United States Government **Department of Energy**

Memorandum

Idaho Operations Office

Date: July 24, 2006

Subject: Recommendation in response to Task 6

To: Jerry McKamy, NNSA
DOE-HQ, NA-171, OFO

This memorandum provides the Criticality Safety Support Group (CSSG) response to Task 6 from the Nuclear Criticality Safety Program (NCSP) manager. The CSSG was tasked to address essential (for criticality safety) and optional (for cost efficiency) needs in the broad areas of criticality safety support and nuclear data that can only be filled by a facility capable of achieving solution criticality with other than pure uranyl nitrate. The complete text of the tasking is provided as Appendix A. However, the CSSG is not in a position nor does it have the resources to address the specific issues in the tasking. Until the development of GNEP is more mature, it is impossible to identify all of the data needs and to be any more specific about programs, projects or facilities that would benefit from the data. In order to identify all international sources of data would be a major research effort, beyond the scope of the CSSG. Also, without knowing the details of what data are required, and what international sources are available, it is not possible to identify the specific types of solutions and experimental capabilities required. As a result of these limitations, the CSSG response is by necessity general.

It is the consensus of the group that the needs for actinide solutions experimental capabilities, to support the GNEP activities of fuel reprocessing and manufacturing, are clear.

Looking back in history, at the major fuel reprocessing facilities, we find that they all relied on the existence of critical facilities (usually a dedicated facility) to support their ongoing operations. In many cases these facilities were used with just enough rigour, without formal experiments of benchmark quality, to support changes to existing processes. This was done in the interest of safety, efficiency and the expediency necessary in the old Cold War days.

Although expediency might not play as strong a role as it did (and maybe it does to alleviate the dependency on foreign oil), safety and efficiency are as important as ever. It is very hard to imagine, without a rigorous in-depth look at the technical issues, that the U.S. could develop all the new missions related to GNEP, without the proper data to validate computer analysis in support of the safety of the proposed operations and equipment.

The CSSG members unanimously support the conclusion that there is a definite need to maintain the capability to perform critical experiments with solutions (in addition to Super SHEBA) in the U.S. Discussions from CSSG meetings are summarized below and indicate that the broad range of criticality safety support and nuclear data needs can only be provided by an assembly capable of achieving solution criticality (other than uranyl nitrate). Appendix B contains statements of current and anticipated needs from individuals who have been active in major NCSP activities over many years and have a strong awareness of both nuclear data and critical experiment needs. This appendix, previously presented as Appendix C in the CSSG response to Task 5, is included here because its contents are also pertinent to Task 6.

The CSSG acknowledges that there are experimental resources worldwide, such as critical experiments facilities in France, Japan and Russia, that could, in principle, supply some, but not all, of the needs identified in Appendix B. A thorough cost analysis of the capabilities presented by these resources was beyond the resources of the CSSG. However, the CSSG was unanimous in its agreement that several key issues made the case both for a facility capable of achieving solution criticality with other than pure uranyl nitrate, as well as the continued operation of the ORELA facility.

Key issues are clearly defined.

1) The Advanced Fuel Cycle concept is that all of the actinides are recycled back into the fuel stream. Generally, this involves dissolving the fuel, separating out the fission products, and conditioning the residual fuel back into solid form. While doing this, the higher-mass isotopes of plutonium, as well as americium and curium build up, and the transition from fluid to solid form in the fuel reprocessing involves systems that have intermediate energy neutron spectra. The nuclear data for these actinides in this energy range are not well known and integral measurements are sparse or non-existent.

   The safety basis for efficiently-sized equipment, in terms of inventory and throughput, will require the demonstration of safe margins of subcriticality. This involves the validation of the NCS transport analyses and nuclear data with benchmark critical and/or subcritical experiments. Certainly, these partially moderated systems, which include the higher actinides, require better differential and integral data to support these validations.

2) Some capabilities lost to the DOE cannot be performed in other countries or with existing U.S. facilities. U.S. integral data for plutonium solutions is generally quite poor. French integral data appear to be much better. The U.S. has lost the capability to perform plutonium solution experiments. Russia has, in large part, relied on U.S. data until the French data became widely available. France is gradually giving up their capabilities and Japan has made a political decision not to allow plutonium into their STACY and TRACY facilities even though that was their original intent and design

3) It was judged by the CSSG that the U.S. must regain Pu solution criticality capabilities within its borders. Critical solution data with the other actinides are also judged important. We note that the shutdown of the Plutonium Critical Mass Laboratory at Hanford handicapped the plutonium processing facilities in their ability to make process and equipment changes. Should we design and build new actinide process plants, we will be substantially more handicapped if we do not correct this deficiency. Having this capability at a dedicated DOE solution critical experimental facility will assure the U.S. that its R&D and production facilities are not held hostage to the availability or schedules of others. It is also judged that this facility would provide more cost-effective support to

the GNEP related missions. Although the details are not yet well defined, GNEP will clearly require nuclear data and high quality critical experiment to support actinide processing and reprocessing as there will need to be benchmark critical experiments for code validation purposes for solution systems with concentrations of actinides far beyond previously measured. As a cautionary note, the ability to generate nuclear data within the U.S. would be lost were the ORELA facility, or other equivalent facility, not maintained in an operational state.

Adolf Garcia
CSSG, Chairman

cc: CSSG Members
J. McKamy, DOE-HQ, NA-171, OFO
J. Felty, SAIC, NA-171, OFO

Appendix A

CSSG Tasking 2006-06

TITLE: Assessment of Criticality Safety and Nuclear Data Needs Requiring Solution Critical Experiments Involving Other than Uranyl-Nitrate Solutions.

TASKING: The CSSG is requested to identify essential (for criticality safety) and optional (for cost efficiency) needs in the broad areas of criticality safety support and nuclear data that can only be filled by a facility capable of achieving solution criticality with other than pure Uranyl-Nitrate. For each need (essential or optional) identified, a specific DOE program/project/facility that would benefit from the information must also be provided. All international sources of the identified data needs should be identified including accessibility to existing data and the potential to acquire data in the future. The CSSG should identify the Types of solutions and experimental capabilities required to address the gaps that remain once the global situation is considered. Considerations of siting and funding for such a capability is NOT within the scope of this tasking.

DELIVERABLE: A formal written report to the NCSP Manager.

DUE DATE: June 30, 2006

Appendix B

Statements from Individuals

<u>**Mike Westfall**</u> (Oak Ridge National Laboratory)

My understanding of the Advanced Fuel Cycle objective is that all of the actinides are recycled back into the fuel stream.

Generally, this involves dissolving the fuel, separating out the fission products, and conditioning the residual fuel back into solid form.

Two situations develop:
1) The higher-mass-number isotopes of plutonium, as well as americium and curium build up.
2) The transition from fluid to solid form in the fuel reprocessing involves systems, which establish intermediate-energy neutron spectra – in which the nuclear data for these actinides are not well known and integral measurements are sparse or non-existent.

The safety basis for efficiently sized equipment, in terms of inventory and throughput, will require the demonstration of safe margins of sub-criticality. This involves the validation of the NCS transport analyses and nuclear data with benchmark critical and/or sub-critical experiments. Certainly, these partially moderated systems, which include the higher actinides, require better differential and integral data to support these validations.

I have attached a rough table of NCS needs for the Advanced Fuel Cycle, which summarizes these issues. The nuclear data testing described above could be performed with both generic critical and sub-critical experiments. Engineering mock-up experiments will also be required to demonstrate the safety of unusual material/geometry combinations in equipment and/or separation/isolation in plant layout.

Potential Criticality Safety Technology Needs Arising from the Implementation of Fuel Reprocessing and Recycling in Advanced Burner Reactors

(Assumed: all actinides go back into fuel, all fission products go into waste stream)

<u>DIFFERENTIAL DATA NEEDS</u>

Transuranic Actinide Data (Cross Sections, Nu, Chi, Decay Data)

- Improved Fuel Exposure Prediction of Spent Fuel Isotopics (Actinides & Fission Products)
- Improved Prediction of Spent Fuel Reactivity Worth for NCS Burnup Credit for More Efficient Sizing & Inventory of Reprocessing Equipment

- Improved Prediction of Neutron Radiation Source Terms, Required Neutron Shielding and Subsequent Neutron Reflection in NCS Evaluations

Improved Nuclide Cross-Section Data for Isotopes Acting as Chemical Reagents (Effects of Neutron Moderation & Absorption)

INTEGRAL DATA NEEDS

Identify, Perform & Verify Critical and Sub-Critical Experiments for Generic Physics and/or Engineering Mockup Applications

- Generic-Physics Critical Experiments to Demonstrate Material Reactivity Effects in Systems with Neutron Spectra Pertinent to These Fuel- Cycle Applications
- Engineering Mock-up Critical Experiments to Demonstrate Capability to Analyze Fuel Cycle Applications with Unusual Material/Geometry Design Features
- Sub-Critical Experiments to Verify the Efficacy of Differential Data in the Analysis of Subcritical Experiments Designed to Simulate Sub-Critical Fuel Cycle Applications

NEUTRON TRANSPORT CAPABILITY NEEDS

Nuclide Separation Process Systems (Wet-& Pyro- Chemistry Processes, Electro-Refining Processes, that is: UREX+, ..., etc.)

- Demonstrate Capabilities for Treating Temperature Effects in Neutron Transport (Neutron Spectra, Absorption Probabilities, Media Densities as Functions of Temperature)
- Application of Sensitivity/Uncertainty Methods & Integral Data to Validate Analytical Capabilities (Transport Methods & Differential Data) Against Pertinent Benchmarks

Advanced Vessel, Storage & Transfer Equipment Geometries

- Geometric Simulation (Efficient Dissolution, Separation Throughput)
- Coupled and/or Single Unit Isolation (Efficient Plant Layout & Maintenance)
- Application of Sensitivity/Uncertainty Methods & Integral Data to Validate Analytical Capabilities (Transport Methods & Differential Data) Against Pertinent Benchmarks

Advanced Fuel Fabrication

- NCS Evaluation of Remote Operations for Highly Radioactive Fuel Fabrication
- NCS Evaluation of Shielded Facilities & Equipment for Storing & Transfer of Highly Radioactive Fuel
- Application of Sensitivity/Uncertainty Methods & Integral Data to Validate Analytical Capabilities (Transport Methods & Differential Data) Against Pertinent Benchmarks

<u>Madeline Feltus</u> (U. S. Department of Energy)

Although the separation chemists may think that they have all the information they need for Uranium and PU/minor actinide separations, and they can "conservatively apply" the ancient test results, we need to be VERY careful about using the historical data we have from previous efforts. The solutions criticality tests at the LANL SHEBA facility used pure uranyl sulfate solutions, Pu solutions, etc, and did NOT have any minor actinides "contaminating" the tests. Theses results may not be suitable for minor actinide bearing solutions, especially with Neptunium, Americium, and further up the periodic table. I would use the following statement as a springboard for discussion, modification etc:

Although criticality experiments have been performed with uranium and plutonium fuels, separate fissile materials, in fuel pin tests and aqueous solution tests (e.g. SHEBA at LANL), the current database does NOT include tests with solutions containing MINOR ACTINIDES that would be present in spent fuel separations activities. Various transuranic isotopes, such as Plutonium Neptunium, Americium, and higher atomic mass elements, will be present in high concentrations as we seek to reprocess spent fuel and recycle fast reactor, deep-burn fuel. Critically benchmark tests, including aqueous solution experiments will be absolutely necessary to assure that criticality safety can be maintained for advanced spent fuel reprocessing, Richard Anderson (Los Alamos National Laboratory)

I think we need such a solution capability mostly because I believe that our influence with other nations comes from our expertise. If we lose the expertise, we lose influence.

Somewhat more specifically, out dealings with the French have been on an exchange basis. We give them our data & they give us their data. Until now, they've not been interested in selling us data. This is how we got CRAC and SILENE data and how we got benchmark data. The (mostly foreign) plutonium solution data in the benchmark handbook came as a result of this exchange process.

Second, the original 93-2 Defense Board recommendation had it right on how to do criticality safety. First is direct comparison with experimental data, second is use of validated calculations (which also requires experimental data), and third is criticality safety analysis with adequate safety margin. The alternative to understanding what you're doing based on experimental results is to do things empirically. We did that for the first 20 or more years of the US nuclear program, so it's not impossible to do (we lost a few people during that time too). What's not at all clear is if that's acceptable today. It is clear that this approach can make costs prohibitive. See AVLIS as an example (we won't know for a few years whether the MOX plant is going down that road as well) and these are projects operating in arenas that we claimed to understand quite well compared to higher actinides.

Finally, I think the higher actinides are in trouble everywhere, not just at intermediate energies.

I think the idea of doing anything that meets a reasonable definition of "new activity" without a US experimental verification simply won't be tolerated.

Hans Toffer (Fluor Government Group, Hanford)

Potential Criticality Safety Technology Needs Arising from the Implementation of Fuel Reprocessing and Recycling in Advanced Burner Reactors

In the planning stages for an Advanced Fuel Cycle Initiative (AFCI), the need for criticality data essential to design considerations has to be firmed up. The AFCI will require facilities where both critical and subcritical measurements can be performed on Plutonium (Pu) and Uranium (U) solutions with and without contaminants. These contaminants can be other actinides, selective fission products, or know additives. The measurement facility should be of dual nature. High precision solution measurement capabilities should be at the Diverse Assembly Facility (DAF). A second facility at the reprocessing plant would provide for quick turnaround measurements for either subcritical or critical solutions. Based on my experience from the Hanford N Reactor the ability to perform measurements on solutions at Hanford was essential to the efficient and safe reactor and reprocessing operation.

Data on Pu – U – other solutions could be procured from other nations, however, such an arrangement could subject the new initiative to undesirable political influences. After all, the basic objective of the new initiative is to support energy independence, therefore control over new reprocessing and supportive data needs has to be domestic.

Use of conservative data on Pu or U solutions would be of some use for initial consideration; however, for detailed design the existing data is too sparse or non-existent especially when it comes to solutions with various elemental mixtures. Use of overly conservative data could limit the efficiency of the new facility. Besides solutions, fuel feedstock materials will require critical/subcritical measurements also.

It is paramount that a decision about critical solution slurry, powder measurement capability for the new initiation be made now. It would take 5 – 7 years before useful data could be obtained from a liquid measurement system at the DAF.

Highest priority has to be assigned to solution measurement capability at the DAF. A commitment is required now for meaningful supportive data for the new fuel cycle.

The above mentioned integral measurements capability is important but of equal significance is the ability to measure differential cross sections of higher actinides and specific fission products. Such data are required for criticality analysis of fuel manufacturing, fuel transport, fuel storage, fuel reprocessing and recycling and waste management.

Blair Briggs (Idaho National Laboratory)

In general it appears that other countries are putting many more resources into improving their nuclear data than we are in the United States. Maintenance and improvement of the U.S. ENDF nuclear data libraries is largely a volunteer effort in the U.S. and has been for decades while other countries have been providing resources for their work.

I offer lead as an example. Lead is widely used in transportation and storage systems for spent nuclear fuels. We have known about deficiencies in the lead cross section for decades. Recently the ICSBEP identified a series of integral experiments that were performed at LLNL that clearly demonstrated an obvious bias when using lead as a reflector. The bias increased as the thickness of lead increased. ICSBEP participants from VNIITF in Russia subsequently offered an independent series of similar experiments that more clearly demonstrated the same bias using U.S. cross section data and MUCH less of a bias using Russian cross section data. The JEFF cross section group immediately dumped resources into re-evaluating the lead cross section and produced a much improved evaluation, even before the Russian benchmarks became available. (I don't know if they included new differential measurements in this case, but they may have.) With a funded staff, they had results for the Russian Benchmarks one weekend (3 days) after I sent them the benchmark. ENDF/B-VII is about to be released. It is my understanding that we will be using the JEFF cross-section data for lead.

With regards to nuclear data, the U.S. is quickly becoming a third world country. That may be an extreme position. Dick McKnight can give you a much more accurate assessment since he works closely with the international community, but my observation is that we are falling behind. My comment is not intended to be a criticism of the efforts of those at BNL, LANL, ANL, ORNL and others who have and continue to volunteer their time to keep us where we are. They have done a remarkable job, but they have not had sufficient resources to do their work since the peak of the reactor development period. Many of the evaluators are aging. The few younger evaluators will likely tire of volunteer work and be attracted to more enticing project that have funding. Begging for funding eventually wears good researchers down. I am not sure that the younger generation will be willing to continue the fight for funding in this manner. Not including resources for Nuclear Data activities on the grounds that we can just be extra conservative is wrong. We need to put resources into both integral and differential measurements.

Lead is an obvious example of a widely used material that we should know much more about. Let me say a few words about plutonium and the higher actinides. The criticality safety community has been struggling with the need for integral benchmark data for damp MOX powders for nearly a decade. The need for integral data for damp plutonium powders has been known for even longer. Existing data has obvious problems. The U.S. lost their capability to do these types integral measurements a long time ago. The OECD NEA sponsored a workshop in the spring of 2004. The experts reached a clear consensus on the need for additional experimental data for these types of systems. The position taken by industry was rather static, thinking only about existing designs and facilities that rely solely on excessive conservatism to compensate for ignorance with little or no concern about future needs. To meet the demands of future United States energy needs, we need dynamic thinking. The static mentality has got us into the situation we are currently in, a super power that either dances to the turn of small oil producing nations or bullies them into giving us what we want.

U.S. integral data for plutonium solutions is generally quite poor. French integral data appear to be much better. The U.S. has lost the capability to perform plutonium solution experiments. Russia has, in large part, relied on U.S. data until the French data became widely available. France is gradually giving up their capabilities and Japan has made a political decision not to allow plutonium into their STACY and TRACY facilities even though that was their original intent and design.

The fact that higher actinide cross sections are, in general, very poor is well documented. As we go to higher burnup, these cross sections will become more important, especially if we start reprocessing the fuel.

With regard to differential data measurement needs, I would refer you to the table in the NCSP Five Year Plan. The NCSP need for Improved Pu-240 cross section data is known. Improved Pu-241 and Pu-242 cross section data are NE needs. We do not know everything there is to know about plutonium. Jerry Cole's preliminary measurements at IPNS on Pu-239 demonstrate that we do not even know everything we think we know about Pu-239. The alpha resonance that he has discovered will likely increase the capture and total Pu-239 cross section over the range of measurement. It will be interesting to see how this discovery will change the performance of our cross section data on certain benchmarks. Even if the effect of this discovery is small for Pu-239, it might not be as small for Pu-240. If we plan to do much with plutonium in the United States in the future, we need Jerry Cole's measurement technique and we need IPNS.

I conclude with a question: By the show of hands, has anyone out there ever built a reactor? Keep your hand(s) up. On the same hand show the number of fingers before you plan to retire. Last year it was GEN-IV, this year it is . . . . If we never really build anything, maybe we won't need more data. Lights out!