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# Signatures of Aging Revisited

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# 1 INTRODUCTION

This study is a follow-on to the review made by JASON during its 1997 Summer Study of what is known about the aging of critical constituents, particularly the high explosives, metals (Pu, U), and polymers in the enduring stockpile. The JASON report (JSR-97-320) that summarized the findings was based on briefings by the three weapons labs (LANL, LLNL, SNL). They presented excellent technical analyses covering a broad range of scientific and engineering problems pertaining to determining signatures of aging. But the report also noted: "Missing, however, from the briefings and the written documents made available to us by the labs and DOE, was evidence of an adequately sharp focus and high priorities on a number of essential near-term needs of maintaining weapons in the stockpile."

JSR-97-320 listed a set of recommendations as to the highest priority program needs to "identify age-induced problems in the stockpile, and prepare means to fix them in a timely fashion." The report concluded as follows:

It is necessary to implement these priorities in a timely fashion; they address the short-term (5-10 years) needs of the SSMP. To do so will require strong, effective leadership — within the three weapons labs and from DOE Washington — to make program choices and to assign appropriate resources among and within the three weapons labs. The individual programs briefed to us by the three labs did not show a balance, a focus, and a coordination consistent with these requirements. The panel has been advised by the labs and by the DOE program office in Washington that extensive efforts (e.g. the laboratories' stockpile life extension and enhanced surveillance programs) have been made to achieve the necessary leadership, focus, and balance that are

required for an efficient national program that appropriately addresses the priorities and coordination we are recommending. Our view is that such efforts are important and should be aggressively pursued. It is of particular importance, as will be clear from our later discussion, to coordinate and balance the diverse activities of stewardship, (enhanced) surveillance, and refurbishment. A future study may want to delve more deeply into this broader issue. We note that such a structure clearly exists for the large new facilities that will be important components of the stewardship program over the longer term.

The JASON revisit to the aging problem in January 1998 focused on the work of the Washington Office (DP-20 under the leadership of Deputy Assistant Secretary of Energy for Defense Programs, responsible for Stockpile Management) to provide a broader context for understanding aging issues and addressing the needs they present for stockpile maintenance. We also reviewed the work of the Laboratories on the Stockpile Life Extension Program (SLEP), which is identified as DOE's planning framework for proactive management of system maintenance activities, and the scope of the Enhanced Surveillance Program (ESP) that supports SLEP and plays a critical role in the overall Stockpile Stewardship and Management Program (SSMP).

The major findings of this limited review can be summarized as follows:

1. There is in place at DOE Headquarters an effective leadership for setting program priorities and integrating the activities between the individual labs, and of the SLEP/ESP into the overall SSMP. In particular we were pleased to see evidence of strong inter-lab cooperation.
2. Concerns raised in JSR-97-320 are being addressed in programs that appear to have been strengthened or initiated during the past six months. We welcome these important developments and are encouraged by what

has been learned that has added confidence to our understanding of aging. Specific technical findings are discussed in the next section. In particular some prior concerns about aging of high explosives have been eased and important progress is being made in study of the time scale of Pu aging.

3. An important new steering committee has been created by DOE with balanced participation by lab scientists with officials at headquarters in setting program priorities. This management structure will be valuable for this program in ensuring proper feedback from elements of the complex and appropriate inter lab coordination. We are pleased to note the evidence that it is already proving its effectiveness.

The program is still young and difficult issues remain to be fully addressed. Two examples are integration of remanufacturing requirements into the SSMP and the coordination of the sub-critical experiments at the Nevada Test Site with the lab work, but overall we are very pleased by what we learned and by progress since our review six months ago.

Maintaining expertise within the nuclear weapons laboratories is a major concern over the long term, so we fully approve of the new efforts now being made within the National Laboratories to identify and train young scientists to become leaders in the relevant technical areas. By strengthening relations with universities, it will be possible to ensure that an ongoing supply of technically talented scientists is maintained within the Laboratories. Also, the long-term reliability of the Stockpile will best be ensured by challenging young researchers to take positions of leadership within the weapons programs, as is now being done in the Laboratories.

## 2 TECHNICAL PROGRAMS

In June/July of 1997 we saw many presentations involving different programs and capabilities at the three labs, all of which had been identified as having potential impact on the surveillance program. In January of 1998, we saw significant progress in bringing these individual programs together to form well-defined cooperative endeavors. Progress includes both coordinating efforts within the Enhanced Surveillance Program, and developing plans for transfer of knowledge and tools to the Core Surveillance Program.

### 2.1 Primaries

Studies of HE related to aging are strong and have shown rapid development in focus. Continuing effort in coordinating work at the three laboratories, should be emphasized.

One area which seems to be in the preliminary stages of development is bringing the understanding of materials aging appropriately into the hydrodynamic codes. Serious thought needs to be given as to what level of materials information is needed for this purpose. It seems likely that a small number of appropriately chosen macroscopic characteristics (e.g. density) will be most useful for hydrodynamic calculations. Prioritizing the experiments and theoretical calculations to be performed based on what is most immediately useful, as well as using a staged approach in which new work systematically builds on prior results, should be emphasized.

One of the most important components of the ESP will be to document the micro-physical effects of aging in Pu in order to understand how the weapon physics package evolves with time. We are pleased to learn that a

program of accelerated aging using  $Pu^{239}$  spiked with 5%  $Pu^{238}$  is under way, including the fact that care will be taken to make sure that samples of spiked and normal plutonium start out at time zero from the same baseline.

We were impressed by the efforts described to characterize the atomic-scale structure (by PAS and synchrotron-based XAFS), microstructure (e.g., by transmission electron microscopy) and the bulk physical properties of both aged and new Pu alloys of different compositions across the range of interest. These analytical methods will provide key information for documenting the effects of aging, not only in actual pits but also in samples designed to have accelerated aging (indeed, in ensuring that the accelerated-aging samples are truly reproducing changes taking place in pits that have been aged without acceleration). The latter will give an important predictive capability that will alert researchers within just a few years of effects that would otherwise take place in decades. This information is also essential for theoreticians, both to help calibrate atomistic models of aging phenomena and to assess the reliability of these models. Moreover, the methods being developed and used to characterize Pu alloys will no doubt prove useful in monitoring the remanufacture of pits over the coming years.

New experiments on the equation of state and the thermodynamic properties of Pu at elevated pressures and temperatures are addressing significant questions that require resolution, and doing so in a program that is both balanced and well coordinated. We approve of the mix of static and dynamic experiments that were described (both underground and above-ground in the latter case), and were impressed by the level of collaboration among groups having complementary strengths in the different National Laboratories.

A crucial question attending such accelerated aging studies remains unanswered: How can we be sure that the effects of accelerated aging are the same as the effects of natural aging? To answer this question, we need to observe and understand in detail the various processes of annealing and mi-



gration that will not be accelerated in the spiked samples. The combination of accelerated damage with unaccelerated annealing and migration might cause either more or less severe effects than unaccelerated damage with unaccelerated annealing and migration. For the purpose of disentangling the effects of damage and annealing, it would be helpful to study the behavior of samples of plutonium with various fractions of  $Pu^{238}$ . Besides zero and 5%, other fractions such as 1%, 2%, 10%, 20%, ... should be studied, so far as environmental and safety regulations permit.

It might appear that these and other accelerated-aging experiments on plutonium could only be done by melting the two  $Pu$  samples in order to mix them, thus in effect resetting the  $Pu^{239}$  to zero age. But this is not necessarily the case. One can bond by physical pressure very flat plates of  $Pu^{238}$  and  $Pu^{239}$  at various temperatures well below melting and study, at different accelerated ages, various defect creation and transport properties in transverse sections of the bonded plates. In this way one can take old  $Pu$  samples and further age them, or study diffusion lengths and other transport properties most easily studied in inhomogeneous systems in plutonium of any age. One might also find that studies of inhomogeneous systems allow for more stringent tests of the modeling of defect properties than can be done simply with homogeneous-sample data: for example, comparing the properties of new  $Pu^{239}$  mixed homogeneously with enough  $Pu^{238}$  to produce an effective age of 100 years with those of 40-year-old  $Pu^{239}$  diffusively bonded to  $Pu^{238}$  and seasoned for a further 60 years of effective age.

## 2.2 Secondaries

Studies leading to a predictive understanding of canned sub-assembly (CSA), or secondaries, aging are driven by the technical difficulty of package disassembly for direct inspection. The required strong correlation of fundamental studies, development and application of advanced diagnostic tools, and coordination with core surveillance activities, is being developed along constructive lines. Continuing emphasis on improving collaboration between diagnostics development with materials characterization efforts is important in this work. Emphasis on two types of diagnostics is required: 1) In-situ diagnostics for characterizing the CSA environment are likely to have a strong pay-off in prolonging life; and 2) Non-destructive ex-situ diagnostics are important for minimizing the cost of identifying units that have undergone significant degradation.

### 3 AGING SCIENCE AND REMANUFACTURING

We emphasize a point made in our last report, which also primarily reviewed and commented on questions of aging rather than on questions of remanufacture. But we did comment there on the need for an early connection between aging studies and remanufacture. We still remain concerned that there is insufficient coupling now between stockpile aging and surveillance issues and issues of remanufacturing and possibly redesigning component parts. A program of selective preventive remanufacture—replacing some components before one knows the full science story about their aging and failure rates—can be cost-effective, if it is well-coordinated with the science studies on aging. One need not wait until aging studies on a specific component are finished before addressing questions of its remanufacture and replacement; preemptive replacement may be a superior approach, provided that there is good evidence that the replacement part really works. We suggest that the ESP Steering Committee create and maintain closer contacts with counterparts in such remanufacturing initiatives as ADaPT.

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