MARTIN MARIETTA

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**Containment and Storage of Uranium
Hexafluoride at U.S. Department
of Energy Uranium Enrichment
Plants**

**UF₆ Containment and Storage
Committee**

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MANAGED BY
MARTIN MARIETTA ENERGY SYSTEMS, INC.
FOR THE UNITED STATES
DEPARTMENT OF ENERGY

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***Containment and storage of
uranium hexafluoride at US
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**CONTAINMENT AND STORAGE OF URANIUM HEXAFLUORIDE
AT U.S. DEPARTMENT OF ENERGY
URANIUM ENRICHMENT PLANTS**

Prepared by
UF₆ Containment and Storage Committee
C. R. Barlow, Chairman

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CONTENTS

EXECUTIVE SUMMARY	iv
INTRODUCTION	1
PROGRAM ELEMENTS	5
MONITORING OF CONTAINMENT INTEGRITY IN STORAGE	5
CORROSION STUDIES ON STORAGE CYLINDERS	7
DEVELOPMENT OF STANDARDS FOR IMPROVED STORAGE	8
IMPROVED PROTECTIVE COATINGS TO EXTEND STORAGE LIFE	9
RUPTURE TESTING AND PROOF TESTING TO SUPPORT CYLINDER DESIGN OBJECTIVES	10
OVERPACK EVALUATION AND IMPROVEMENT	11
NEW PROJECTS FOR IMPROVED STORAGE	12
SUMMARY	14
DISCUSSION	15
REFERENCES	18
Appendix: PHOTOGRAPHS	20

FIGURES

1 CYLINDER STORAGE AT OAK RIDGE K-1066-E	21
2 STORAGE CYLINDERS AT PORTSMOUTH	22
3 DEPLETED UF ₆ IN STORAGE AT PADUCAH	23
4 TYPICAL 1-IN. UF ₆ VALVE	24
5 RAYGO-WAGNER CYLINDER STACKER	25
6 STRADDLE CARRIER FOR IN-PLANT CYLINDER HANDLING	26
7 SCALE ACCUMULATION RESULTING FROM GROUND CONTACT	27
8 PITTING ALONG LINE OF GROUND CONTACT	28
9 LEAKAGE VERIFICATION/MONITORING ON STORAGE CYLINDER	29
10 RUST ACCUMULATION IN SKIRTED CYLINDER	30
11 BREACHED CYLINDER IN K-25 SITE STORAGE	31
12 CORROSION TEST COUPONS AT K-25 SITE	32
13 TEST COATINGS ON STORAGE CYLINDERS	33
14 RESULT OF BURST TEST ON MODEL 48T CYLINDER	34
15 HANDLING DAMAGE IN CYLINDER STACKING	35

EXECUTIVE SUMMARY

Isotopically depleted UF_6 (uranium hexafluoride) accumulates at a rate five to ten times greater than the enriched product and is stored in steel vessels at the enrichment plant sites. There are approximately 55,000 large cylinders now in storage at Paducah, Kentucky; Portsmouth, Ohio; and Oak Ridge, Tennessee. Most of them contain a nominal 14 tons of depleted UF_6 . Some of these cylinders have been in the unprotected outdoor storage environment for periods approaching 40 years. Storage experience, supplemented by limited corrosion data, suggests a service life of about 70 years under optimum conditions for the 48-in. diameter, 5/16-in.-wall pressure vessels (100 psi working pressure), using a conservative industry-established 1/4-in.-wall thickness as the service limit. In the past few years, however, factors other than atmospheric corrosion have become apparent that adversely affect the serviceability of small numbers of the storage containers and that indicate the need for a managed program to ensure maintenance of containment integrity for all the cylinders in storage.

The program includes periodic visual inspections of cylinders and storage yards with documentation for comparison with other inspections, a group of corrosion test programs to permit cylinder life forecasts, and identification of (and scheduling for remedial action) situations in which defects, due to handling damage or accelerated corrosion, can seriously shorten the storage life or compromise the containment integrity of individual cylinders. The program also includes rupture testing to assess the effects of certain classes of damage on overall cylinder strength, as well as ongoing reviews of specifications, procedures, practices, and inspection results to effect improvements in handling safety, containment integrity, and storage life.

The program is intended to assure the retention of storage cylinder containment integrity and design function until the point of final disposition. The inspection activities have identified several breached cylinders, two at Portsmouth and four at Oak Ridge, in which the damage was caused by improper handling or storage practices. They have also identified a number of cylinders in ground contact where accelerated corrosion can seriously shorten storage life. In addition, early stacking practices gave cylinder arrays that are not accessible for proper visual inspection. These situations will be corrected as funding and manpower become available. The aim of program oversight efforts is to assure that all cylinders are handled and stored so that the maximum storage life, as controlled by normal atmospheric corrosion rates, is realized. Present plans anticipate conversion of the depleted UF_6 to oxide beginning at some time before the year 2020, if alternative uses are not developed.

It should be emphasized that the "end of storage life" for a UF_6 cylinder is only a nominal concept, or definition, and does not in itself suggest an imminent hazard

or danger of loss of containment. At the end of its storage life, the cylinder still conforms to its code design and is still fully qualified for the pressure cycle associated with liquefaction of its contents. The accelerated corrosion processes that have been observed, however, are seen as presenting a real danger of penetration and loss of containment if not properly managed and corrected. Handling damage presents a similar, but more immediate danger. The inspection procedures, therefore, are largely directed toward the detection and amelioration of these factors.

INTRODUCTION

Uranium occurs in nature in two principal isotopes of atomic weights 235 and 238. The fissionable isotope ^{235}U comprises about 0.711% of the total, and isotopic enrichment by the gaseous diffusion process yields a product in the neighborhood of 3 to 5% ^{235}U for reactor fuel and a depleted stream in the general range of 0.2 to 0.5% ^{235}U , depending on a variety of economic factors in effect at the time of processing. The enriched product (in the form of UF_6) is shipped to the consumer in steel cylinders of 2-1/2-ton capacity enclosed in protective overpacks designed to mitigate the foreseeable effects of most transport accident scenarios, while the five-to-ten-times-more voluminous isotopically depleted material is stored in steel vessels of 14-ton capacity, usually at the enrichment facility plant site (Figs. 1-3; all figures are located in the appendix). Because of space requirements, storage is outdoors and largely unprotected from the elements. The cylinders are arrayed side-by-side in rows of 50 to 100 units, supported on contoured wood or concrete saddles for separation from the ground and for control of spacing and stability. A second row of cylinders rests on top of each ground-level row, with each upper-row cylinder resting in the valley between each pair of lower-tier cylinders. The storage surfaces are either concrete or compacted gravel.

Although new cylinder procurements for many years have specified the type (and later, the method) of paint to be applied by the cylinder manufacturer, this paint has not been intended as protection in the storage environment and in practice has been observed to blister and peel because of steam exposure in the autoclave. Thus, it gives only short-term protection in storage. The storage life of UF_6 cylinders is controlled, therefore, by atmospheric corrosion rates of the steels used in their construction. The present strategy for managing the growing numbers of stored UF_6 cylinders is aimed at identification, avoidance, and correction of the factors that cause corrosion in excess of atmospheric rates and thus detract from this maximum storage life. In addition, storage management must also identify and correct the causes (and results) of mechanical damage that may compromise containment integrity. With these factors under control, the optimum storage life can be achieved and the most economical planning effected for ultimate conversion and disposal of the depleted uranium.

The uranium enrichment program has used steel cylinders for shipping and storing natural, depleted, and enriched (product) UF_6 since the late 1940s. The early accumulation rate of fewer than 1000 cylinders of partially depleted UF_6 per year from the enrichment cycle accelerated to more than 2000/year at the inception of the toll enrichment program in 1969. Currently, more than 55,000 steel cylinders of 10- and 14-ton capacity are in service, mostly for long-term storage of depleted uranium. The present inventory of depleted material contains approximately 570,000 tons of UF_6 , or 390,000 tons of uranium.

UF₆ handling in the enrichment plants for more than 40 years has been remarkably free of incidents with potential for damage to the environment or to plant or community health or safety. The risk of releases of UF₆ to the environment because of valve failure was recognized as a potential problem early in the enrichment program. The industry-standard fluorine valve used for UF₆ cylinders was redesigned, and the new valve has been subject to only minor problems since its introduction in the mid-1950s (Fig. 4). Hazards associated with potential cylinder rupture because of handling accidents were reduced by administrative procedures prohibiting intraplant transport of liquid-filled cylinders (dating from about 1978), and the recognition of a potential for brittle fracture in winter-season cylinder handling operations gave rise to the specification of a steel with certified low-temperature impact response in all cylinder procurements since 1979. The observation of lamellar tearing as a failure mechanism resulting from low-level impact during handling operations in shipping/storage led to a further modification of the specification for cylinder steels in 1984. Greater resistance to damage from handling contacts is now assured through the use of steels with a low sulfur content, which improves ductility and impact strength in the short transverse (through-thickness) direction.

An integrated program of activities related to the containment and storage of depleted UF₆ has been established to provide oversight of objectives and reports of activities and progress in projects that have as a goal the advancement of safety and integrity of long-term storage. These projects include increasing storage capacity to alleviate overcrowded conditions in the storage yards, renovating and upgrading storage yard surfaces, improving the handling and stacking of cylinders, restacking to correct present deficiencies (e.g., degraded support saddles, ground contact, and inaccessibility for full inspection), identifying cylinders in which accelerated corrosion has thinned wall areas below minimum specified levels, and implementing uniform procedures and practices used in storage of UF₆ throughout the enrichment complex. The projects are supported by experimental evaluation of stresses resulting from normal handling and stacking procedures, and by the steel corrosion test programs at the three plants, the cylinder life-cycle study at the Paducah Gaseous Diffusion plant, and the cylinder rupture test program at the Oak Ridge K-25 Site. Also included are the three-plant activities relating to upgrading the Department of Transportation (DOT) Specification 21PF-1* protective overpack, the development of inspection criteria to assure continuing safe storage and containment of depleted UF₆, and the development of standards for long-term retention of UF₆.

The present storage arrangement for UF₆ cylinders was instituted primarily for inventory control. Specially designed or adapted cylinder handling equipment was developed for the storage of depleted UF₆ (Figs. 5 and 6), but the stored cylinders were not arrayed to facilitate inspection or monitoring for other features. Inspection

*Title 49 *Code of Federal Regulations*, part 178.352, specification 21PF "Fire and Shock Resistant, Phenolic-Foam Insulated Metal Overpack."

and maintenance were not seen as needs of the storage system until corrosion damage resulting from direct ground contact (Figs. 7 and 8) was observed in the 1970s during cylinder movements associated with the reprocessing of partially depleted UF_6 and with the sale of depleted materials. In an early attempt to correct this deficiency, some 3000 cylinders stored on asphalt paving in a one-time parking lot at the K-25 Site were removed in 1982 and restacked in a part-concrete, part-compacted gravel storage yard. Continuing observations of cylinders in storage have prompted the recent development of monitoring and inspection programs for early detection of incipient or active valve leaks¹ (Fig. 9) and tracking of cylinder wall thickness to act as a guide in scheduling maintenance or planning for material transfers to new cylinders if needed to maintain containment integrity.

As the foregoing listing of current projects indicates, a substantial level of ongoing effort is directed toward the assurance of safe containment and storage of UF_6 . These activities are the outgrowth of many years of attention to safe handling of this material and are based on a long-term and continuing policy that has given priority to safety in handling UF_6 . For more than 20 years, an interplant UF_6 Handling Committee has been responsible for ensuring safety in plant operations involving UF_6 outside of the diffusion process cascades. Among the committee's accomplishments have been the establishment of uniform practices regarding the following:

- operation of autoclaves to liquefy UF_6 for impurity and isotopic sampling and for feeding to the cascades,
- operation of freezer-sublimers for power and inventory control,
- cylinder weighing activities, and
- cooling requirements for liquid-filled cylinders before transfer or transport operations.

A subcommittee to the UF_6 Handling Committee, the Cylinder and Valve Specifications Subcommittee, from the early 1960s to the early 1980s, exercised oversight over all designs, specifications, and procurement activities relating to cylinders and valves for UF_6 storage and transport. This subcommittee developed the Model 30B cylinder design for transport of ^{235}U -enriched UF_6 , as a successor to the Model 30A (DOT-specification chlorine) cylinder that was originally obtained from U.S. Department of Defense World War I-era surplus chemical warfare stockpiles. They were also responsible for administering quality criteria for procurement of all code cylinders and for procurement of the cylinder valves whose design and material specifications grew out of K-25 Site concepts from the early 1950s. These valves incorporated the stem threads and packing within the valve body, rather than in a separate bonnet, and were made from a single-phase aluminum bronze to reduce the tendency to failure by stress-corrosion cracking. The subcommittee initiated the certified volume and fill-limit concepts that ensure safe filling and reliquefaction of UF_6 in all standard cylinders in the feed and product cycles of the enrichment plants. They developed and evaluated design changes in UF_6 cylinders to enhance safety of containment, ease of handling, and durability in transport and storage.

The current array of projects relating to containment and storage of UF_6 is administered by the Containment and Storage Committee, whose charter comes from the Martin Marietta Energy Systems, Inc., (Energy Systems) Interplant Operating Committee. This committee, established in 1990 from a working group representing the three storage sites, had functioned informally for several years before issuance of its formal charter. The group evolved because of a growing awareness of the need to assure the safety of containment during in-plant storage and the suitability for conversion to alternate forms at such time as continued storage in the form of UF_6 is no longer an option. A major aim of the UF_6 Containment and Storage Committee is to identify and develop the technologies that can be employed to optimize the integrity of the containment vessels and to improve the precision in the forecast of their useful life.

PROGRAM ELEMENTS

MONITORING OF CONTAINMENT INTEGRITY IN STORAGE

For the past 15 years, storage cylinders for depleted UF_6 have been purchased with a zinc chromate primer and an enamel topcoat, but these coatings are only intended to protect the cylinder until it is filled and deployed in a storage yard. The coatings are not resistant to steam in the autoclave cycle applied to single-use feed cylinders, which constitute a significant fraction of new storage cylinders, and are not touched up to repair incidental handling damage before storage deployment. In unprotected outdoor storage, therefore, the cylinders are subject to continual rusting, and their service life in this environment is limited. Present plant operating procedures prohibit autoclaving for liquid UF_6 transfer if the cylinder wall thickness falls below 1/4 in. (this applies to all 100-psig cylinders designed for storage of partially depleted UF_6). Observations at the Paducah storage site indicate that atmospheric corrosion processes consume about 0.001 in. of steel per year, giving an estimated storage life for a new cylinder of some 60 to 70 years.²³ A few thousand of the cylinders now in storage have served in this capacity since the latter part of the 1950s, and thus have a remaining life expectancy of 20 to 30 years.

While a general thickness reduction of about 0.001 in./year, resulting from atmospheric weathering, will give a storage cylinder life of 60–70 years, other factors are recognized that promote localized attack at significantly higher rates. Pitting attack and crevice corrosion have been observed beneath the cylinder nameplate and in the areas of contact with the wooden support saddles of bottom-tier cylinders. Recent inspections have shown that the stiffening channel on the Model O cylinder is particularly susceptible to stacking damage, and this, in turn, may have promoted accelerated attack in areas of the cylinder wall covered by the channel. Damage to the containment envelope can also result from impacts with the lifting lugs of adjacent cylinders during cylinder placement. Because of the nature of the stacking process, these will usually occur in the most sensitive areas, near an adjoining stiffening ring or lifting lug. The load concentration in an area of high lateral restraint can result in a through-thickness crack in the cylinder wall. Unless such a crack is detected immediately, it will be sealed by reaction products and then will grow slowly through corrosion of the steel by the HF produced from the reaction of UF_6 and moist air. As the hole enlarges, uranium compounds as well as HF are released from the cylinder.

Probably the major promoters of accelerated corrosion on storage cylinders are ground contact and storage in persistently damp areas. Significant corrosion occurs only in the presence of moisture, and the protracted or continuing presence of moisture along the cylinder-to-ground interface results in wall thinning rates that may be 10 to 20 times higher than those where surfaces can dry freely. Several hundred cylinders have been found to be in contact with, or settling into, support surfaces in

cylinder storage yards. Many more are within a few inches of the ground, in the zone where sustained high humidity is known to accelerate corrosion of steels. An additional area of concern is the head region on skirted cylinders (models T, H, and HX). Heavy rust deposits trapped in the skirt (Fig. 10) have been found to promote accelerated corrosion of the adjoining cylinder head because of moisture retention and increased time of wetness.

Ultrasonic thickness measurements will be obtained from a select number of problem cylinders in storage at each of the three storage sites (these measurements have already begun at Paducah and Portsmouth). The ultrasonic technique will also be applied to evaluation of areas of severe corrosion that are found during visual monitoring. In this manner, it will be possible to track the progress of both general corrosion and specific areas of localized attack.

Visual monitoring of cylinder integrity will also include conditions of valves and plugs. It is known from a recent cylinder storage audit that a number of valves at each of the storage sites show external uranium contamination at levels above plant-allowable limits. Work is now in progress to identify whether these valves are actually leaking, and work is planned to further characterize the extent and magnitude of valve leaks in storage cylinders. Potential or suspected plug leaks are also to be monitored.

Visual monitoring of stored UF_6 cylinders has resulted in the discovery of six breached cylinders, two at Portsmouth, found during the initial formal inspection in mid-1990, and four at Oak Ridge, during an enhanced inspection in 1991-92 using improved procedures and inspection techniques. All were discovered through the observation of reaction products beneath the breached cylinders, with further scrutiny revealing the actual locations of the holes. Both of the Portsmouth breaches appeared to be the result of handling damage incurred during stacking for storage. Two of the Oak Ridge cylinders show evidence of similar origin (Fig. 11), while the other two appear to be the end result of pitting corrosion incurred during protracted ground contact. It should be noted that, in each case investigated, the amount of uranium material that escaped from the breached cylinder was minor, and most of this was subsequently recovered.

Upgrading of the cylinder storage facilities is under continual review to identify and correct problems that are seen as having adverse influence on storage integrity, with regard to containment and the ability to safely transfer the stored material for final disposition. Cylinder designs and materials, and handling practices, are being examined and evaluated to furnish more precise estimates of cylinder storage life. Cylinder yards have been prioritized for restacking to correct ground contact situations and stacking geometries that inhibit full visual access for periodic inspection. A high priority is given to upgrading of storage surfaces (an ongoing program at Paducah) to provide stable support and good drainage, and cylinders from deteriorated storage situations are being relocated to the improved surfaces.

These efforts are directed toward optimization of storage life, so that the general atmospheric corrosion rates will be the major factor in determining cylinder lifetimes.

CORROSION STUDIES ON STORAGE CYLINDERS

Unprotected steel corrodes in outdoor environments at rates that depend on a large number of factors, including atmospheric chemistry and the presence and character of rainfall and of condensable moisture, as well as the steel composition and microstructure. Rates may range from a few tenths of a mil per year to 10 mils per year, depending on the specific location. Paducah plant personnel measured wall thickness on several storage cylinders in 1974, and again measured the same cylinders in 1988. They concluded from these observations that the thin wall cylinders would still be serviceable (i.e., the wall thickness would still exceed 0.250 in.) in 30 years: 2018. The observed corrosion rates support a life estimate of 60–70 years for storage cylinders. These Paducah cylinders constitute the basis for a “life-cycle” study, along with a test cylinder that was emptied and cut open for accurate micrometer thickness measurements.

The life-cycle study is supplemented by the initiation of atmospheric corrosion test programs that use the testing procedures and coupon geometries given in ASTM G-50.* Test coupons are presently deployed in storage yards at all three principal storage sites (Fig. 12). The coupons are part of a coordinated program to develop site-specific corrosion rate data through both weight-loss and thickness-loss measurements. Ultrasonic measurements of cylinder wall thickness will be expanded at the Paducah plant and will be extended to the Portsmouth and Oak Ridge sites as well. Some 500 cylinders representing the entire production history of storage cylinders will be surveyed periodically to monitor the wall thickness-loss rate. Periodic visual inspections now monitor corrosion of the steel cylinders, along with other factors affecting the quality of long-term cylinder storage, and will eventually include observations of localized attack and pitting, primarily in the areas of cylinder support and in restricted regions associated with skirts, lifting lugs, and areas of handling damage. Time-of-wetness probes and instantaneous corrosion rate probes will be strategically deployed in these areas to support the coupon tests and the life-cycle studies now under way.

The corrosion monitoring programs are being developed and conducted to provide a rational basis for storage management, including the economic scheduling of retirement or maintenance where necessary as cylinders approach the end of their safe-storage life, as well as for recommending a starting schedule for final disposal of the depleted material.

*ASTM-G50, Standard Practice for Conducting Atmospheric Corrosion Tests on Metals, American Society for Testing and Materials.

DEVELOPMENT OF STANDARDS FOR IMPROVED STORAGE

During some 30 years of UF_6 storage at the three enrichment facilities, a number of problems have materialized. Originally addressed on a case-by-case basis, these deficiencies have been more thoroughly characterized during the past two years of close scrutiny of the storage yards. The unprotected steel cylinders have a finite life in outdoor storage, and any long-term storage program whose duration exceeds this finite lifetime must consider both the economics and the logistics of either periodic cylinder exchanges to correct the effects of atmospheric corrosion and localized attack because of other factors or the prevention of corrosion through painting, protected storage, or other appropriate means. Either option requires implementation of a formal program of monitoring the condition of the storage system. When disposal of the stored material is planned to occur at, or before, the end of the cylinder lifetime, the storage system must still be monitored to provide the necessary assurance that optimum corrosion life is achieved. This type of surveillance program cannot be fully implemented at present because many of the stored cylinders are not situated to provide adequate access for a detailed visual inspection. The stacking arrays that were originally provided were designed only to facilitate inventory and accountability activities. Thus, while aisle space has been provided for easy access to the valve end (and nameplate) of all cylinders, the plug ends are in some cases too closely spaced to permit inspection. It is apparent that, if visual monitoring of all cylinders is to be a viable means of supporting storage management, many of the approximately 55,000 cylinders now in storage will have to be restacked to provide full access to both valve and plug ends. This improved accessibility would also provide space for service equipment (such as a pump cart) for occasional maintenance activities such as cold pressure tests, field pump-down, and valve changes.

Upgraded storage should provide for improved cylinder support saddles. The creosote-treated wood saddles used in the past support a degree of pitting corrosion because they retain moisture in areas of contact. With creosote no longer environmentally acceptable as a wood preservative, present procurements of wood saddles specify pressure-treated materials. It is not certain that the pressure treatment chemicals will be benign in (wet) contact with steel. These are being evaluated for their influence on possible pitting corrosion. The Paducah-developed concrete saddles, in use as replacements for the past 15 years, are also under evaluation for possible corrosion effects. Upgraded storage should also provide for improved drainage on both concrete- and gravel-paved storage yards. Most of the present facilities do not have effective drainage systems. Relocation of the cylinders to upgraded facilities should also consider the economics of painting versus the possible need for cylinder exchanges because of excessive corrosion losses in a number of cylinders.

IMPROVED PROTECTIVE COATINGS TO EXTEND STORAGE LIFE

While the normal rate of atmospheric weathering appears to be 0.001 in./year or less, giving an optimum storage life in excess of 60 years, factors that increase time-of-wetness cause an accelerated attack that may substantially shorten this storage-life expectancy. The inspection and monitoring programs for cylinder storage are therefore structured to identify and correct these factors, and to assess the degree of damage to the affected cylinders. It is anticipated that cylinders that show significant thinning (i.e., wall thickness approaching the limiting 1/4 in.) can be stabilized for their term of storage by application of a suitable protective coating. Cylinders with wall thickness already below 1/4 in. can also be stabilized by cleaning and painting, but must then be segregated and identified for special handling when they are scheduled for emptying.

Early cylinder production procurements specified a coating of green paint, without spelling out any details on quality, type, or preparation. Specifications for the Type-T cylinder (1956-1958) called for a coat of zinc chromate primer and a coat of jade green paint (Pittsburgh # 54-130 or approved equal). A 1979 revision to the cylinder specification (JSP 553, Rev. 5) called for a surface "free from all foreign material such as oil, grease, soap, loose mill scale, welding slag, and moisture." This cleaned surface was to be primed with one coat of zinc chromate, Pittsburgh 6-204, followed by one coat of avocado, Pittsburgh 54-304. The current specification, Rev. 16, has increased the requirements for surface preparation and paint quality:

Prior to painting, the exterior surface of the shell and heads shall be sandblasted to a commercial finish. . . . The cylinder shall be primed with one coat of Carboline 858 Primer (three component, zinc-filled, epoxy primer). The finish coat shall be Carboline 834 (acrylic aliphatic polyurethane).

None of these coatings has yielded any significant degree of protection against corrosion in the storage environment. With the lack of control over surface preparation, they have been prone to blistering and peeling, particularly in the autoclave cycle. Field studies have recently been initiated to test both organic and inorganic zinc coating systems to provide extended protection to corrosion-damaged (or new) storage cylinders.

Fourteen new model 48G cylinders (the current design) were set aside from a current order for storage cylinders. These have been processed through a commercial facility, where they were first cleaned by abrasive blasting and then coated with the presently specified zinc chromate primer/epoxy enamel topcoat or with one of two metallic zinc-bearing systems. These cylinders have been in outdoor storage at the Paducah Gaseous Diffusion Plant since October 1991 (Fig. 13). They have been filled with depleted UF_6 , and some are to be processed repeatedly through an autoclave cycle in an accelerated durability test.

Close monitoring of these cylinders in the storage environment can give the needed evaluation in a fairly short time. The principal characteristic needed for storage durability is adherence, and this feature receives a severe test in the steam autoclave. If the coatings survive multiple cycles in the autoclave without blistering, subsequent damage will occur only during handling for storage deployment. Other users' field experience shows that the zinc-containing coatings will protect steel structures for periods of 25 years or more when properly applied. The inorganic zinc coatings on the test cylinders have shown a remarkable resistance to handling damage. Where scratches, scrapes, and gouges in the other coatings have manifested their presence by rust development in the short exposure period since deployment, the cylinders with the inorganic zinc coatings have shown no rusting, although the same equipment and procedures were used in their handling and placement.

RUPTURE TESTING AND PROOF TESTING TO SUPPORT CYLINDER DESIGN OBJECTIVES

The cylinders used for UF₆ containment for both transport and storage are American Society for Mechanical Engineers (ASME) code-design vessels rated for service at maximum pressures of 200 psig or 100 psig. The code qualifies a cylinder on the basis of design, manufacturing surveillance, and a proof pressure test; industry practices, primarily as given in ORO-651⁴ and ANSI N14.1,⁵ require that periodic inspections and tests be performed to maintain this qualification.

The ASME design limits the maximum wall stress to 1/4 of the minimum ultimate tensile strength of the material of construction at the working pressure, and requires a post-manufacture proof pressure test that is defined by ORO-651 and ANSI N14.1 as double the working pressure. With the added thickness for corrosion allowance, new code vessels may therefore be expected to give a nominal burst pressure of about 8 times the working pressure (e.g., 1600 psig for all of the "shipping" cylinders and 800 psig for the thin-wall 48-in. "storage" cylinders). Since 1959, rupture tests have been conducted on all sizes of the 200 psig product cylinders, for design verification, evaluation of the influence of modifications or handling damage, and testing for the effects of accidental fire exposure. All of these cylinders were found to give burst pressures exceeding the design criteria, in one case by a factor of five. A non-ASME vessel, the 30A cylinder, was found to be a deficient design (for UF₆ service) in a number of areas and is being excluded from use in toll enrichment product shipments after December 31, 1992.

Large-capacity cylinders of 48-in. diameter require external stiffening rings to support the loads imposed by evacuation for filling. Rupture tests on several of the 48-in. designs have shown that this stiffening ring also becomes a critical feature in the event of internal overpressure. An incompletely penetrated butt weld in the stiffening ring, typical of the cylinders tested to date in this program, is subject to early failure and serves as the rupture locus (Fig. 14), sometimes at pressures well

below the nominal value of eight times the working pressure. The eight-times factor is recovered if a full penetration weld is used, but the consequence is a large-area rupture that can result in rapid loss of most of the cylinder contents.

Other rupture tests on 48-in. cylinders have been and are to be directed toward evaluation of handling damage and of corrosion damage resulting from inadequate storage practices. Continuation of the test program will allow for further evaluation of these features to develop a data base for special handling requirements for defective cylinders identified during normal enrichment operations and during periodic inspections of the ever-increasing number of stored cylinders of depleted UF₆. Design efforts directed at solution of the apparent dilemma of the stiffening ring weld will also be supported by the rupture test program.

OVERPACK EVALUATION AND IMPROVEMENT

Uranium hexafluoride enriched in the fissionable isotope is transported in protective shipping packages, the DOT Specification 21 PF-1 overpacks, which are designed to protect the contents against all foreseeable shipping accidents. The packages are filled with an insulating foam to protect the cylinder contents from significant heating in a 30-min, 1475°F fire exposure. The structural integrity of the overall package and the dampening properties of the foam give additional protection to the contained cylinder against damaging impact loads that can result from transport or other mishaps.

The packages have been in service for periods of up to 20 years, and it has been apparent for some time that the owners' handling, maintenance, and repair practices vary widely. In some cases, the packages accumulate moisture in the insulating foam during long service periods, and this moisture accumulation promotes rusting that degrades the structural integrity. Deficiencies in some of the features of the package design have also become apparent during use. A group of packages rejected for further service by the K-25 toll enrichment operation (and concurred with by the package owners) has been subjected to destructive sampling to assess the level of corrosion damage and to measure the moisture content of the insulating foam. Drying procedures were developed in cooperation with a package manufacturer, thermal conductivity tests were run on the moisture-laden insulation, and repair standards were developed. These observations, along with an improved package design, were presented at a fall 1989 conference at Portsmouth.

The development of standards for inspection and repair of existing packages is a continuing effort that involves interaction with package manufacturers and owners. In addition to the continuing work on packaging standards and design, an effort is being made to improve the handling and transport of these packages, including both trailers and tie-downs.

The severe rusting that instigated the renovation efforts on the steel-covered 21 PF-1 overpacks also prompted the design and construction of a group of stainless steel packages. Some of these packages have been in service for as long as 10 years without problems. It has recently been observed, however, that some of the newer stainless steel packages are showing evidence of pitting damage on both inner and outer surfaces. Both users and fabricators of these packages are currently investigating service histories, fabrication records, and component sources in efforts to identify the source of the damage.

NEW PROJECTS FOR IMPROVED STORAGE

The program for improving storage and the assurance of containment is under a continuous process of modification and improvement, as new concerns and new understandings develop. In addition to the program elements already outlined, several more have been assessed, planned, or suggested for implementation as manpower and funding become available.

- A study of the stress loadings associated with the normal activities of cylinder filling, handling, and stacking is being conducted. It proposes that several cylinders be fitted with accelerometers and strain gages that will be monitored during cylinder transport and placement in storage. The data will be useful in refining stacking procedures to minimize the potential for damage. Preliminary analysis of early data from the study indicates that the stresses resulting from handling during filling and cooling are very low.
- Nondestructive evaluation techniques will be examined for possible application to in-place assessment of containment integrity. Under consideration are acoustic techniques for detection of leaks, ultrasonic or vibration measurements for determination of fill level or distribution, and remote visualization techniques for corrosion evaluation of cylinder surfaces near the ground.
- Cylinders identified as approaching or already past the 0.250-in. limiting wall thickness are to be segregated for painting to arrest further corrosion. This identification will be made primarily during restacking operations to effect full visual access for the baseline and periodic inspection requirements. Schedules for these activities have not been established.
- Cursory sampling of areas surrounding a cylinder storage yard, as well as sampling associated with breached cylinders, has not suggested any problems with dispersal of materials from stored UF₆. This program should be expanded to give quantitative and statistically meaningful data to support the integrity of long-term storage. No resources have been allocated to this effort.

- Compilation and maintenance of a file on all applicable regulations, procedures and other documentation for handling, packaging, transport, and storage of UF_6 is seen as a potentially useful tool for storage management. Source materials would include engineering standards and procedures, Department of Energy (DOE) orders; ANSI standards; Environmental Protection Agency, DOT, Nuclear Regulatory Commission, and other federal regulations; publications of international standards organizations; and any other sources of pertinent information. No funds have been allocated.

SUMMARY

The need for formal planning and surveillance of depleted UF_6 storage has grown with the number and age of the containers involved. Approximately 55,000 steel cylinders are currently in use for storage of natural, and enriched UF_6 , with the oldest storage cylinders approaching 40 years of service. The cylinders are supported on wood or concrete saddles (bottom tier) in double-high rows on paved concrete or compacted gravel surfaces.

Observations and tests over nearly 20 years have shown that the probable cylinder life, as determined from measurements of atmospheric rusting, is 70 years or more, based on the minimum allowable thickness of 1/4 in. for a 100-psi storage cylinder whose initial wall thickness is 5/16 in. Other factors in the storage environment, however, may shorten this life expectancy through accelerated localized attack, and programs directed to assurance of safe containment and storage for the required time have been established to identify and correct these situations. Among these factors the principal sources of damage are the pitting attack that occurs as a result of ground contact and the head thinning in rust drift areas of skirted cylinders. Other areas of accelerated attack include the restricted spaces covered by the support saddles and by the cylinder nameplate. Other identified containment issues are leaking valves and/or plugs and leaks initiated from handling and stacking damage.

An integrated program of activities related to containment and storage has been established under the aegis of the Energy Systems Interplant Operating Committee. The program includes oversight of objectives and reports of activities and progress in projects that have as their goal the advancement of safety and integrity of long-term storage of UF_6 . The program is directed and administered by the Energy Systems UF_6 Containment and Storage Committee, with representatives from the two operating plants, the K-25 Site and the Energy Systems Enrichment Technical Operations Division. Projects have begun throughout the enrichment complex to increase the size of storage yards to alleviate overcrowded conditions and facilitate needed inspections, renovate and upgrade storage yard surfaces, improve handling and stacking of cylinders, and implement uniform procedures and practices used in storage of UF_6 . These projects are supported by evaluations and tests to determine the stresses resulting from normal handling and stacking operations, by the steel corrosion test programs now under way at the three storage sites, by the cylinder rupture test program for evaluation of designs and characteristic defects, and by three-plant activities relating to upgrading of the DOT Specification 21PF-1 protective overpack. The committee is also involved in the development of enhanced inspection criteria for added assurance of continued safe storage and containment of depleted UF_6 .

DISCUSSION

Most of the cylinders now in storage are not properly arrayed to permit the levels of monitoring necessary to assure continued containment integrity, and a priority need for such monitoring is the restacking of many of these cylinders. This need in itself presents a problem in containment integrity because a small but significant hazard in operation of a storage facility is that which comes from cylinder handling activities (Fig. 15). Bumping contacts during stacking or unstacking can, under a limited set of circumstances, generate small cracks that extend through the cylinder walls, thus setting the stage for leakage of the cylinder contents. If these cracks are not detected at inception, they can grow through corrosion of the steel by the action of HF that is generated by reaction of moist air with the cylinder contents. Handling accidents can also result in bending (and consequent leakage) or breaking of valves. Neither situation gives leaks of catastrophic dimensions, since the solid contents have a rather low vapor pressure. Both types tend to be self-healing, but they release small amounts of uranium compounds and HF from the cylinder.

Valves can be changed in the field with proper equipment and controls, provided that the location is accessible. Leaking cylinders, however, must be taken to transfer facilities, and this may require unstacking of other cylinders, perhaps 100 or more, to provide access, if the leak is not detected at its inception. Each cylinder movement also presents a small added risk of damage to the cylinder or its valve. A test program is being developed to evaluate the stresses resulting from the normal range of impact contacts because of cylinder stacking. The results of this program will furnish guidance in the development of improved handling procedures. In addition, the current stacking procedures require better control of cylinder spacing and provide for an immediate post-placement inspection for handling damage. The probability of undetected leaks from stacking accidents is thus greatly reduced from that of earlier experience.

In addition to restacking cylinders to facilitate a monitoring and inspection program for proper assurance of containment, a priority need of the storage program involves the development of quality assurance procedures that must precede disposition of a number of nonstandard cylinders. This group of irregular cylinders includes an unknown number that were filled to higher limits than presently specified, a few thousand noncode-stamped and noncertified volume cylinders from early phases of the enrichment/storage program, and a few hundred containers built in-house from surplus converter parts. The noncode and nonvolumated cylinders should be identified for special handling at the time of final disposal. The "overfilled" cylinders could represent a substantial safety hazard for any program involving emptying cylinders and should be handled as a special group, for which procedures must be developed before final disposal. The 19T converter shell cylinders cannot be accommodated in present autoclaves and will require special handling, special

procedures, and possibly special equipment for disposal. The 12.8T cylinders, however, are of a size that can be handled in the present containment autoclaves.

Containment and storage issues have historically been the subject of standards development and regulatory attention on local, state, and national levels within the nuclear fuel and power industry and in international channels. One of the earliest efforts at dissemination of information to the fuel industry produced ORO-651, currently in its 6th revision and now titled *Uranium Hexafluoride: A Manual of Good Handling Practices*. This document, in worldwide distribution since first issued in 1966, is regarded as the standard guide to handling and packaging of UF_6 . Diffusion plant personnel were also prime movers in the adoption of ANSI standards for shipping and storage cylinders, for trailers for highway transport of UF_6 , and for tiedowns for securing shipping packages to transport trailers.

A comprehensive program of UF_6 cylinder testing and protective packaging development was initiated at the K-25 Plant in the mid-1960s,⁶⁻⁹ and this work is continuing¹⁰ at the request of DOE. There is also significant interest in this work from both domestic and international communities and their respective regulatory agencies. International interest in the storage of UF_6 was prompted by "Monitoring of Corrosion in ORGDP Cylinder Yards," a paper presented at the 1988 Oak Ridge conference on Uranium Hexafluoride—Safe Handling, Processing, and Transporting.¹¹ Helen Henson, who presented the paper, is serving as an invited consultant to the International Atomic Energy Agency in preparation of a chapter on UF_6 storage for an agency guidebook to be issued in 1992.¹² Further interest in UF_6 cylinder corrosion studies was also apparent during the PATRAM '89 international symposium in Washington, D.C.¹³ Five papers covering containment and storage activities at the DOE diffusion enrichment plants were presented at this conference.

A second Oak Ridge conference on UF_6 handling devoted most of two sessions, 10 papers, to storage issues.¹⁴

A continuing and growing interest and concern in matters relating to UF_6 containment and storage is apparent both from historical experience within the project and from recent developments and current trends in environmental awareness in legislative and regulatory activities. An integrated program is thus seen as necessary to maintain each of the activities in proper perspective, to ensure continuing progress in the achievement of the goals seen as necessary for proper disposition of a valuable energy resource, to maintain compliance with applicable regulatory directives, and to maintain and enhance the operation's community image. The program is based on inspection and continued monitoring of cylinders in storage, and includes auxiliary projects to do the following:

- improve inspection and surveillance of the stored cylinders,¹⁵
- assess the consequences of the observed degrees of handling and storage damage or deterioration on containment integrity and safety, and
- arrest or correct the deterioration that has resulted from past storage practices.

The objectives of the present storage program are to ensure that each cylinder is situated to permit effective full visual inspection and to identify and correct those situations that interfere with this objective. Further, all cylinder placements will be monitored during and immediately following stacking operations to detect and correct the results of damaging impacts at the earliest possible time. Through such proactive efforts, the primary causes of shortened storage life and loss of containment integrity (damage during placement and accelerated corrosion resulting from extended time of wetness) can be eliminated and the maximum available storage life realized for all cylinders.

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Appendix
PHOTOGRAPHS

FIGURE 1

CYLINDER STORAGE AT OAK RIDGE K-1066-E

Approximately 5,000 large cylinders containing UF_6 are stored in three separate areas at the Oak Ridge K-25 Site, with about 2,000 of them at this location. The storage yards are of compacted gravel (left) and concrete (right).

FIGURE 2

STORAGE CYLINDERS AT PORTSMOUTH

The depleted UF_6 from the Portsmouth Gaseous Diffusion Plant is stored in a single concrete-paved facility. There are more than 12,000 large cylinders in long-term storage, and about 1,000 new cylinders are added each year.

FIGURE 3

DEPLETED UF_6 IN STORAGE AT PADUCAH

Most of the depleted UF_6 from the Paducah Gaseous Diffusion Plant is stored here. There are about 35,000 cylinders of depleted, enriched, and feed materials stored in 14 separately accessible yards. Thirteen of these have compacted-gravel surfaces.

FIGURE 4

TYPICAL 1-IN. UF₆ VALVE

This valve, used on all feed, transport, and storage cylinders, was designed specifically for UF₆ service. The one-piece body and the location of the stem thread minimize the risk of breakage due to external forces, as does the inherent ductility of the single-phase aluminum bronze from which the valve is constructed. Few problems have been encountered in the 40 years this design has been in production.

FIGURE 5

RAYGO-WAGNER CYLINDER STACKER

FIGURE 6

STRADDLE CARRIER FOR IN-PLANT CYLINDER HANDLING

FIGURE 7

SCALE ACCUMULATION RESULTING FROM GROUND CONTACT

The continuous presence of moisture at the cylinder surface when the cylinder is resting on or near the ground results in greatly accelerated corrosion. The development of a heavy layer of scale indicates a corresponding reduction in wall thickness.

FIGURE 8

PITTING ALONG LINE OF GROUND CONTACT

Pitting corrosion results from protracted direct contact with the ground because of high levels of moisture. Wall thickness levels of 0.20 in. have been observed in several such cylinders, and this thinning mechanism is suspected to be the major cause of two recently discovered breaches of containment in storage cylinders.

FIGURE 9

LEAKAGE VERIFICATION/MONITORING ON STORAGE CYLINDER

Leaking is indicated by uranium deposits (yellow-green) on the exterior surfaces of the valve. The Teflon bag will allow capture and identification of uranium salts for verification of leakage, which can then be corrected by tightening or replacing the valve. The bag can also furnish a container for a more sensitive HF detector to verify an active leak.

FIGURE 10

RUST ACCUMULATION IN SKIRTED CYLINDER

The rust buildup trapped in the skirt retains moisture for long periods of time, causing accelerated corrosion of the skirt and adjacent area of the cylinder head.

FIGURE 11

BREACHED CYLINDER IN K-25 SITE STORAGE

The location of the hole suggests impact with another cylinder during stacking operations.

FIGURE 12

CORROSION TEST COUPONS AT K-25 SITE

Similar arrays of coupons are deployed at both the Paducah and the Portsmouth storage locations. The tests are as given by ASTM G-50, and rate determinations are to be made from both thickness and weight-loss measurements after exposure times of 1 to 16 years.

FIGURE 13

TEST COATINGS ON STORAGE CYLINDERS

Organic zinc (white) and inorganic zinc (gray) coatings are to be compared with the currently specified enamel coat (green) in their ability to protect the steel cylinders in storage service. All of the coatings were applied over the manufacturer's recommended primer on a grit-blasted surface.

FIGURE 14

RESULT OF BURST TEST ON MODEL 48T CYLINDER

Rupture testing has verified the design safety factors on all cylinder types in use in the U.S. uranium enrichment operations. Failure in large cylinders with external stiffening rings, however, was found to initiate at the stiffening ring butt weld because of incomplete penetration. As the weld quality at this location improves, the size of the rupture becomes larger, with a greater potential for material loss.

FIGURE 15

HANDLING DAMAGE IN CYLINDER STACKING

The channel-type stiffener on the Model O cylinders was particularly susceptible to damage in bumping contacts with adjacent cylinder lifting lugs. This opens to corrosion an area of the cylinder wall that cannot be checked for thickness.

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