POSSIBILITY OF EXPERIMENTAL VALIDATION OF CRITICALITY SAFETY METHODOLOGY IN SUPPORT OF UNDERGROUND FUEL STORAGE EFFORTS

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ABSTRACT

Critical systems which might be formed in geologic repositories as a result of long-term degradation of the storage media, leaching of plutonium from the storage media, and the redistribution of low concentrations of plutonium into underground sand layers or lenses can be characterized by positive reactivity feedback. Formation of such a systems can not be excluded when considering the burial of high enriched uranium or plutonium contaminated wastes or spent nuclear fuels. Although the probability of formation of a critical systems under such conditions is very low, the reliable prediction of neutron multiplication properties appears to be of great interest from a criticality safety viewpoint. At the present time, all estimations of criticality are based only on evaluated neutron data because critical experiments are not available for large systems containing small quantities plutonium distributed throughout a typically encountered matrix material such as silicon dioxide. The possibility of providing such an experiment using the large Russian critical assemblies, BFS-1 or BFS-2, is considered. It is shown that critical systems containing small amounts of hydrogenous material (polyethylene) with positive reactivity feedback can by modeled in the BFS Facility.

I. INTRODUCTION

In a recent Joint Russian/U.S. Plutonium Disposition Study\(^1\) the possibility of underground burial of plutonium in deep bore-holes was investigated by Russian and American specialists. In all considered options, plutonium was introduced into a glass or ceramic matrix with the possible addition of neutron absorbers to prevent the development of a chain reaction and some of long-lived radionuclides such as Cs-137 to prevent unauthorized withdrawal. The concentrations of plutonium considered in these studies varied from 3.8 - 29.4 wt.% for Russian glass logs and 5 wt.% for U.S. glass logs. The total amount of plutonium buried in one bore-hole can be measured by tens of metric tons. On a geological time scale, logs of vitrified plutonium will degrade and plutonium can be leached from the logs and dispersed in the surrounding rock. If, in this process, plutonium is separated from the absorber material, critical configurations can be formed. C.D.Bowman and F.Venneri\(^2\) described a number of scenarios leading to criticality in systems composed of SiO\(_2\), water and plutonium.

On Figure 1, the dependency of critical plutonium weight fraction in water-contaminated SiO\(_2\) matrices are shown for
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Figure 1. Dependencies of critical plutonium weight fraction from water molar fraction for the spheres of diameter 4 m (thick lines) and 1.4 m (thin lines). Solid lines correspond to the pure $^{239}$Pu homogeneously distributed in mixtures of SiO$_2$ with water (as in Ref.2). Dotted lines corresponds to weapons grade plutonium (5% $^{240}$Pu) homogeneously distributed in the mixture of SiO$_2$ + 0.3 Al$_2$O$_3$, with water. Dashed line corresponds to the case when weapon grade plutonium is concentrated in the layers 0.24-cm thick divided by matrix material. Critical plutonium weight fractions for four proposed BFS-1 critical assemblies discussed in the text are shown by the circular points. Proposed BFS-2 critical assembly are shown by triangular point.

reflected spherical volumes of different radii. The data obtained in Reference 2 are confirmed by our calculations. It is easy to see that, if a large volume of wet SiO$_2$ (~4 m in diameter) with a low (~0.001) weight fraction of plutonium is formed, the loss of water from this volume brings the system nearer to criticality. When criticality is actually achieved, water is rapidly evaporated from the volume and the system becomes prompt critical.

For smaller systems of wet plutonium-contaminated SiO$_2$ sand (~2 m in diameter), criticality can be achieved only at larger plutonium weight fractions (smaller critical mass values). In this case, positive reactivity feedback also exists; however, the necessary water volume fractions are much larger and evaporation of the water, as result of fission chain reaction, leads to the formation of a subcritical dry system. It is clear that energy release in this case is not very large, but as it is pointed out in Reference 2 it can be sufficient for further dissipation of plutonium and the eventual formation of a more dangerous system similar to the 4-m sphere considered above.
Critical systems like described above can be formed not only with plutonium but with any thermally fissile material (FTM) in particularly with FTM contained at small concentration in the radiative wastes. Of course, the probability of formation of autocatalytic critical systems is very small (see for example Reference 3). However, estimation of probability is nearly always required of those who want to realize a definite technical project such as underground burial of plutonium. From a criticality safety point of view it is necessary to have full confidence that parameters of the discussed autocatalytic critical systems can be predicted with acceptable reliability because it is not excluded that such systems can be formed as the results of accidents in some other sectors of the fuel cycle beyond underground burial. At the present time, all estimations of criticality are based only on evaluated neutron data because critical experiments are not available for large systems containing small quantities plutonium or other FTM distributed throughout a typically encountered matrix material such as silicon dioxide. Even though neutron cross section data for silicon, oxygen and FTM in the energy region below 1 keV (where the most of fissions and captures take place in the systems of interest) seems to be known rather well, it is uncertain that the absorption, by FTM, of neutrons that are slowed down in the SiO₂ matrix is described with the desired accuracy. Thus performing of critical benchmark appear to be of interest. We will consider, in this paper, only the possibility of providing critical experiments with plutonium. Similar experiments can be performed with uranium.

II. EXPERIMENTAL POSSIBILITIES

It is clear from the above consideration that the systems of major interest are sand (SiO₂) systems with low concentrations of the plutonium and water. At first glance the best method of investigation of such a system is the determination of kₑ by insertion of investigated media in the critical assembly using the null-reactivity method. However, the migration length in such a system is rather large (about 35 cm). The dimension of the insertion must be much greater than the migration length because, only in this case, would the neutron spectrum and spectrum of the adjoint flux in the center of insertion be sufficiently close to those for an infinite media. Even in this case, if the driver region surrounding the central insertion region has a neutron spectrum that is not much different from the spectrum of the infinite media of insertion material, the average radius of the insertion region must exceed, at least, three migration lengths (i.e. must be greater than 1 meter). Thus, the dimension of the whole assembly, including the insertion region, driver region, and the reflector, becomes close to 2 meters. Under these circumstances, it seems more reasonable to increase slightly the content of plutonium in the investigated media and perform an actual critical experiment. In either case, it is necessary to use a rather large critical facility (such as ZPPR for example) to perform such an experiment. This is shown in Figure 1 were only large SiO₂ -Pu systems have positive reactivity feedback for all water concentrations. Unfortunately, as we know, ZPPR is no longer in operation. Therefore, we consider only the large Russian critical facilities, BFS-1 and BFS-2.

The BFS critical assemblies consist of large hexagonal lattices of stainless steel or aluminum circular tubes, 5 cm in diameter, 0.1-cm thick, with a pitch of 5.1 cm. The tubes are
filled with pellets of materials under investigation. The outer diameter of pellets is, as a rule, equal to 4.7 cm. Thus, the volume fractions of the pellets, tube material, and the remaining void space are 77.02%, 6.83%, and 16.15%, respectively. The total volume of the BFS-1 assembly is 6.5 m³ (2 m in diameter and 2.1 m in height). If a reflector thickness near 30 cm is chosen, the volume of the core region becomes equal to approximately 2.5 m³ (1.45 m in diameter and 1.5 m in height). The volume of BFS-2 facility is much greater (50 m³ approximately) and the volume of core region can be equal to 35 m³ (4.2 m in diameter and 2.5 m in height) with a reflector thickness of at least 30 cm.

BFS critical facilities were designed and used for investigations in the fast reactor physics and thus a large amount of plutonium pellets are available. The majority of these pellets were manufactured from weapon grade plutonium (near 5% of $^{239}$Pu). The amount of available plutonium is approximately 800 kg. In addition, some amount of plutonium with a greater contamination of $^{240}$Pu (near 10%) is available. All plutonium pellets represent metallic disks 4.3 cm in diameter and 0.24 cm thick and are canned by a 0.04 cm thickness of stainless steel.

At the present time, there are no SiO₂ pellets available for testing in the BFS Facility. The best material for these pellets is quartz but manufacturing the quartz pellets is very expensive. In addition, the core, of necessity, will be contained in aluminum tubes. If the pellets are manufactured from a much cheaper material such as SiO₂-Al₂O₃ ceramic (20% Al₂O₃) and aluminum tubes are used the BFS lattice represents a matrix with an average density 1.86 g/cm³ and an average composition SiO₂+0.3Al₂O₃. The capture cross section of this material is greater than that of pure quartz by only about 24%. Such a matrix is more representative of real rock material than pure quartz and the isotopic composition of the BFS plutonium pellets is about the same as that for weapons grade plutonium contemplated for burial. On Figure 1, the dotted lines show the dependence of critical weight fraction on the water molar fraction, as calculated for the BFS matrix material and isotopic composition of plutonium. It is easy to see in this case that, even in very large critical systems such as a sphere of 4 meter in diameter, an autocatalytic chain reaction can not be developed. The reason is that the strong neutron absorption in $^{240}$Pu which, in a system without any hydrogen, dominates the absorption in the matrix material. Replacing a small portion of matrix material with water leads to an increase in the $^{240}$Pu resonance escape probability and a reduced critical plutonium weight fraction. When large portion of matrix material is replaced with water, the effect of neutron absorption in the hydrogen becomes the major factor and the critical weight fraction increases with water content. Thus, if homogeneous systems of weapons grade plutonium are considered, autocatalytic critical systems can be formed only after a long time when the concentration of $^{240}$Pu will be reduced by radioactive decay of this isotope ($T_{1/2} = 6550$ y compared to $T_{1/2} = 24110$ y for $^{239}$Pu).

However, it is necessary to point out that such a favorable situation takes place only for homogeneous systems. Let us consider systems in which weapons grade plutonium is not uniformly distributed in the matrix material, but is concentrated in thin layers, for example 0.24-cm thick (such as in BUS pellets). The results of corresponding calculations are shown on the Figure 1 by dashed lines. As can be seen in this case, autocatalytic critical systems can be formed only if the dimension of the plutonium contaminated matrix is sufficiently large. Thus if we want to model systems with positive hydrogen reactivity effect using weapon grade
plutonium we must consider heterogeneous systems like that which can be assembled on BFS. We begin with consideration of the experimental possibilities in BFS-1.

On BFS-1 it is possible to design the assembly of 721 aluminum core tubes arranged in a hexagonal configuration. Each core tube contains core cells. Each core cell contains one plutonium pellet above and below which ceramic pellets are placed. Thickness of one cell is equal to 10 cm. Ceramic pellets are also placed above and below the core region to provide a 30-cm-thick reflector region. Around of core region, five rows of tubes filled only by ceramic pellets will form the lateral reflector. This assembly is practically the largest one which can be done using BFS-1. Such a system is close to the 70-cm-radius spherical systems considered in Ref. 2 (see Figure 1). The average weight fraction of plutonium is equal to 11 wt.%. Heterogeneity of the core has only a slight influence on criticality. If heterogeneity effects are not take into account, the dependence of critical plutonium weight fraction on the hydrogen molar fraction would be very similar to those for the 50-cm-radius spherical systems considered in Reference 2 (see Figure 1). But in fact, the heterogeneity reactivity effect strongly increases with increasing hydrogen concentration. For example if thin cylindrical polyethylene twigs, 0.8 cm in diameter, are inserted into half of the holes between core tubes, criticality will be achieved when only 405 tubes with plutonium pellets are loaded into the core and the other tubes in the core contain only ceramic pellets. If polyethylene twigs are inserted into all holes between tubes than 316 plutonium contaminated tubes would be necessary for criticality. At last if plutonium contaminated tubes will be diluted by the tubes with ceramic and polyethylene pellets (15 volume percent of polyethylene) criticality would be achieved with 241 fuel tubes in the core. These critical systems are shown on Figure 1 by circular points. All these points lie above the dotted curve. In spite of fact that heterogeneity significantly changes the dependence of critical plutonium weight fraction on hydrogen content in the matrix, the general character of this dependence is conserved. Thus, it is possible, in BFS-1, to model undermoderated critical systems in the more interesting regions (when hydrogen content is low or absent).

For modeling the situation in which the introduction of hydrogen into the core decreases the $k_{\text{eff}}$, it is necessary to use the larger critical facility, BFS-2. We have considered an assembly with 4447 core tubes arranged in the circle of 357 cm in diameter. Each core tube contained three plutonium pellets divided by 55 cm of ceramic. These pellets formed three thin plutonium layers in the core. Above the upper plutonium pellet and below the lower plutonium pellet, thick (about 1 m) ceramic columns was represented in each core tube. Monte Carlo calculations have shown that this assembly is critical. The critical loading is equal to 671 kg of weapon grade plutonium. The weight fraction of plutonium is 2.2 wt.%. If polyethylene twigs, 0.8 cm in diameter, are inserted between core tubes, $k_{\text{eff}}$ is reduced by about 4% and the radius of the core must be increased to achieve criticality. Thus, the BFS-2 assembly shows the same dependence of criticality on hydrogen content as the dashed curve on the Figure 1. Therefore, this system represents a good benchmark for validation of computer codes and neutron data used for criticality safety estimation of large overmoderated systems.

If the critical experiments described above are performed, a number of supplemental measurements can and must be made. First of all it would be of great interest to measure the influence of $^{240}\text{Pu}$ concentration on reactivity.
This can be done by substituting part of plutonium pellets with those having an increased concentration of $^{240}$Pu. Secondly, reactivity worth of plutonium isotopes and hydrogen must be accurately measured. Thirdly, spatial distributions of plutonium isotopes fission rates and the aluminum capture rate must be measured with the help of small track detectors and foils. At last a number of spectral indices must be measured for neutron spectrum estimation. This supplement experimental data allow more reliable extrapolation of experimental critical data to systems with other plutonium isotopes and water content values in SiO$_2$.

Experiments like those described above can also be performed with highly enriched uranium. The only problem necessary to keep in mind in this case is that one of major reasons for underground burial of highly enriched uranium fuel is large concentration of uranium-236 that is found in highly enriched spent fuel from research and naval reactors. Direct measurements of the influence of this isotope on criticality in the experiments on the BFS critical assemblies is not possible. The estimation of the influence of this isotope can be done only indirectly - by measurement of uranium-236 reactivity worth and fission rate ratio.

III. SUMMARY

In contrast with pure $^{239}$Pu, weapon grade plutonium (5% of $^{240}$Pu) cannot be configured into an autocatalytic homogeneous critical system when mixed with rock materials. Nevertheless such a system can be formed if plutonium is heterogeneously distributed in a large rock matrix.

Experimental capabilities of the Russian BFS facilities allows the performance of critical benchmark experiments for verification of codes and neutron data used for criticality safety predictions for large systems of low neutron absorbing matrices containing low concentrations of TFM. Such systems can be, in principle, formed as the result of degradation of buried, vitrified, TFM barring logs or perhaps in some other scenario. No benchmark critical experiments of such a type exist today.

REFERENCES


