June 1, 2015

To: J. N. McKamy Manager, US DOE NCSP
From: D. G. Erickson, Deputy Chair, US DOE NCSP CSSG dge

Subject: CSSG Tasking 2014-03 Response

In Tasking 2014-03 a subgroup of the Criticality Safety Support Group (CSSG) was requested to draft a revised chapter on Criticality Safety for a revision to the Plutonium Handbook, last revised in 1980.

The drafting team consisted of the following CSSG members:
- T. P. McLaughlin (lead)
- D. G. Erickson
- M. Brady-Rapp
- A. S. Garcia

The new Chapter was reviewed by the entire CSSG. Comments were incorporated into the final 'draft' version of the Chapter that is attached to this memo.

cc: CSSG Members
M. Dunn
A. N. Ellis
L. Scott

Attachment 1: Response to CSSG Tasking 2014-03 (Draft Chapter)
Attachment 2: Approved Tasking 2014-03

NUCLEAR CRITICALITY SAFETY OF PLUTONIUM

By the USDOE Criticality Safety Support Group

Introduction

Materials that can support a nuclear chain reaction, commonly referred to as fissile or fissionable materials, present a unique hazard to those who work with those materials and those in close proximity to them. Plutonium and enriched uranium are the most common fissile materials, both as pure materials and when mixed with other materials, such as in solutions during various chemical processes.

This unique hazard is the accidental generation of a fast, often instantaneous, fission chain reaction, that is, a criticality accident. Such events can result in injurious and sometimes lethal doses of both neutron and gamma radiation to workers in the immediate vicinity of the accident. This radiation results predominately from the fission process itself, but subsequent, lesser amounts can be associated with the radioactive decay of the resulting fission products. Nuclear criticality safety, commonly known simply as "criticality safety," is usually defined as:

Protection against the consequences of a criticality accident,
preferably by prevention of the accident.

The underlying principles that are associated with conditions necessary to achieve a fission chain reaction are complex and may often be counter-intuitive. Furthermore, studies of past criticality accidents have shown that conditions that may be conducive to a criticality accident have usually not been apparent, resulting in no forewarnings of the accident. However, in spite of the potential risks, experience has shown that extensive operations with fissile material can be performed safely and economically when proper precautions are exercised.

It is noted that essentially all facilities that process quantities and forms of fissile materials that pose a criticality hazard rely on criticality safety expertise either within the facility or company, or under contract to the company. Operations with fissile materials are also strictly regulated by government requirements and subject to regulatory oversight organizations. This chapter is intended for persons who are not criticality safety subject matter experts and is intended to introduce them to this unique hazard. Design efforts and actual work with more than gram quantities of plutonium need to be conducted in concert with guidance from the appropriate criticality safety subject matter experts.

This chapter has been significantly abbreviated from its predecessor in the two earlier editions of this handbook. This reflects the fact that extensive criticality safety documentation is now available in a small number of significant reference documents that have been generated over the approximately 50 years since the original text of this handbook was produced. Summaries of key points from these reference documents are presented in the following sections of this chapter. This chapter also includes a list of current criticality safety national consensus standards with selected subcritical limits from those standards, plus a review of process criticality accidents and lessons learned from those accidents.

Process Facility Criticality Accident History and Lessons Learned

There have been only 22 criticality accidents reported worldwide in the processing of enriched uranium and plutonium (Ref. 1). Of these twenty-two, eight involved plutonium and fourteen involved enriched uranium. A chronology of these non-reactor criticality accidents is provided in Figure 1.

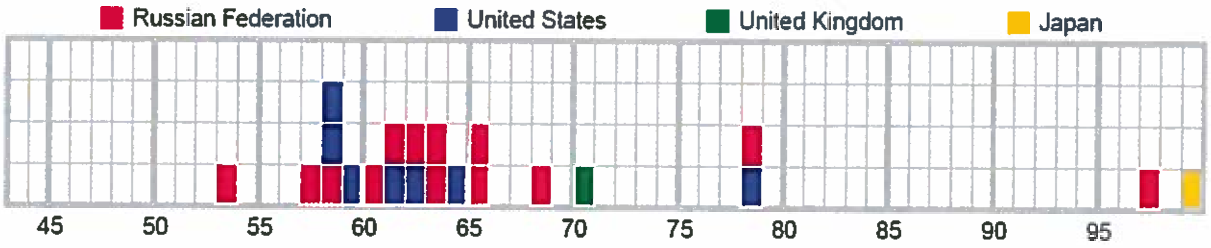

Figure 1. Chronology of process criticality accidents.

Figure 1 Chronology of Process Criticality Accidents

Notable features of these process accidents include:

- Twenty-one occurred with the fissile material in solutions or slurries.
- One occurred with metal ingots.
- None occurred with powders.
- Eighteen occurred in manned, unshielded facilities.
- Nine fatalities resulted.
- Three survivors had limbs amputated.
- No accidents occurred in transportation.
- No accidents occurred while fissile material was being stored
- No equipment was damaged.

As is evident from Figure 1, there was about one accident per year in both the USSR and USA from the mid-1950s through the mid-1960s and since then there has been a dramatic reduction in the accident frequency. This was a direct result of replacing large vessels (so-called "unfavorable" geometry vessels) for the processing of fissile-bearing liquids with vessels more conducive to neutron leakage from the system (so called "favorable" geometry vessels). Examples of favorable geometry vessels include long pipes of limited diameter, typically 15-cm diameter or less, and slab or annular tanks with the smallest dimension being only several cm thick. It is noted that when reliance is on favorable geometry, challenges to the stability of the geometry, such as overpressure events, must be considered. For small batch solution processing, limited fissile mass (<500 grams of plutonium) and solution volume (5 liters or less) can also readily control criticality accident risks.

In addition to the highlights listed above, two of the major observations from studying these 22 accidents are:

- No accident resulted in significant radiation consequences beyond the facility site, either to people or to the environment. This reinforces a commonly-held contention that criticality accidents are similar to small, bench-top scale chemical explosions in their personnel and environmental consequences; i.e., they are immediate-area worker safety issues.
- No accidents were solely attributed to equipment failure. To the contrary, all accidents were predominately influenced by some combination of the following design, managerial and operational attributes: communications, procedures, fissile material accountability and accumulation, vessel geometry and volume, operator knowledge, new, restarted, or one-of-a-kind operations, and unanticipated movement of solutions.

Major lessons learned from the process criticality accidents are:

- Unfavorable geometry vessels should be avoided in areas where high-concentration (grams of fissile per liter of liquid) solutions might be present.
- Transition from favorable to unfavorable geometry needs specific attention.
- Important instructions, information, and procedural changes should always be in writing.
- The processes should be well-understood so that credible abnormal conditions can be foreseen.
- Criticality control should be part of an integrated program that includes fissile material accountability, conduct of operations, configuration management, training, oversight, etc.
- Operations personnel should know how to respond to equipment malfunctions and to their own errors.
- Operations involving both organic and aqueous solutions require extra diligence in understanding possible upset conditions if mixing of the two phases is credible.
- Operations personnel should be made aware of criticality hazards and empowered to implement a stop-work policy.
- Operations personnel should be trained to understand the basis for and to adhere to the requirement for always following procedures. Lack of adherence to procedures, either inadvertently or intentionally, has been a major contributor to several accidents.
- Process supervisors should ensure that operators under their supervision are knowledgeable and capable.
- Equipment should be designed and configured with ease of operation as a key goal.
- Senior management should be aware of the criticality accident hazard and its consequences.
- Operations personnel should be trained in the importance of not taking unapproved actions after an initial evacuation.
- Readouts of radiation levels in areas where accidents may occur should be considered.

- Hardware that is important to criticality control but whose failure or malfunction would not necessarily be apparent to operations personnel, should be used with caution.
- Criticality alarms and adherence to emergency procedures have saved lives and reduced exposures.
- Process supervisors should ensure that the operators under their supervision are knowledgeable and capable.
- Policies and regulations should encourage self-reporting of process upsets and to err on the side of learning more, not punishing more.
- Regulations should exist which promote safe and efficient operations.
- Regulators, like process supervisors, should ensure that those they regulate are knowledgeable and capable.

While not having been a contributor to an accident, when performing operations with plutonium solutions, generally during separations operations where concentration is being controlled, the plutonium could form a gel if acid molarity is insufficient. This could significantly increase the effective plutonium concentration as the solution becomes a slurry in a very little time.

Criticality accident risks will not vanish as long as significant quantities of fissile material exist. However, sufficient knowledge has been gained from planned experiments and from accidents to provide a high degree of confidence that, with appropriate support from senior management, reasonable diligence on the part of the criticality safety staff and operating personnel, and adherence to codified fundamental safety principles and guidance, accident likelihoods can be maintained at the current low level or possibly be reduced even further.

Fundamental Guidance for the Control of Criticality Accident Risks

All facilities handling gram quantities (or more) of purified fissile material today should have dedicated, or contracted, criticality safety specialists who provide specific process evaluations and guidance for process vessel design and layout to prevent criticality accidents. While regulations governing criticality accident prevention vary somewhat by country, the guidance contained in the American National Standards in Nuclear Criticality Safety, i.e., the ANSI/ANS-8 series of national consensus standards, has generally been adopted worldwide and is discussed below.

The ANSI/ANS-8 series of national consensus standards provide the general basis of all criticality safety guidance worldwide. These standards were an outgrowth of company policies and procedures nationwide in the US from the 1940s and 1950s and were first issued as a single document, the N16.1 Standard, in 1964. The series has now grown to the 18 documents listed in Table 1, with ANS-8.1 being the one from which all the others have been spawned.

A fact that is implicitly woven throughout the lessons learned from past accidents is that all accidents have been dominated by design, managerial and operational failures. Thus, the most significant first step in the control of criticality accident risks is management's establishment of responsibility for nuclear criticality safety. This administrative practice is discussed in ANS-8.1 and elaborated on in ANS-8.19. A key requirement stated in both of these standards is:

> *Before a new operation with fissionable (fissile) material is begun, or before an existing operation is changed, it shall be determined that the entire process will be subcritical under both normal and credible abnormal conditions.*

Since the advent of the American National Standards, the International Standards Organization (ISO) has begun issuing similar guidance, generally patterned after the ANS-8 standards. The key requirement quoted above is stated similarly in the corresponding international document with the use of the words "reasonably foreseen" in place of the word "credible." This is mentioned since the determination of "credible" is always highly judgmental and the words "reasonably foreseen" might assist in providing insight into this issue. Accident risks will never be entirely eliminated and must be controlled to the lowest practical level.

Other administrative issues discussed in these two standards include: written procedures; fissile materials control; operational control; operational reviews and emergency procedures. ANS-8.1 also discusses technical practices that are often more fully developed in the individual standards listed below. These technical practices include: controlled parameters such as fissile mass and vessel geometry; neutron absorbers; the double contingency principle (defense in depth) and neutron moderation.

Table 1 ANS-8 National Consensus Standards

Standard	**Title**
ANS-8.1	*Nuclear Criticality Safety in Operations with Fissionable Materials Outside Reactors*
ANS-8.3	*Criticality Accident Alarm System*
ANS-8.5	*Use of Borosilicate-Glass Raschig Rings in Solutions of fissile Material*
ANS-8.6	*Safety in Conducting Subcritical Neutron-Multiplication Measurements In Situ*
ANS-8.7	*Nuclear Criticality Safety in the Storage of Fissile Materials*
ANS-8.10	*Nuclear Criticality Safety Controls in Operations with Shielding and Confinement*
ANS-8.12	*Nuclear Criticality Control and Safety of Plutonium-Uranium Fuel Mixtures Outside Reactors*
ANS-8.14	*Use of Soluble Neutron Absorbers in Nuclear Facilities Outside Reactors*
ANS-8.15	*Nuclear Criticality Safety Control of Selected Actinide Nuclides*
ANS-8.17	*Criticality safety Criteria for the handling, Storage, and Transportation of LWR Fuel Outside Reactors*
ANS-8.19	*Administrative Practices for Nuclear Criticality Safety*
ANS-8.20	*Nuclear Criticality Safety Training*
ANS-8.21	*Use of Fixed neutron Absorbers in Nuclear Facilities Outside Reactors*
ANS-8.22	*Nuclear Criticality Safety Based on Limiting and Controlling Moderators*
ANS-8.23	*Nuclear Criticality Accident Emergency Planning and Response*
ANS-8.24	*Validation of Neutron Transport Methods for Nuclear Criticality Safety Calculations*
ANS-8.26	*Criticality Safety Engineer Training and Qualification Program*
ANS-8.27	*Burnup Credit for LWR Fuel*

Criticality Data Compendia and Good Practices Guides

The original version of the Pu Handbook was published in 1964, and a revision was published in 1980. However, the chapter on Nuclear Safety and Criticality, Chapter 27, did not receive any updates. Since the time of the original publication access to critical parameter data for plutonium (and uranium) systems has improved dramatically. Additional critical experiments have been performed, and the capabilities of the computer codes have improved. With the advent of the internet, practitioners can access digital copies of relevant guides and handbooks from many locations. Some of the more commonly utilized handbook references are: TID-7016 (Ref. 2), ARH-600 (Ref. 3), LA-10860 (Ref. 4), and LA-12808 (Ref. 5). Summaries of the contents of and differences among each of these are provided in the following paragraphs.

TID-7016, Rev. 2, *Nuclear Safety Guide* (Ref. 2) was originally issued in 1956 as a classified Atomic Energy Commission report, then in 1957 was published in an unclassified format, with almost no changes. It was a collaborative effort by practitioners from Oak Ridge National Laboratory (ORNL), the Hanford Site, Los Alamos Scientific Laboratory (LASL) and the Rocky Flats Plant. The purpose of the Guide was to help improve the nuclear criticality safety of operations with fissile materials. Since then there have been two revisions, the last of which, Revision 2, was published in 1978. The purpose for the revisions was to update the data as more experimental information became available, and to utilize improved computational methods.

The Guide laid the groundwork for the N16.1 Standard and what has now become the ANSI/ANS-8 series of standards. In later years the Guide expanded upon the requirements of the original N16.1 Standard, with guidance related to implementation, the 'how' vs. the 'what' of the standards. The Guide also presents 'single-parameter subcritical limits'. Based on current information it is presumed these subcritical limits equate to a calculated k_{eff} of approximately 0.98 based on the codes and cross sections available at that time.

ARH-600, *Criticality Handbook*, (Ref. 3) was last published in 1968, and was based on calculations performed by practitioners at the Hanford site, and for use by other Hanford criticality safety practitioners. Most of the data presented were presumed to be critical data ($k_{eff} = 1.00$) based on the available codes and cross sections. When available critical experiment data was available it was utilized as a check. ARH-600 presents many unique curves of data/parameters not generally found in other references.

LA-10860-MS, *Critical Dimensions of Systems Containing* ^{235}U, ^{239}Pu, *AND* ^{233}U (Ref. 4) was published in 1986 and was a revision to, and expansion of, a predecessor document, TID-7028. The expansion was modest, since most of the relevant data was generated prior to the issuance of TID-7028, with a few calculational interpolations and extrapolations added. The data presented are critical data ($k_{eff} = 1.00$) with no safety margins included. Two figures from this document are presented below to assist the reader in appreciating how the critical state of ^{239}Pu varies with the medium.

Figure 2 depicts the mass that will just be critical, first as a sphere of high-density (alpha phase) metal, and then as an idealized homogenous mixture of plutonium and water. The dilution with water increases as one progresses from right to left until one reaches the asymptote at about 7.5 grams of plutonium per liter of mixture. At lesser densities a homogeneous mixture cannot be made critical. The two curves are for unreflected spherical systems and for systems reflected by thick (i.e., ~30 cm) water. Also shown is some actual data highlighting the effect of both ^{240}Pu and nitrate radicals on the critical mass. Figure 3 shows how the system volume varies over this same density range.

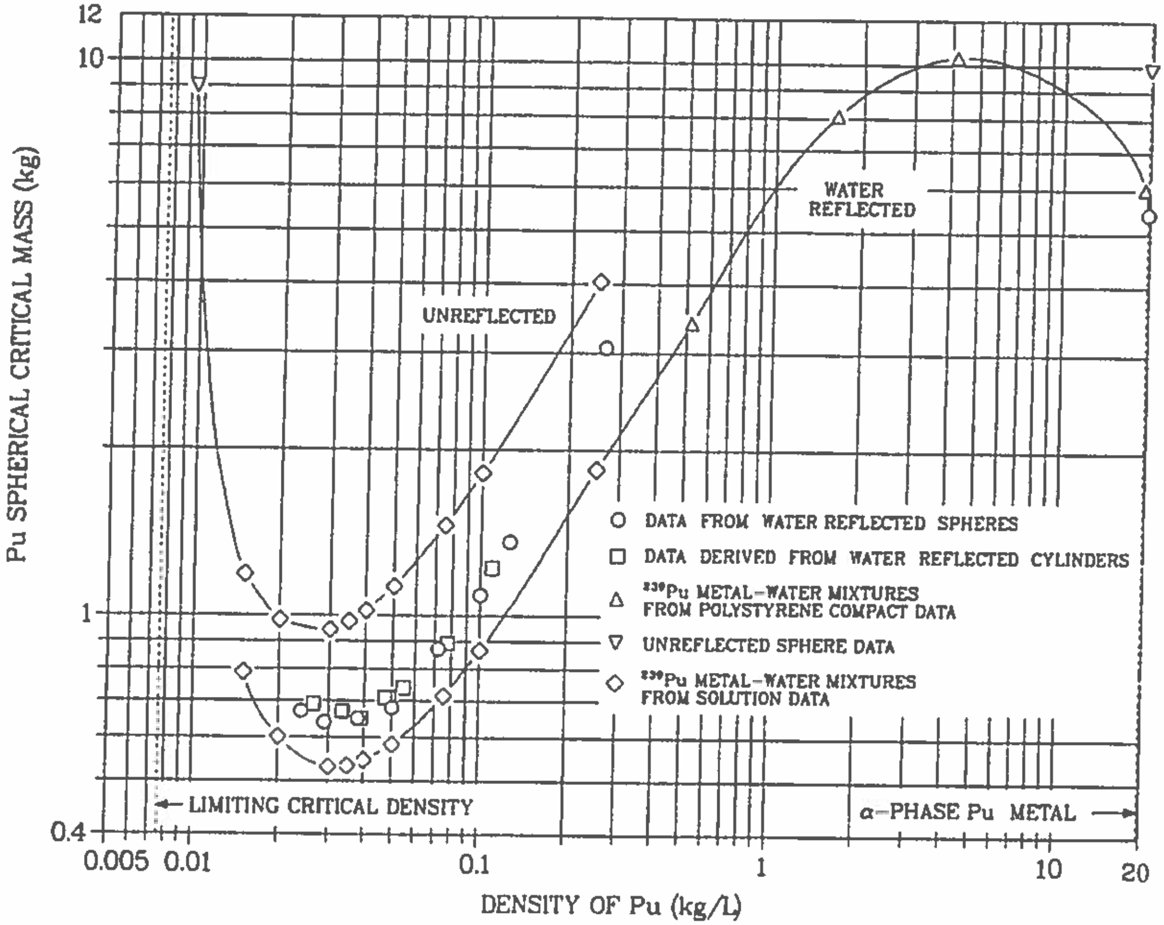

Fig. 31. Critical masses of homogeneous water-moderated plutonium spheres. The points suggesting an intermediate curve apply to water-reflected Pu $(NO_3)_4$ solution with 1 $\underline{N}$ HNO_3 and 3.1% ^{240}Pu content of the plutonium.

Figure 2 **Critical Masses of Homogeneous Water-Moderated Plutonium Spheres [LA-10860-MS]**

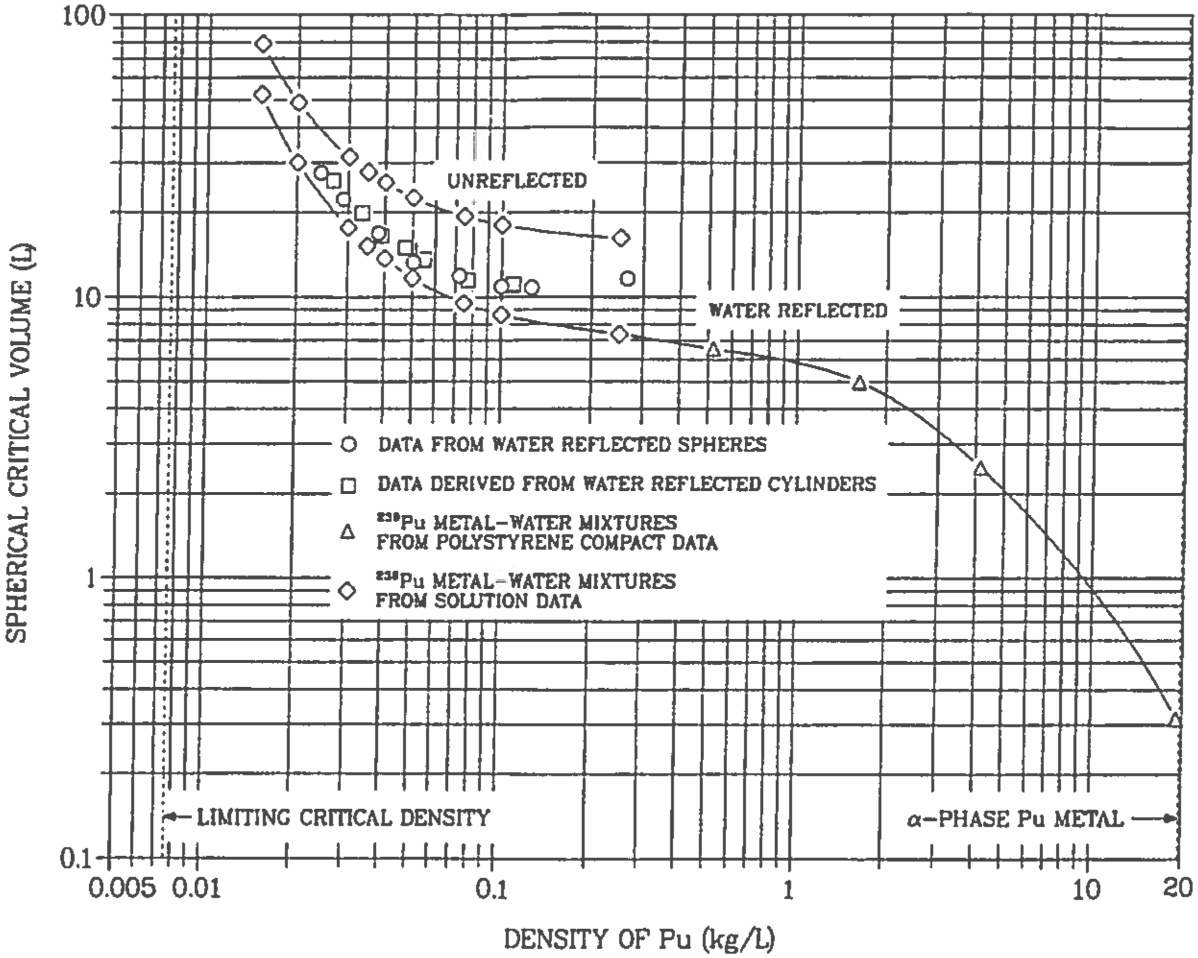

Fig. 32. Critical volumes of homogeneous water-moderated plutonium spheres. The points suggesting an intermediate curve apply to water-reflected Pu $(NO_3)_4$ solution with 1 N HNO_3 and 3.1% ^{240}Pu content of the plutonium.

Figure 3 **Critical Volumes of Homogeneous Water-Moderated Plutonium Spheres [LA-10860-MS]**

LA-12808, *Nuclear Criticality Safety Guide*, (Ref. 5) was published in 1996, and is, partially, Revision 3 of TID-7016. This document corrected all known errors from the previous TID-7016 (Ref. 2), and incorporated many changes suggested by the criticality safety community. Some chapters and sections were chosen to not be carried forward to this new document. There was little new experimental data but extensive revised calculations were incorporated to extend the document's usefulness.

Critical Experiment Capabilities in the US

The landscape, related to performing critical experiments, has changed significantly over the decades since the 'Plutonium Handbook' was first published. At one time most of the primary DOE facilities had critical experiment capabilities. Today there is one primary critical experiment facility in the United States, the National Criticality Experiments Research Center

(NCERC) located at the Nevada National Security Site (NNSS). NCERC currently maintains four critical assembly machines, with a substantial special nuclear material (SNM) inventory, to support a variety of configurations. There are also some other, more specialized, facilities, such as a water moderated lattice facility at Sandia National Laboratory.

There were also many international critical experiment facilities. Today most of those facilities have closed, and the remaining have uncertain futures.

<u>References</u>

1. McLaughlin, T.P., et al, *A review of Criticality Accidents – 2000 Revision.* LA-13638, Los Alamos National Laboratory, Los Alamos, NM, May 2000.

2. Thomas, J.T., ed. *Nuclear Safety Guide.* TID-7016, Rev. 2, U.S. Nuclear Regulatory Commission, June 1978.

3. Carter, R.D., Kiel, G.R., and Ridgway, K.R. *Criticality Handbook.* ARH-600, Atlantic Richfield Hanford Company, Richland, WA, June 1968.

4. Paxton, H.C. and Pruvost, N.L. *Critical Dimensions of Systems Containing U-235, Pu-239, and U-233.* LA-10860-MS, Los Alamos National Laboratory, Los Alamos, NM, July 1987.

5. *Nuclear Criticality Safety Guide.* LA-12808, Los Alamos National Laboratory, Los Alamos, NM, September 1996.

Attachment 2: Approved Tasking 2014-03

CSSG TASKING 2014-03, Rev 1
Date Issued: February 27, 2015

Task Title: ***Draft Criticality Safety Chapter for Revised Pu Handbook***

Task Statement:

The current revision of the Pu Handbook came out in 1980 and was unchanged from the original issue in 1967. Chapter 27 of that revision addressed criticality safety. An effort has been started to revise the handbook, and the criticality safety chapter/section should be updated commensurate with changes in knowledge and abilities over the last decades.

The CSSG is being tasked with drafting the updated criticality safety chapter for a revised Plutonium Handbook.

Tasking revised due to member availability and deliverable dates revised accordingly.

Resources:

CSSG Task 2014-03 Team Members:

- Tom McLaughlin (Team Leader)
- Mikey Brady-Raap
- David Erickson
- Adolf Garcia

Contractor CSSG members of the team will use their FY14 NCSP CSSG support funding as appropriate; DOE CSSG members of the team will utilize support from their site offices. It is up to the team members to utilize other expertise, or include other interested parties, as can be made available to support the tasking, without incurring additional CSSG expenses. No travel is anticipated to be necessary to support this tasking.

Task Deliverables:

1. CSSG Subgroup to hold task 'kickoff' telecom by August 26, 2014
2. CSSG Subgroup to provide draft chapter to full CSSG for review: March 5, 2015
3. Full CSSG to provide review of chapter to Task Team Leader: March 24, 2015
4. CSSG Subgroup to provide finalized chapter to NCSP Manager: March 31, 2015

Task Completion Date: March 31, 2015

Signed: 2/27/2015
Jerry N. McKamy, Manager US DOE NCSP
Director NA-00-10