 11 kg. In either kind of shipment, adequate separation from other, similar units is determined by using an iron frame or "bird-cage" surrounding the actual package, such as those shown in Figure 6. The Nuclear Safety Guide recommends that the U-235 density in a shipment of metallic, enriched uranium units should not exceed 4 kg/ft³. Hence, one can safely ship a reasonable number of units of enriched uranium in the 20-in. bird-cages if each cage contains 11 kg of U-235 since the density, 2.4 kg/ft U-235, is less than the recommended value.

In one of its regulations, the AEC recommends a modified 55-gallon drum for shipping uranium-bearing scrap. Figure 7 is a diagram of such a container. A centrally fixed stovepipe arrangement assures adequate separation among similar units. Since the central container has a diameter greater than 5-inches, the safe diameter value under any conditions of moderation and reflection, the uranium loading of the drum must be limited as to mass.¹⁰

Fig. 5 Uranium Metal Chip Storage Area (Courtesy of
Nuclear Metals Inc.)

Fig. 6 Bird-cages for Shipping Enriched Uranium (Courtesy of Nuclear Metals, Inc.)

WOOD SPACER DETAILS
MATERIAL - PINE 2x4

Fig. 7 Uranium Scrap Drum Shipping Container (AECM-7430)

SUMMARY

The ideas on criticality given above can now be summarized by expressing a few general guides to nuclear safety. The mass parameter of criticality has been stressed because many fabrication plants, which employ a series of batch processes, use it in determining nuclear safety. It is often more desirable to use dimensional limitations, since this approach reduces the administrative control needed. However, dimensional limits are more easily applied to fairly continuous processes or static situations, such as storage arrays. In either case, safety factors are applied to critical parameters to achieve safe values.

First, allowance must be made for the possibility of double-batching caused by human, analytical, or sampling errors. Therefore, there should be a factor of at least two between the critical mass and the mass value used. The workers at Oak Ridge use a safety factor of 2.3 on mass.¹¹

Secondly, the safety factor on volume or dimensional parameters is usually 1.3. The lower factor is used here since operating errors are less likely when these limits are feasible and errors in design and construction are quite unlikely.

When dimensional limits are used, great care must be taken that the values are correct. Dimensional limits usually apply to solution-like system and must not be used for unmoderated uranium. For example, the recommended maximum thickness of a slab-like solution container is 1.4-in., while the maximum thickness of an enriched uranium metal slab is 0.7-in..¹²

Present practice dictates that total flooding of a process or storage area by water should not result in a critical configuration. For this reason, the data and analyses of fully reflected experiments are most useful. However, in some arrays the partially flooded situation can be more reactive and account must be taken of this possibility.

The smallest critical configurations involve finely divided and dispersed material. For this reason, the data derived from study of uranium solutions are usually applied to processes involving scrap solutions, chips, turnings, and sludges. Here, extreme care must be taken to account for the possible increase in the infinite multiplication of a system, due to changes in concentration caused when such systems are disturbed.

Nuclear safety should not depend on the occurrence of less than two unlikely events. As an example, consider the critical masses of the massive, fully enriched uranium metal sphere shown in Table II.

Recall that the minimum critical mass, unreflected, is about 48 kg of U-235. When completely water-reflected, the critical mass is about 22.5 kg. Therefore, the storage of 20 kg would be safe, unless more material were added concurrently with flooding. However, to account for double-batching during packing or unpacking, the usually recommended maximum mass is 11 kg.

In conclusion, I wish to emphasize that the ideas presented here do not represent a complete picture of nuclear safety problems. The general bases for criticality have been described. There are virtually an infinite number of possible fissionable systems in processing plants. Only a very few have been indicated. Further, the related problems of radiation alarm systems, of evacuation procedures, and of monitoring after a critical burst have not been described. However, it is hoped that some appreciation of the scope of preventing unwanted criticality has been presented.

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