Signatures of Aging

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13. ABSTRACT *(Maximum 200 words)*

The Department of Energy and its three weapons laboratories (LANL, LLNL, and SNL) have developed aStockpile Stewardship and Management Program (SSMP) in response to their designated mission of maintaining an effective, i.e. reliable and safe, nuclear deterrent without underground nuclear tests (UGTs). The need to ensure the effectiveness of an aging stockpile presents new challenges of major importance.

In this study we review what is known about the aging of critical constituents, particularly the high explosives, polymers, and metals in the enduring stockpile. We discuss data that are required to provide a fuller understanding of aging, and how to obtain that data as a basis for anticipating and addressing potential stockpile problems. Our particular concern is problems that may arise in the short term, i.e. within the next 5-10 years, and their implied requirements for preventive maintenance and remanufacture.

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Contents

1 SUMMARY AND RECOMMENDATIONS

In accord with present policy, the U.S. must have the ability to detect potential problems in the enduring nuclear stockpile with sufficient lead-time to fix them and maintain confidence in the stockpile. It is the aim of the Stockpile Stewardship and Management Program (SSMP) to meet this goal by:

1. Developing and implementing a surveillance program that can detect the effects of aging and identify ways to fix them.
2. Identifying the changes in an aging stockpile that could lead to degradation of performance, reliability, or safety.
3. Developing a science-based understanding of the materials and their interactions in warheads.
4. Bringing on line the required evaluation facilities and remanufacturing capabilities so that the needed repairs, refurbishing, or remanufacturing can be accomplished before the weapons degrade.
5. Determining the long term need for advanced test facilities, and developing facility specifications and construction schedules based upon the best scientific understanding of aging problems and findings of the SSMP surveillance program.

An effective SSMP must both identify age-induced problems in the stockpile, and prepare means to fix them in a timely fashion. In a number of cases the remanufacturing processes will differ from those used in the original manufacture of the warhead, as a result of new environmental and safety regulations or because the original materials and industrial sources are unavailable. There may also be changes encouraged by a need to reduce cost. However, changes in the manufacturing processes cannot be allowed to reduce confidence in the stockpile.

Not all of the work needed to maintain confidence in the stockpile performance and safety over the coming decades qualifies as forefront science or technology. But it is critical for responsible, annual certification of each type of warhead in the enduring stockpile and must not be short-changed in assigning resources relative to other parts of the program that are major initiatives, such as the Accelerated Scientific Computing Initiative (ASCI), the National Ignition Facility (NIF), or an Advanced Hydrodynamic Facility (AHF). Similarly, the pursuit of fundamental science at the weapons labs must serve, and not dominate, the essential tasks of assessment, surveillance, and refurbishment.

Recommendations:

1. Highest priority attention must be given to those changes due to aging that affect the performance of the primaries. If the yield of the primary is inadequate, the secondary (main stage) will not ignite and the weapon will fail. There should be appropriate emphasis placed on sampling the oldest weapons in the stockpile for diagnostic purposes in order to identify changes due to aging.

   (a) There is no hard evidence of aging effects in high explosives that will affect performance of primaries in our current nuclear weapons and that also require immediate or near-term action. However, the existing data on aging high explosives (HE) is inadequate to set a time-line for the possible emergence of such problems and to prepare for the replacement of HE on the needed scale. In particular there is need to establish the effect of binder aging in the 9501 formulation of HE. Binder aging might change the HE response to high mechanical stresses that are encountered in the stockpile-to-target sequence of specific systems.

(b) There is urgent need to do experiments on aged, weapons-grade Pu^{239} in order to measure void swelling and surface damage arising from the recoiling U and He products of Pu radioactive decay. The only way to make such measurements within a reasonable time and with high fidelity is to accelerate the aging of Pu^{239} by spiking it with Pu^{238} and taking care not to alter the thermal and phase properties relevant to weapons conditions. The rate and incubation period of damaging changes need to be determined to set the capacity of a Pu refurbishment and pit remaufacturing facility.

(c) Appropriate sub-critical experiments will provide data on possible differences between old and young Pu in surface properties, ejecta, and phase diagrams. Such differences could affect the behavior of primaries.

2. Components external to the nuclear package should continue to be thoroughly tested and improved when necessary in order to retain confidence and reduce costs during weapon remanufacture. Under a CTBT such activities can continue.

3. Although current data does not suggest that aging will lead to diminished performance of secondaries in the near future, continued careful surveillance of secondaries must be maintained in order to detect, and thereby avoid, uranium hydriding or structural degradation of components due to corrosion or other changes.

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.

2 INTRODUCTION

The Department of Energy and its three weapons laboratories (LANL, LLNL, and SNL) have developed a Stockpile Stewardship and Management Program (SSMP) in response to their designated mission of maintaining an effective, i.e. reliable and safe, nuclear deterrent without underground nuclear tests (UGTs). The need to ensure the effectiveness of an aging stockpile presents new challenges of major importance.

In this study we review what is known about the aging of critical constituents, particularly the high explosives, polymers, and metals in the enduring stockpile. We discuss data that are required to provide a fuller understanding of aging, and how to obtain that data as a basis for anticipating and addressing potential stockpile problems. Our particular concern is problems that may arise in the short term, i.e. within the next 5-10 years, and their implied requirements for preventive maintenance and remanufacture.

All three weapons labs (LANL, LLNL, SNL) presented excellent technical analyses that addressed a broad range of scientific and engineering problems pertaining to determining signatures of aging. 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.

Here are guidelines for a plan to fill this void:

1. Develop an understanding of the opportunities, costs, and needs of remanufacturing with available technology. An adequate understanding of the sensitivity of warhead performance and safety to the manufacturing process is necessary to provide a context for the Enhanced Surveil-

lance Program (ESP), the Stockpile Life Extension Program (SLEP), and to provide a baseline by which the success of the SSMP can be evaluated.

2. Develop estimates of how much it would cost to remanufacture the appropriate weapons components to ensure a reliable stockpile, at START-controlled levels, over 30 year time periods. Historical experience gives confidence that individual weapons in the enduring stockpile have at least a 20 year life-span (See Appendix A). With confidence based on design margins, including planned improvements in progress, the life-span can probably be extended to 30 years or longer. A better understanding of aging, and of the costs of R&D on aging and remanufacture, will provide one of the important components needed for comparing with the savings that might result from less frequent remanufacturing of the stockpile.

3. Develop and assign high priority to a program to determine the important aging phenomena, including an accelerated aging program where possible with fidelity to physical conditions appropriate to warhead environments, to enable the factors associated with aging to be predicted with some confidence. Diagnostics of aging need not be atomistic in character and may be phenomenological, as long as they are validated by experiments in the laboratory and by measurements on the stockpile. This part of the program should focus on accelerated aging tests for components that are highest in priority when the risk of failure and the cost of extending lifetimes are appropriately weighted. It should also include studies of the value for lifetime extension of maintaining a controlled thermal environment during storage of the active reserve force and, as practical, into the stockpile-to-target sequence of the weapons. The accelerated aging program should yield concrete results within 5 years for the components and systems it is designed to address. It

should not rely on long-term science projects in order to be successful. Close cooperation between theory and experiment is crucial.

4. For the longer term, the stockpile stewardship program should work toward a more complete understanding of all components of the physics package. This will require adequate diagnostics for above-ground and underground experiments (i.e. sub-critical measurements) consistent with the comprehensive test ban treaty. The purpose is to understand the sensitivity of stockpile weapons to changes resulting from aging or from variations in the remanufacturing processes. The focus of effort should be on areas with the highest payoff relative to cost and risk. The existing program for testing and replacing components outside the physics package is not restricted by a CTBT and should continue to receive priority attention and support commensurate with the benefits.

5. Computer models of materials will be important for longtime stockpile stewardship. With such models, together with non-nuclear experiments, one can assess the effects on performance and safety of observed changes in materials. One can also use measurements on a small scale to predict behavior on the large scales relevant to materials performance in weapons. In the long term, such modeling may become accurate enough to replace accelerated aging experiments that can never precisely reproduce the effects of natural aging. The deeper understanding gained from modeling should allow the optimization of maintenance schedules to match subsystem and component lifetimes.

As a final comment, we express a serious concern about the SSMP and the studies of aging of the physics package briefed to the JASON study group. The laboratories and the scientists who are doing this work are starting from a difficult position. We do not wish to point a finger at its causes or to place blame. Archiving of data, characterization of materials, and preservation of samples lacked proper attention during approximately 150 tests of modern weapon designs over the past two decades. During these years new design and diagnostic programs commanded priority attention.

As a result, important knowledge of our stockpile, with basic understanding based on data, is now lacking. It is imperative that the SSMP focus necessary resources on the routine analytical and stockpile maintenance program. However desirable such a focussed program would have been in the past, it is now crucial for maintaining confidence in our aging stockpile in an era without UGTs. Such a program requires strong laboratory management and inter-lab coordination to set priorities and focus on essential tasks. It also requires strong DOE program leadership in Washington.

3 ORGANIZATION OF REPORT

In the following we consider what is known, what needs to be known, and how important data can be gained about potential aging problems that could affect performance of the

1. Energetic Materials
2. Organics and Plastics
3. Pits
4. Secondaries
5. Detonators, Initiators, and Electronics

Throughout, we provide examples of priorities that must be set, distinguishing the essential from the less important activities. We also consider what should be done to collect valuable information from the records of past activities in the weapons program that are relevant to stockpile management and stewardship. We conclude with general observations on the relation between investigations into aging science and stockpile remanufacturing.

4 MATERIALS SCIENCE ISSUES IN STOCKPILE MAINTENANCE

The key policy problem in all of the studies of materials properties is defining how the information gained from these studies will be used in making decisions concerning stockpile maintenance.

Major goals in the stockpile surveillance program as related to materials and components are to:

(a) Establish diagnostics for aging in which readily measurable physical quantities can be uniquely related to the required functional performance of the materials.

(b) Establish diagnostics for aging in which readily measurable physical quantities can be uniquely related to the safety of the materials.

(c) Establish the stage of materials aging at which decomposition products endanger the performance of other components of the weapon.

(d) Establish criteria and procedures for the replacement of the materials and components.

In addition, confidence in the reliability of the stockpile can be enhanced by increasing performance margins for many of the primaries by means of improved boost gas delivery systems (see JSR-95-320). Adequate performance margins are needed to guard against unknown issues and problems yet to be identified. On the other hand, potential benefits from enhanced surveillance, advanced diagnostics, and improved processes for remanufacturing must be quantified in order to evaluate which are worth pursuing. One measure of the significance of any such efforts is through comparison with the potential and cost of directly enhancing the performance margins as discussed in JSR-95-320.

4.1 Energetic Materials

The stability and performance of energetic materials are essential to the safety and reliability of the weapons in the stockpile.

In principle, a fair amount of information on the aging of the HE materials is already available, because the historical surveillance program includes a number of standard characterization techniques which have been used (and which should continue to be used to provide continuity) in order to monitor the state of the HE components of the physics package. These include measurement of density, binder molecular weight, and tensile strength, as well as a number of other diagnostics which have been applied to at least some subset of the HE materials, or some subset of the weapons systems.

To be most useful, this information should be correlated with as much information as can be found (for the same unit) concerning the manufacturing and handling history, and, most importantly, performance studies, especially studies of initiability and detonation-front propagation. Unfortunately, as discussed in Section 5 of this study, it is unclear how much historical information of this sort exists, and retrieving and interpreting the old records will require substantial effort. As it becomes available, such information will prove invaluable in documenting the range of variability in physical characteristics that in the past was proven to be consistent with functioning weapons.

At the most conservative level, the criteria for maintenance of the stockpile energetic materials could be set by requiring physical characteristics to fall within the ranges that were previously observed for materials giving satisfactory performance. It would be more valuable to combine modelling and experiment to determine the most likely effects of aging that could be detrimental to safety or reliability of performance. Such studies would ideally serve to anticipate signs of degradation, which could then be monitored but should at the very minimum lead to explicit characteristics (ranges in values of measurable properties) considered acceptable for the HE. A clear program for determining how decision criteria will be defined is essential, and must be completed in a timely manner.

4.1.1 Aging Studies

There is a large number of enhanced surveillance activities focused on performance characteristics (including initiability, propagation, shock safety), as well as physical characteristics, such as stress-strain behavior, shape changes, chemical composition, etc. These studies serve the immediate purpose of defining the present-day baseline of the energetic materials in the stockpile, and also serve as the basis for studies of aging. Experimental simulated aging studies appear to be only in their early stages, as existing stockpile-to-target sequence (STS) tests are not by themselves adequate for quantitative prediction of stockpile aging.

A constructive approach being undertaken at Los Alamos is to formulate special batches of PBX 9501 (using both strategic reserve and commercial estane binder), and to vary the properties systematically to mimic known effects of aging such as decreases in binder molecular weight and depletion of nitroplasticizer. These samples should then be evaluated with the same tests used in the enhanced surveillance studies. The results of these studies should make it possible to determine directly the quantitative levels of physical/chemical changes that result in unacceptable changes in performance characteristics. At Livermore, simulated aging will be studied in special environmental cells (PBX Environmental Test Units) in which the energetic materials can be subjected to variable conditions. The utility of such tests will be determined by how well physical characteristics and performance diagnostics on the "aged" materials are correlated, and by how well the accelerated aging process can be correlated with real time.

Neither of the above two points was discussed directly in the briefing. However, it was indicated that dynamic measurements of elastic moduli as a function of temperature are underway, and these, as well as long-time measurements at different temperatures, should be used to characterize the time-temperature correlations for the study. It is clearly important to develop and maintain strong coordination between the aging studies which are planned for the same materials at the weapon design laboratories.

Although the materials under study, and the nature of the performance requirements are somewhat different at Sandia, the similarities in the required studies are strong, and coordination of aging studies at Sandia with those at the other laboratories is clearly needed. The specific aging studies now beginning at Sandia involve tests of XTX8003 samples aged at three different temperatures for two years. A variety of physical and chemical characterization techniques will be correlated with a performance measurement of the resulting sample detonation velocity. Systematic comparison of the aging characteristics at three temperatures will in principle serve to characterize the time-temperature correlation and allow extrapolation to longer times.

4.1.2 Aging in the Radiation Environment

One issue of aging which clearly needs to be addressed for all energetic materials in the stockpile is the effect of the radiation environment. Livermore scientists have completed one study of the effect of radiation on the molecular weight distribution of Kel-F 800. The results showed that significant changes occur only at radiation doses much higher than those expected in the weapon environment. Similar studies should be given a high priority for all the components of energetic materials in the stockpile, as this is an aspect of aging that cannot be independently judged from thermally-accelerated aging tests. For materials in which molecular weight distribution is not an appropriate property, the physical characteristics most likely to be relevant to weapons performance are the mechanical properties (e.g. stress-strain). Thus determining the radiation dose effects on the elastic and other mechanical properties should serve as a useful measure of radiation-induced materials aging.

4.1.3 Fundamental Studies

The discussion in the preceding paragraphs has focused on the development of diagnostics that can be correlated with weapon performance, and on experimental accelerated aging studies that can also be directly correlated with weapon performance. These studies are of the highest priority for the stockpile program as they can be used to set immediate criteria and diagnostics to certify reliable performance of the energetic materials in the stockpile.

Because such focused studies may fail to detect unexpected aspects of aging, fundamental experimental studies correlated with modeling of the energetic materials in the stockpile also must be pursued. The extent to which the results of such fundamental studies can be correlated to performance characteristics such as initiability can be used to set priorities within the more focussed research.

Study of HMX

A long-term fundamental study of the properties of HMX (a major component of both the PBX and LX materials) has been carried out at Sandia over a period of many years, and can serve as a model of the potential, the limitations, and the costs of fundamental studies in predicting stockpile performance. Research on HMX dates back at least 50 years, and the present concentrated thrust at Sandia has been underway for approximately 10 years. The fundamental studies include a concentrated effort to apply methods of chemical analysis to understand the molecular mechanisms and rates of decomposition of HMX. A special approach was used in which the mass of the sample was monitored simultaneously with evolution of gaseous products during reaction. Correlated studies of the evolution of the structure of the solid particles of HMX during reaction were performed with transmission electron microscopy (TEM). The results of this study show a complex interplay between the reaction chemistry of the HMX and the evolution of physical structure of the solid phase.

Continuing work on HMX to relate known age-dependent changes in properties of the material to changes in the underlying chemical/physical processes of decomposition is likely to be extremely helpful in understanding the effects of aging. Coordination of void measurement with Livermore positron annihilation spectroscopy (PALS) experiments would be useful as well. Efforts to develop mathematical models to describe HMX decomposition were mentioned, but the procedures to be used in developing such models were not discussed. Given the complexity of the system, developing effective descriptive, much less predictive, models represents a serious challenge to state-of-the-art capabilities in theoretical materials chemistry. If this modeling aspect of the program is undertaken seriously, its main impact is likely to be an improved generic understanding of the materials processes, which may reduce costs by guiding improvements in testing, rather than in any short-term impact on the surveillance program.

Efforts in using shock physics to model the performance of granular HE materials including HMX are also underway both at Sandia and at Livermore. These modeling studies provide methods for simulating initiation behavior for simplified models of the granular particles of energetic materials, and need to be better coordinated. At the present state of modeling, highly graphical and intuitive information can be provided about the effects of generic variables such as particle size and packing, density, and geometry of sample. This type of information is useful as feedback to what key variables need to be emphasized in experimental studies. Including more detailed input of specific material properties, such as strength, particle interactions, dissipation and chemical reactions is an area of current research. Appropriately incorporating reliable information about specific materials may be extremely difficult, given the complexities of real materials. However, doing so is essential to providing feedback to experimentalists and in improving predictibility in materials performance.

Progress in this direction can be accomplished only via a close interaction between experimentalists performing studies of the fundamental properties, and the theorists carrying out the modeling. There is also the need for considerable caution in developing procedures for incorporating the appropriate fundamental properties into such simulations. Normal procedure in developing the phenomenological models is to tune the input parameters to reproduce some known property of the system under study. The quality of the theoretical predictions made with the given parameters can be highly variable, in unpredictable ways. Thus, validation of the simulations by careful comparison with experiments distinct from those originally used to define the input parameters is an essential part of any simulation program.

Another point of considerable concern is that at the present level of capabilities, the appropriate role of computation and modeling must be as an aid in interpretation and guide for future measurements. In spite of the appealing visual presentations of first-principles, atomistic simulations that can be prepared with high-powered computational capabilities, we are far from being able to make quantitative predictions of material properties at the level needed to certify stockpile performance. Both experiments and simulations/theory will be needed to make valid judgments about certification of the stockpile.

Our description of the HMX studies is intended to provide detailed examples of how current experimental and theoretical programs are being pursued, and how they can be improved. To summarize, theory is essential to the degree that it provides timely and reliable results, so as to be useful for aging-related experiments and for surveillance. Unless linked to high-quality experimental measurements, theory cannot play a strong role.

4.1.4 Integral Experiments

Integral above ground experiments (AGEX) can study the implosion hydrodynamics prior to criticality if the Pu in the primary is replaced by a suitable surrogate non-fissioning material. Such core punching (radiography) experiments can provide important data on aging effects in the HE by comparison of data from aged samples against young ones. The importance of core punching experiments at PHERMEX and FXR is well established for documenting important parameters of the implosion, such as energy and symmetry. Recent detector and accelerator upgrades will add to knowledge of the energy and symmetry in the implosion process by providing higher resolution and several successive images. DARHT will add the important capability of imaging from two different angles at two different times, as well as providing a deeper technical understanding of the potential need for more advanced facilities allowing multiple images during the implosion.

4.2 Organics and Plastics (other than energetic materials)

A large number of polymeric components in the various weapons systems serve roles in structural support, electrical isolation, vibrational isolation, etc. It is unclear what, if any, archival information exists as to the criteria used in the selection of these materials. However, in the absence of specific information to the contrary, and given that the materials list seems to involve very standard formulations, it seems reasonable to assume that the materials were chosen, or engineered for specific weapons needs, on the basis of standard engineering requirements and routine availability

Aging of the polymeric materials is a complex and long-studied engineering problem, which may be intensified in nuclear weapons by the special conditions of the radiation environment or by the impact of decomposition products from aging energetic materials and other organic materials and salts. Routine inspection of the components, as under the existing surveillance program, is likely to be sufficient to indicate when replacements are needed, and all three weapons laboratories have priorities in place as to the evaluation of these components. Diagnostics of the gaseous environment in the various compartments of the weapon which are being developed to monitor other materials systems will have the secondary benefit of indicating changes in environment that may accelerate degradation of the polymeric materials.

Therefore, it does not seem warranted to undertake extensive scientific studies of these materials. Instead it seems more appropriate to limit the investigations of the aging of these materials to standard engineering methods, augmented as appropriate with controlled exposure to radiation, or degradation products of other materials. The standard engineering procedure for characterizing accelerated aging of polymer materials is the time-temperature (and the corollary frequency-temperature) test of elastic response. In this approach, measurements of an elastic property, typically the stress relaxation modulus, are made over a limited time range with the sample held at different temperatures. The time considered is then extended from the finite measured time to longer times by scaling the time axis for each temperature by a temperature-dependent coefficient. Similarly, measurements of the frequency dependence of an elastic parameter, typically the storage compliance, can be made over a limited frequency range at different temperatures, and then rescaled to a wide frequency range. An experimental verification of frequency scaling is thus a useful baseline (e.g. necessary, but not always sufficient requirement) for the applicability of time-temperature scaling.

4.3 Pits

4.3.1 Plutonium aging

The central task of stockpile stewardship is to make sure that aging of pits does not significantly degrade the performance of weapons. Sooner or later, pits inevitably deteriorate because of the alpha-decay of plutonium. Symptoms of deterioration are microscopic disordering of the metal, migration of helium and gallium to and from grain boundaries, recrystallization of grains, local conversion of plutonium from delta-phase to alpha-phase, formation of helium bubbles, and macroscopic swelling or shrinking and creep of the entire pit. These symptoms could change the equation of state, the elastic moduli and the conditions of melting for the bulk metal. As far as pits are concerned, the program of stockpile stewardship must have three parts to be pursued concurrently. Part 1: Study of the symptoms of deterioration by static observation of samples of metal of various ages. Part 2: Study of the effects on weapon performance by dynamic sub-critical experiments using samples of metal of various ages. Part 3: Construction of a remanufacturing facility to make new pits out of old ones as needed.

Up to now, the weapons laboratories have made little progress in implementing the three parts of this program. Activities related to Parts 2 and 3 of the program have only recently begun. We heard a number of briefings related to Part 1, from Los Alamos, Livermore and Sandia. The briefings mostly described experimental facilities, some already operating and some planned, that are to be used for enhanced surveillance of pits and other weapons components. The surveillance program has been mainly directed toward finding defects caused by manufacturing errors and chemical corrosion. Finding such defects is necessary and important, but the work done up until now has not produced much hard data concerning effects of aging of plutonium. The enhanced surveillance program is being designed to develop a capability to find defects caused by aging and to estimate the lifetimes of weapon components.

The little data that exists is summarized in Livermore report UCRL-LR 118812 (S) and Sandia report SAND95-8712. The Livermore report contains mostly qualitative evidence indicating that pits free from manufacturing defects can be stored for twenty years without significant deterioration. The Sandia report estimates the deterioration of plutonium due to alpha-decay by comparing it with observations of deterioration in metals exposed to neutron bombardment or ion implantation. The observations are quantitative, and show that swelling and creep occur when the integrated radiation dose exceeds a certain threshold. Unfortunately the threshold dose causing large-scale damage varies widely from metal to metal, and none of the observations were made on plutonium. The extent of damage depends on the effectiveness of processes of annealing which repair damage and restore stability of the metal. The annealing processes depend sensitively on the microscopic structure of the material. Since plutonium is an unusual metal with unique properties, the extrapolation of the observational results to plutonium is highly uncertain. The Sandia report concludes that the age at which plutonium will show serious effects of aging may lie anywhere between twenty and a hundred years. Theoretical estimates based on observations of other metals are not an effective substitute for direct measurements using aged plutonium.

We have been told that the main reason why observations of effects of aging in plutonium have not been made is that samples of old pits are not available. The oldest pits in the enduring stockpile are only thirty years old. The processes of deterioration are highly non-linear, and significant effects on weapon performance might appear rather suddenly at some point in the aging process. Little can be learned from observation of samples younger than twenty years. A possible supply of forty-year-old plutonium has recently been located in the UK [see Los Alamos report NMT-5:97-234(S)]. A substantial quantity of this plutonium may become available for study in the US. We recommend that the program of experiments described in the Los Alamos report be carried out as soon as possible. These experiments will substantially increase our general understanding of the aging process in plutonium, although differences in the chemistry and metallurgical history of the plutonium in UK samples and in US pits must be taken into account in interpreting the results and applying them to the SSMP.

Fortunately, there exists a method for obtaining samples of plutonium in which the process of aging is greatly accelerated. This can be done by mixing ordinary weapons-grade plutonium with the separated isotope plutonium 238. The alpha-decay of Pu 238 produces the same types of material deterioration as the decay of Pu 239, but the decay of Pu 238 is 280 times faster. If one uses weapons-grade plutonium mixed with f percent of Pu

238 then the effects of aging will be accelerated by a factor (1+2.8f). Using samples with f=10 percent Pu 238, one can study in four years the effects of aging of ordinary plutonium up to 116 years. We recommend that such studies begin immediately, using samples with various fractions of Pu 238 up to 10 percent to provide a solid basis for Parts 1 and 2 of the stewardship program for pits.

According to Livermore report UCRL-LR-118812 (S), page 13, some studies of aging using samples of plutonium enriched in Pu 238 were carried out in the 1960's. The simulated age of the samples did not exceed twenty years and no significant effects of aging were seen.

A study comparing aging effects in pure Pu 238 and weapons-grade plutonium is described in an unclassified Russian paper ["Influence of Intensive Alpha-decay on Properties of Plutonium Metal", A. G. Seleznev et al., Radiochemistry, Vol. 37, 449-453, (1995)]. The results are not directly relevant to the US stockpile since all the samples were initially in alpha-phase rather than delta-phase. The measurements show that in pure Pu 238 the effects of radiation are so disruptive that the alpha-phase does not survive intact. The aged Pu 238 is a mixture of phases, while the aged weapons-grade plutonium is still alpha-phase. The aging effects seen in pure Pu 238 are quite different from the effects in weapons-grade plutonium accelerated by a factor of 280. The Russian results show that accelerated aging must not be too rapid if it is to be a valid model of natural aging. In order to have measurements of accelerated aging that can be trusted to predict effects in US stockpile plutonium, it is essential to use mixtures of Pu 238 with weapons-grade plutonium that will reliably remain in delta-phase during the aging process. Mixtures containing up to 10 percent Pu 238 will probably stay in delta-phase, but this must be verified as the experiments proceed.

Plutonium containing 10 percent Pu 238 will generate a heat output of about 70 watts per kilogram, compared with the 2.5 watts per kilogram of weapons-grade plutonium. To simulate the aging of pits in storage, the samples containing Pu 238 must be artifically cooled to the same temperature. This can be done.

Two major problems may arise in implementing the recommended program of accelerated aging studies. One problem will be to ensure a continuing supply of adequate quantities of Pu 238. The weapons laboratories state that they have ample supplies for the experiments that they are now planning to do. But this does not mean that they have ample supplies for the experiments that they will need to do in the future. We recommend that experiments using Pu 238 should be a central part of the pit stewardship program for many years to come. Pu 238 is a scarce resource and must be carefully husbanded. Experiments using Pu 238 should be designed to use small samples. To the extent practical, material studied under dynamic conditions should be recovered.

The second problem in carrying out experiments with Pu 238 is to deal with the associated radiological and environmental hazards. Pure Pu 238 is difficult and dangerous to handle. The hazards will be reduced if the fraction of Pu 238 in the samples is kept below 10 percent. If supplies of Pu 238 are maintained by extracting it from the spent fuel of naval reactors, there is no reason why the enrichment of Pu 238 need ever exceed 10 percent.

We recommend that the program of accelerated aging measurements using Pu 238 should be funded with a priority comparable with other essential elements of the stockpile stewardship program. It should be a substantial and permanent part of the stewardship of plutonium.

One potential problem with aging is failure of seals, or outgassing of other components of the weapon. Maintaining the warheads in a steady, controlled thermal environment at reduced temperatures could be beneficial in slowing the rate of such processes.

The existing program of work on the properties of plutonium includes diamond anvil studies of pressure response, positron annihilation studies of atomic void formation, transmission electron microscopy (TEM) of void formation, gas gun studies of equation of state, and studies of HE driven ejecta production. Additional high-resolution TEM studies of atomic structure, hydrodynamic tests, and gas-gun studies of spall formation are planned. There is clearly a need to organize this variety of tests into a coordinated program with a focus on setting specific, conservative criteria for retiring pits from the stockpile, and on defining the least expensive diagnostics which can be used to determine when the criteria are met.

4.3.2 Pit remanufacturing

Los Alamos has undertaken a pit remanufacturing program with excellent results. Using an advanced electrorefining technique, they are able to retrieve Pu from old units, achieving higher tolerances for purity than were possible in the past. They will use an improved casting process, which should yield savings in fabrication costs over old methods. Their stated goal is, "The pit rebuild project will deliver a functionally equivalent pit compared to those produced by the previous manufacturer at the Rocky Flats Plant." They presented evidence to the effect that the processing changes they are implementing are consistent with this goal. The specific 5 areas of process change are:

- Pu composition
- Pu casting
- Welding by pulsed laser
- Dry Machining
- Cleaning Solvent

The expected impact of these process changes are:

The Pu composition will be changed slightly from past processing by increasing the Ga component by approximately 0.1-0.2 percent and reducing the level of impurities by the same amount. This change is within specifications for the Pu, and should increase the workability of the material.

The use of the casting process is different from the most recently used wrought processing. However, both cast and wrought processes were used successfully in producing tested pits in the present stockpile. The only material property difference expected due to the change in processing is grain size distribution. The new casting process generates a Pu grain size distribution which is well within the variability observed in the existing stockpile.

The welds created using a pulsed laser beam rather than the previous technique of electron beam welding meet strength specifications, and appear to be clean and uniform. As there is considerable variability in weld structure within the existing stockpile, it is likely that the characteristics of the new welds fall within the range of characteristics in previously tested weapons. This should be confirmed.

The possible negative effect of dry machining is a pressure induced phase transition from the delta to the alpha phase. Checking for such damage will be a normal part of the manufacturing process.

The change in cleaning solvent is desirable for environmental reasons. The possible negative effect of such a change is a modification of the surface finish or composition. This seems unlikely, but can and should be checked.

The processing changes in the pit rebuild should result in a functionally equivalent pit as planned; this can be confirmed by an appropriate certification and qualification process.

4.3.3 Non-destructive diagnostics

For assessing the status of the pits, as well as of other metallic structural elements in the units, an efficient method of determining whether there are cracks, voids or areas of corrosion in or on the structure is important. There are several standard industrial probes for such structural defects, three of which, eddy current inspection, x-ray tomography, and ultrasound testing, were mentioned in the briefings as potentially useful in monitoring the status of pits. Each of these techniques has well known strengths and limitations in industrial applications, and each would require some development effort for the specific needs of the surveillance program. Because such development is likely to result in improved technologies with potential industrial applications, close interaction with industrial and academic partners in developing these tools is strongly recommended.

In determining which methods of non-destructive evaluation (NDE) to use, the goal should be to minimize cost while maintaining (but not exceeding) the quality of information needed to assure stockpile performance. Thus in assessing the usefulness of development of these techniques, an important step will be defining what level of information is needed, and how it will be used in decision making concerning the stockpile. Subsequent evaluation of the technology will require determination of the costs of its use. These will include the investment of developing and assessing the technology, of equipment, transport costs for moving the weapons to the test site (or the test equipment to the weapons), and training costs for personnel who run the equipment, and analyze and archive the data.

Scanning Squid Microscopy

Eddy current inspection is a widely used engineering technique for evaluating structures for cracks and other defects in metallic structures. It is especially useful for structures which must be tested in place, such as bridge reinforcement bars or airplane wings. The basic technique involves inducing a time-varying eddy current in the structural element, which in turn generates a magnetic field. The perturbation of current flow around cracks and defects leads to variations in the field. By scanning a probe of magnetic field across the surface of the element, it is possible to detect the spatial variations in field and relate them to underlying structural defects.

A major limitation of standard eddy-current testing is limited depth sensitivity. Because the skin depth scales as the inverse square root of the frequency, while the sensitivity of most magnetic sensors scales linearly with the frequency, standard eddy current testing is most sensitive to near-surface defects. The development of superconducting quantum interference device (SQUID) sensors for eddy current testing offers the potential for significant improvement in performance. There are two reasons for this. First, SQUID detectors are extremely sensitive. For instance a 10 micron SQUID loop can be constructed with sensitivity less than $10^{-5}T/\sqrt{\mathrm{Hz}}$. In addition, the SQUID signal amplitude is independent of frequency. Because of this, it is possible to perform eddy current testing at lower frequencies, allowing sensing of defects at depths on the order of at least 10 mm.

The LANL development project for scanning squid microscopy is expected to yield an instrument with spatial resolution of 0.02 mm. Whether this resolution is actually needed for NDE of weapons, and how it will be coupled to eddy current excitation were not discussed. Development of appropriate geometries for inducing the eddy current in the structure under test and analyzing the results will need equal emphasis to the development of the sensor technology for this program. One possibility that should be evaluated is the development of an eddy-current tomographic technique, which could be implemented (at least in principle) by the use of variable eddy current frequencies and induction geometries.

X-Ray Tomography and Ultrasound Testing

X-Ray tomography has the benefits of sensitivity to a wide range of materials, excellent depth penetration and well-developed understanding of procedures and analysis. It is likely therefore to be valuable for at least some testing of components of the stockpile. However, it does suffer from the drawbacks of expense of equipment, and the need to transport the item to be tested.

Ultrasound testing for industrial materials is carried out in a rather primitive form (simple recording of individual reflections as a function of time) compared to the relatively well developed techniques of ultrasound imaging used in medical studies. Like eddy-current testing, it has the benefit of relatively low cost, and portability of equipment. Also like eddy-current testing, it will need development effort to be useful to the specific needs of the surveillance program, and such development is likely to have useful industrial spin-offs. Developing a more sophisticated ultrasound detection technology for these applications by using time-resolution techniques to obtain depth resolution and tomographic imaging capability appears straightforward. Many of the techniques which can be used to improve medical ultrasound imaging (see JASON report JSR-95-145, "Ultrasound") can be also be applied to problems in NDE using ultrasound.

4.3.4 Fundamental studies and Modeling of Pu Materials Aging

The materials issues presented by aging of Pu constitute a multi-length scale problem, the kind of problem that is considered a "Grand Challenge" of materials science. Significant advances in materials prediction and design are now expected, due to advances in understanding how to extrapolate from observations (or calculations) at one length scale, to behavior at the next length scale. The philosophy of these advances is to recognize that very little of the wealth of information that governs the behavior at one length scale is important in detail at the next larger scale. To make significant progress, it is necessary to learn how to extract the "relevant" information at the smaller length scale and apply it appropriately to predict behaviors of interest at the next scale. Theoretical studies are needed to learn how to make the correlations across length scales. Then, even though theoretical calculations are seldom extensive or accurate enough to make quantitative predictions, one can use accurate experimental measurements at one length scale to make quantitative predictions about the behavior at the next scale.

Progress on this broad problem in materials science is in the early stages, and it is unlikely that the questions necessary to maintain the stockpile will be answered directly by theoretical studies alone. However, theoretical studies can provide corollary supporting information to guide experimental studies, and also have the potential to have significant impact on materials science. Such studies are worthy of support at a second level of priority if they are performed at a high level of scientific excellence.

The Livermore basic science studies of Pu address challenging problems beginning with ab initio calculations of energetics. Extension of this work to obtain a thermodynamic understanding by appropriately introducing thermal properties, and extension to larger length scales by introduction of models of dislocations, are under development within Livermore and with university collaborations. To yield a greater impact, more supporting experimental work, such as measurements of dislocation structures and their evolution at the micro- and meso-scale is needed and is in progress.

4.4 Secondaries

The dominant fraction of the yield of a modern nuclear weapon comes from its secondary; yet in many ways secondaries present a less formidable task for stewardship in a CTBT era than do primaries. Provided sufficient radiative energy is delivered to the secondary during the correct time interval, and the integrity and physical alignment of secondary components are within specifications, the performance of the secondary of a tested nuclear design can be guaranteed with high confidence.

Stewardship issues associated with secondaries may be grouped into three categories: (1) secondary remanufacture capability; (2) monitoring and prevention of the corrosion of uranium components of secondaries; and (3) verifying and maintaining the physical and chemical integrity of other materials in the secondary. We discuss these three categories in turn.

4.4.1 Remanufacture of Secondaries

The secondaries (commonly referred to as Canned Secondary Assemblies or CSAs) of all nuclear weapons in the US strategic nuclear stockpile have been manufactured in the Y-12 facility in Oak Ridge, Tennessee. US secondary manufacturing capability will remain where it has always been, in contrast to the case of pit fabrication. Therefore, less will change for CSAs. Nevertheless good management and continued support remain essential. The Y-12 facility is currently in the process of being restarted after being down since 1994 for environmental and other reasons. After detailed planning it has been decided that the future Y-12 facility will be sized to allow remanufacture (and repair) of secondaries at a normal capacity of 100 CSAs per year, with a surge capacity well beyond that.

In addition to existing manufacture and surveillance tools, two new tools are being developed for Y-12. The first new tool is a neutron radiography facility that Y-12 will use to nondestructively examine secondaries for voids and other internal problems. LLNL is developing the facility using its Enhanced Surveillance Program funding in cooperation with LANL, the Savannah River Technology Center, and Y-12. A complete facility will be delivered to Y-12 in about 4 years. The second new tool is an ultra high peak-power (petawatt) very short pulse (femtosecond) laser that can make thin, precision cuts in metallic components without heating the metal on either side of the cut to the melting point. Secondary components that have been cut apart in this fashion can be laser welded for reuse as desired. Both the neutron radiography facility and the cutting laser will be valuable additions to the capability of Y-12 in carrying out its stewardship responsibilities for secondary assemblies.

Current plans for secondary remanufacture and stewardship appear to be well founded. Providing that necessary support and management leadership is maintained, the planned annual capacity will allow conservative criteria to be used for replacement of canned secondary assemblies based on surveillance findings.

4.4.2 Prevention of Uranium Hydriding

Corrosion of uranium in the context of secondaries refers to its chemical reaction with hydrogen according to the process: $3/2\ H_2 + U \rightarrow UH_3$. In secondaries, hydrogen is generated by the reaction of moisture on lithuum deuteride: $LiD + H_2O \rightarrow LiOD + H_2$ or the corresponding process on lithium hydride, and perhaps by other sources. Sources of the moisture in canned secondaries include outgassing of water molecules from other components in the secondary. The hydride preferentially forms on edges and microfeatures on the surfaces of metallic uranium. Prevention of hydriding is important in secondaries, and has been managed effectively by careful manufacturing processes.

4.4.3 Secondaries

Development of preventive maintenance diagnostics in the form of (insitu) gas sampling sensors, especially for hydrogen and water, clearly should be a high priority task for enhanced surveillance and could be implemented in SLEP. There are several other materials in secondaries that need monitoring. Most ubiquitous among these is ^{6}LiD, a long studied and well characterized material, whose replacement when necessary raises no difficult problems.

Development of predictive capabilities to evaluate aging rates more accurately, and to prevent aging effects more aggressively (e.g. improved gettering), can be used to reduce costs by increasing the lifetime between replacements, and to help maintain a more stable level of quality in the stockpile. These goals should be kept in mind when evaluating the collaborative work in progress at LANL, LLNL, Y12, and SRTC to develop rigorous procedures for computer modeling of molecular transport and reactivity of gaseous species with components in the CSA. Molecular and microscopic level experimental data will be used to fix rate coefficients in the model. The fundamental studies of reaction processes which are being performed as part of this modeling effort are well focused on the key issues in hydriding.

4.5 Detonators, Initiators and Electronic Systems

The non-nuclear parts of a weapon must function with high reliability, and they are at least as complex as the nuclear parts. Even without a nuclear test ban treaty, a vigorous program of research, surveillance and maintenance would be called for, and therefore should be adequately supported. Where security considerations permit, advances in this area should be transferred to the civilian economy, with ample credit to the DOE innovators. Not only will this help to maintain the enthusiasm and motivation of DOE personnel, but it could have worthwhile payoffs, since the civilian economy pays a heavy price for failures associated with material aging – from roads and bridges, to automobile tires and bodies, to computer disk drives.

Relays, strong links, accelerometers, etc., contain mechanical elements that are designed to move appropriately on demand. The gears, lubricants, lock-and-key mechanisms, springs, etc., must function as designed indefinitely, with no deterioration due to accumulated dirt, viscosity changes of aging lubricants, corrosion or seizing of normally quiescent bearings, etc. The task of the DOE will be made more difficult by continuing replacement of mechanical devices in the civilian economy with modern solid-state counterparts. There will be a decreasing experience base with such mechanical devices in American industry.

Any component with the potential for electrolytic corrosion or recrystalization – for example batteries, igniters, electrolytic capacitors, solder joints, etc. – is at increased risk for failure and should receive special attention. With some components such as bridge wires, redundancy can be relied on to ensure reliable operation.

Polymeric materials, ranging from lubricants to O-ring seals, are widely used in the nonnuclear parts of weapons. They deserve very close attention, since many degrade slowly with age, chemical oxidation, sulfidization, etc.

The residual gas composition in the non-nuclear parts of the weapon should be monitored closely. This will provide useful clues to unsuspected decomposition of materials. Various corrosive gases like hydrogen sulfide may be generated.

It would be useful for DOE to institute a program to monitor the experience of some commercial devices, like air bags for passenger cars. These contain weapons-relevant accelerometers, ignitors and gas generators, and they are so numerous that good statistics on failure modes could be collected.

Integrated tests of the operation of the non-nuclear parts of nuclear weapons can and should be done from time to time to build confidence and to detect unsuspected failure modes. It would be bad economy to scrimp on the study of non-nuclear parts.

The DOE program should be especially careful about seemingly innocuous changes in the construction of non-nuclear parts. In the 1950's an intended new series of relays with silver-plated ceramics was introduced by the Bell Telephone System. Instead of achieving improved system reliability as hoped, many of these relays failed due to unanticipated dendritic growth of silver "whiskers" through the insulators and across the gaps. We fully expect that some replacements will be needed in the non-nuclear parts of the weapons. This would be true with or without a test ban. Our point here is that even with seemingly minor changes in the components, the DOE should maintain a paranoid suspicion of the capabilities of the new parts and of the overall, slightly modified system, and should insist on continued rigorous testing of the parts and of the system as a whole.

SNL has prime responsibilities for the many components of a weapon system (i.e. firing sets, neutron generators, etc) outside of the physics package, and for overall system integration. The lab's stated strategy appropriately focuses on identifying components that are most likely to wear out and need changes. It is important that programs to improve such components in search of longer lifetimes with modern (micro-) mechanical techniques should not sacrifice confidence in performance nor take necessary resources away from more mundane needs such as thermal battery performance. Equally important is program support for well-instrumented High Fidelity Joint Test Assembly (HFJTA) flights conducted with DOD to confirm overall performance of the weapon systems in meeting established military requirements.

5 PRESERVING THE HISTORICAL RECORD

The historical record relevant to stockpile stewardship and aging of weapon components includes data from all parts of the weapon complex, past and present, as well as from the commercial sector. Some parts of the historical record are being mined for important information, as in LANL's program to scan Rocky Flats documents; however, there is no aggressive effort to utilize the record more effectively. We learned that many records are in boxed storage in some unspecified, DOE warehouses.

The more obvious records of interest are as follows:

1. Production records for all weapon components from all plants, past and present.
2. Nuclear test records, including properties and conditions of the warheads recorded just prior to the tests as well as test results.
3. "Life history" records of weapons that have been or soon will be dismantled or inspected, as well as those remaining in the stockpile.
4. Disassembly/decommissioning records.
5. Records of relevant non-nuclear hydrodynamic tests.
6. Condition of spare parts and subsystems "on the shelf".
7. Personal interviews with personnel involved in production of critical components or subsystems.
8. Aging information from the commercial sector about plastics, potting materials, etc., that might be relevant, such as components subjected to low level radiation for many years at nuclear reactors, or in storage facilities for radioactive waste.

9. Information from non-US sources, especially from the UK, France and Russia.

The following are ways in which that historical record can be put valuable use:

1. Production Records

It would be useful to know the range of sizes that key components were "as-built." Was this range well within specified tolerances, or barely so? It would also be useful to know if any key item that should have been considered outside the established specification range was ever in a weapon that was tested and how closely manufacturing process specifications were met for weapons that were tested. (See also item 7, Personal Interviews)

2. Nuclear Test Records

Did pre-test radiographs of tested weapons ever show any "anomalies" that would raise concerns today about reliability and performance? If so, what were the results of these tests? Would the more sensitive "eyes" of the Enhanced Surveillance Program indicate anomalies not recognized previously, and, if so, what were the test results in those cases?

3. "Life History" Records

Rather than picking warheads for dismantlement and inspection by random selection, it would be useful to select those whose life history and production records suggest that they would be particularly interesting for aging studies. If would be of particular interest to compare two warheads of the same design and age, but different thermal histories.

4 . Dismantlement/decommissioning records

Components that survive weapon dismantlement can be examined and tested for the effects of aging, the results being correlated with their life histories. Likewise, any records that indicated that components from dismantled weapons were no longer "as-built" should be put together with production and life history records to look for useful information on aging.

5. Records of Relevant Non-Nuclear Tests

The historical record of past hydro-tests, i.e. pin-dome, core punch and other experiments involving HE and IHE in current stockpile weapons, provides a basis for comparison with future experiments, to observe whether aged explosive or replacement explosive manufactured by a different process has changed performance.

6. Condition of Spare Parts

Many stockpile reserve components have been "on the shelf" for many years. There may be some useful histories among those components (e.g.– some stored at high or low temperature) that can provide insight into aging processes.

7. Personal Interviews

Although formality of operations is the practice in the production environment, experienced production workers may know enough about what they are doing to take "short cuts" because they know they can achieve the same final product in less time. Therefore, we think it would be wise to interview some workers from key production lines while they can still remember what they really did. They are unlikely to recall anything about any particular component (pit, for example). However, they might recall whether they ever did something "a little different" from the standard process, and what that variation was.

8. Aging Information from the Commercial Sector.

The question here is whether aged plastics and other organics (including high explosive materials) used or stored in various environments might yield useful information for the weapon program. Are there support pads, encapsulants, etc., that have been subjected to radiation while being used at nuclear power plants, in the storage racks at commercial isotope suppliers, or in nuclear waste storage facilities? Perhaps the suppliers of plastic components to DOE production plants such as Pantex will be able to supply the names of other customers who bought the same materials for supports, spacers, etc., and those parties who might have had uses under conditions relevant to the stockpile could be contacted to find out how those materials have aged. The database available may be very large on some materials.

9. Information from Non-US records.

It would be worthwhile to seek information from the UK, France and Russia on any aspect of Pu metallurgy. Project Redbeard is indicative that the UK is willing to work with us in understanding the properties of Pu, and the Russians have published relevant studies too.

Collecting and organizing the vast historical records is not a task for scientists and engineers at the national laboratories, nor are they likely to be eager to do that job. However, those responsible for stockpile stewardship will certainly benefit from having it done. Hiring contractors to do the work should be relatively low cost compared with, for example, even a few major experiments on Pu. Once collected and organized, the records should be made accessible to anyone studying the effects of aging on the stockpile.

6 GENERAL OBSERVATIONS ON AGING SCIENCE AND REMANUFACTURE

The present study was chartered to focus on problems of the aging stockpile, and how to address them. But aging cannot be dissociated from questions of remanufacture and preventive maintenance. In crudest terms, the annual cost to remanufacture and maintain is inversely proportional to the reliable age of the stockpile weapons

The question is whether there is a minimum-cost point for a mix of remanufacture and science investigations of stockpile deterioration. In particular there is a need to analyze replacement costs of specific components. A natural approach to this problem is to work out goals and priorities, not for the aging program by itself and not for a remanufacture/preventive maintenance program by itself, but jointly.[1]

6.1 Goals and Priorities

Goals and priorities can be established by considering the degree of risk to weapons function, costs of alternative ways to reach the same goal, and applicability of scientific aging studies to maintaining confidence in the stockpile.

There is general agreement among the labs on the degree of risk associated with age-related failure of weapons components. The greatest worries in

[1]In any replacement or remanufacturing program, consideration should be given to "canning" various components – e.g. with a thin laminate or foil – to reduce loss of volatiles and to limit interactions among components caused by vapors. Of course this should be done only when analysis shows that there is no possibility of an added problem due to this barrier and that such an effort would be cost-effective.

the physics package concern the high explosives and pits; the organic (non-HE) materials and CSAs pose lower risks at this time. Of equal importance as the physics package are the electronic and mechanical components outside the physics package, which in general are not redundant and are subject to single-point failure. Failure of any of the highest priority components could lead to zero-yield.

Now consider the question whether it is better to prove by extensive aging studies (experiments and modeling) that a certain high explosive or energetic material has a specific reliable age, or to institute a sophisticated remanufacture/preventive maintenance program with only limited aging studies. The distinction here is one of emphasis: Of course, HE has a finite age, and eventually must be replaced, but if it can be determined that this is a very long age, then not much attention need be paid to streamlining the replacement process, deferred into the future. On the other hand, if the age is short, or if definitive answers are not reached, remanufacture may be more frequent, and should be made as easy and cheap as possible. The remanufacture program should have answers to such important questions. This will require many detailed scientific studies of a type that only the weapons labs can do.

6.2 How the DOD Can Help

The DOD is a customer with a great deal of influence over certain nuclear weapons matters, such as stockpile maintenance and the STS of various devices, but with very little say about weapons design and science investigations, once weapons requirements are set. It is important that DOD and DOE work cooperatively in setting requirements, especially those that drive large costs.

In particular there is an inertia resisting change stemming from operational requirements set long ago during the era of a large Soviet threat. In those days, it was considered that our weapons had to have very substantial radiation hardness to resist Soviet ABM attacks. This needs to be continually reexamined in the context of changes in international force and posture; the Soviet ABM threat, such as it is, still exists, but only at the level of 100 nuclear interceptors, and it is hard to believe that any other power, including China, would install a similar or larger nuclear ABM system. There are few, if any, commercial suppliers of electronics hardened to the dose-rate standards of the cold-war era (although total-dose hardening is important for non-military space electronics). It would be costly and, in our opinion, unnecessary to insist on replacement electronics as hard to radiation as was required in the past.

7 OVERVIEW OF RECOMMENDATIONS

The fundamental set of recommendations is simple: Provide a quantifiable set of priorities which combine the two key streams of i) scientific understanding and ii) remanufacture/preventive maintenance. Analyze the development of advanced remanufacture and maintenance technologies now, not at some time to be specified as a result of a decade or more of scientific studies. Apply this analysis to evaluating which long-term efforts are likely to be cost-effective, relative to remanufacture and replacement; support only those long-term efforts that have been evaluated in this manner and shown to be effective within useful (pre-established) time scales. Generate an understanding of this advanced remanufacture and maintenance technology to inform the DOD which changes in components are acceptable, and in some cases required, for continuing confidence in the stockpile. Provide a budgetary plan which makes it clear how the labs' expenditures over the coming decades contribute specifically to stockpile confidence, a plan which tightly integrates the labs among themselves and with the plants. Approach the questions of remanufacture and maintenance with the same spirit of scientific inquiry which has served the labs so well when their function was weapons design.

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LANL/LLNL/SNL Briefing Charts

A APPENDIX A: Likelihood Fits to Aging Data

We performed a maximum-likelihood analysis of aging data provided to us by the Sandia group[1]. This information consists of numbers of weapons sampled and numbers of "actionable findings" observed, sorted by age of weapon in one-year bins. Our purpose was to use these data to estimate as accurately as possible the rate of actionable findings and, in particular, to look for any significant change in defect-rates with age.

In analyzing these data, we assume that the probability, p, of a defect being recorded as an actionable finding during a given inspection is a *smooth function* of the age, y, of the sample group being examined. Specifically, we assume:

$$p = a_0 + a_1 y + a_2 y^2 + a_3 y^3$$

where the a_n are parameters to be to be fitted to the data. Further, we assume these parameters are non-negative, implying that defect rates will only grow over time, in the same way for all types of weapons. The probability that r actionable findings are observed in a sample of n weapons in the age bin, y, is given by the binomial distribution,

$$f(r; n, p) = \frac{n!}{r!(n-r)!} p^r (1-p)^{n-r}$$

where p is the defect rate evaluated for the age, y, being studied.

Our likelihood function is the product over age bins of the individual binomial probabilities for the observed numbers of findings. The fitting parameters, a_n, are varied to maximize the logarithm of the likelihood function†

†This form correctly handles binomial statistics and corresponds (up to a factor of -1/2) to a χ^2 or least-squares fit when the r_i are reasonably large.

(up to an overall constant that does not influence the fit), given by:

$$\ln \mathcal{L} = \sum_i r_i \ln p_i + (n_i - r_i)\ \ln(1 - p_i)$$

where the summation is over all age bins, i, contributing to the fit; r_i is the number of actionable findings in the sample of n_i weapons examined in the given age bin and p_i is the actionable finding rate evaluated for the i-th bin.

The MS Excel tool, "Solver", was used to find fitting parameters that maximize $\ln \mathcal{L}$. Various fits were performed in order to study the stability of the results[‡].

The final results, shown in Figure 1, included data in the age range 5 to 32 years. Data from years 1 to 4 are well represented by the parameters determined from the older weapons, but were excluded from the fitting process to avoid possible biases arising from the "youngest" samples. It is interesting to note that the fits consistently found solutions where $a_2 = a_3 = 0$, presumably, a consequence of the positivity constraint. The best fit values for the other parameters are:

$$a_0 = 0.002642 \text{ findings/trial} \quad \mathrm{a}_1 = 0.000355 \text{ findings/trial/year}$$

To estimate the errors on the fitted rate of actionable findings, the values of a_1 that change $\ln \mathcal{L}$ by -2 units (corresponding to a $\pm 2\sigma$ variation in likelihood) were found. Curves (straight-lines) for these parameters are included in Figure 1. A proper calculation of the covariance matrix for these fits is beyond the scope of this report, but we believe the above procedure gives a reasonable estimate for the 90% confidence-level bounds. The two parameters, a_0 and a_1, are highly correlated; by using the slope parameter, a_1, to estimate errors, we should be conservatively setting confidence-level bounds for the oldest weapons under consideration.

[‡]We also performed fits with a likelihood function based on Poisson statistics. The results were indistinguishable from the binomial case.

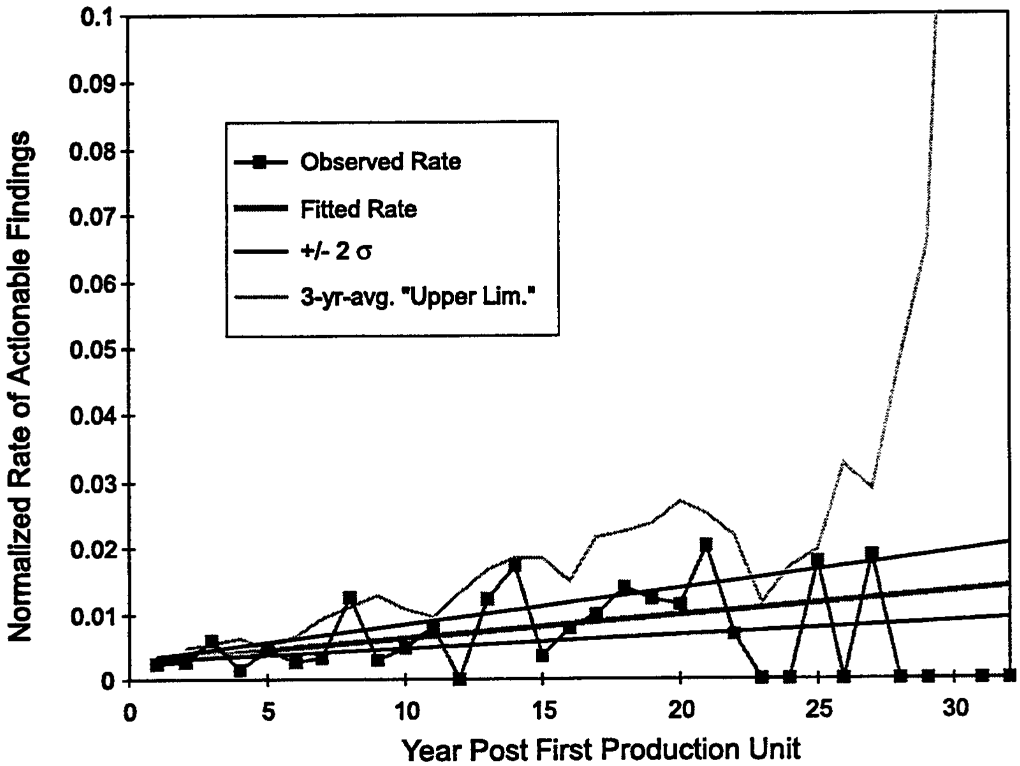

Figure 1. Summary of results on age-dependence of rate of actionable findings. The square boxes indicate the observed number of actionable findings[1] divided by number of samples, plotted against the cohort age. The line labeled "Fitted Rate" is the actionable findings rate versus age determined from the maximum-likelihood fit described here. The lines labeled "+/- 2 σ" indicate our estimate of the 90% confidence-level bounds for the rate actionable findings. The curve "3-yr-avg labeled Upper Lim." is the previously reported estimate [1] of the 90% upper limit on the rate of actionable findings, based on a three-year running average.

The results of our fit suggest a straight-forward picture of the aging process in these devices: A given weapon is a very complicated assembly consisting of large numbers of components, some of which are aging and may trigger actionable findings when examined. With many possible and nearly independent pathways leading to an actionable finding, the weapon, in this picture, behaves much like a sample of unstable particles: after a time, t, the fraction surviving is given by the exponential decay law,

$$f(t) = \exp(-\lambda t)$$

which is a consequence of the independent nature of the decays, as described by Poisson statistics. The parameter λ is the mean decay rate, or inverse of the mean-lifetime, $\tau = 1/\lambda$. In this picture, which we call the "Poisson model" of aging, the individual "decays" of components lead to a "build up" over time of actionable findings. Thus, the time dependence of the probability for actionable findings should have the form:

$$p(y) = 1 - \exp(-\lambda y)$$

In the limit of small λy, this reduces to the linear term, found in our fits. It also naturally explains why the fits found zero-values for the higher-order fitting parameters. Our fits also find a constant term, a_0, which can be interpreted as arising from some different, non-aging mechanism, such as "infant mortality" or defects that were hidden in post-production inspections[§]. Interpreted via this simple picture, which we believe to be plausible, the data suggest a constant defect probability of 1/4% and an aging lifetime of 2800 years.

Also plotted in Figure 1 is the 90% confidence-level "upper limit", proposed in SAND95-2751 (UC-700). This limit was computed from a 3-year running average of the observed findings rate. As expected, the higher statistical power of the maximum-likelihood method yields a considerably more

[§] Alternatively, the constant term can be interpreted as the "clock" for defects in the Poisson model starting 7 years before the weapon is declared "produced" accurate estimate of the rate of actionable findings, particularly for the older weapons. The basic assumption underlying our procedure—smoothness in the age-dependence of the underlying failure rates—is physically reasonable and gives no indication of significant increase, beyond a linear rise with age. in the probability of age-related findings for weapons greater than 27 years old, where, in fact, no such defects have been observed in the data available to us.

References

[1] Data on Actionable Findings, kindly provided to us by Robert Paulsen of Sandia.

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