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INVESTIGATION OF BREACHED DEPLETED UF_6 CYLINDERS

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ABSTRACT

In June 1990, during a three-site inspection of cylinders being used for long-term storage of solid depleted UF_6, two 14-ton cylinders at Portsmouth, Ohio, were discovered with holes in the barrel section of the cylinders. An investigation team was immediately formed to determine the cause of the failures and their impact on future storage procedures and to recommend corrective actions.

Subsequent investigation showed that the failures most probably resulted from mechanical damage that occurred at the time that the cylinders had been placed in the storage yard. In both cylinders evidence pointed to the impact of a lifting lug of an adjacent cylinder near the front stiffening ring, where deflection of the cylinder could occur only by tearing the cylinder. The impacts appear to have punctured the cylinders and thereby set up corrosion processes that greatly extended the openings in the wall and obliterated the original crack. Fortunately, the reaction products formed by this process were relatively protective and prevented any large-scale loss of uranium. The main factors that precipitated the failures were inadequate spacing between cylinders and deviations in the orientations of lifting lugs from their intended horizontal position.

After reviewing the causes and effects of the failures, the team's principal recommendation for remedial action concerned improved cylinder handling and inspection procedures. Design modifications and supplementary mechanical tests were also recommended to improve the cylinder containment integrity during the stacking operation.

INTRODUCTION

Depleted uranium hexafluoride (UF_6) produced at the uranium enrichment gaseous diffusion plants has been stored in steel cylinders since the diffusion plants first went into production more than 40 years ago. As the number of cylinders became significant, each of the three diffusion plants created outdoor storage yards as long-term depleted UF_6 repositories. These yards consist of concrete or compacted gravel storage pads, with lower cylinders positioned on wooden or concrete saddles and a second level of cylinders supported by the lower level. In June 1990, during inspections at Portsmouth, Ohio, two steel cylinders, each containing 14 tons of depleted UF_6, were discovered with holes in the barrel section of the cylinders. The holes in both cylinders appeared to have originated very close to the stiffening ring nearest the valve end. An investigating team was appointed and, in association with Portsmouth personnel, developed action plans to permit detailed examinations of the cylinders and their contents, as well as an assessment of the environmental impact of the incidents on the storage yard and neighboring cylinders. Evidence was gathered through the process of removing the failed cylinders from the storage yard, checking their internal

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pressure, analyzing gas and salt samples, emptying one cylinder, and sampling internal reaction products from the other cylinder. Details of the procedures followed in these examinations and related problems are described in a companion paper presented at this Conference (1). Another companion paper (2) presents the chemical findings uncovered in these examinations and their general implications for continued cylinder storage. The present paper discusses the failure cause and the associated lessons learned that can be applied to improvements in cylinder design and cylinder storage practices. Members of the investigation team who were instrumental in the failure analysis consisted of Energy Systems personnel (Jack DeVan, Chairman, John Barker, John Googin, Tim Butler, and Mike Taylor) and Department of Energy representatives Bob Dyer and Joe Russell. An interim final report covering their investigation was issued in September 1991 (3).

INVESTIGATION STRATEGY

The lesser damaged of the two cylinders was investigated first, since it was hoped that the initial cause of failure would be less obscured by subsequent exposure of the cylinder contents to the ambient atmosphere. To access this cylinder, over 200 stacked cylinders had to be moved with the Raygo Wagner loader. Several nearby cylinders were observed to be dented at, or near, the location where both failures had occurred. These other cylinders were examined for cracks; however, none was found. Once access had been gained, the cylinder was pressure checked and a gas sample taken for chemical analysis. As would be the case for an undamaged cylinder, the internal pressure was still less than atmospheric (-4 psig at 85°F), and the gas analysis showed negligible oxygen or nitrogen contamination. The cylinder was then patched, removed from the storage yard, and valved at ambient temperature into the cascade. After removal of the UF_6, the breached area was X-rayed and the internal cylinder surface in the area of the hole was imaged in the X-705 annex using a miniature video camera mounted on a flexible cable. Presently, this cylinder is in storage awaiting sufficient radioactive decay to allow removal and evaluation of the material still plugging the hole and metallographic examination of the cylinder wall.

To clear a path to the cylinder with the larger hole, 60 neighboring cylinders had to be moved. The pressure in this cylinder was above atmospheric (+8 psig at 85°F) and contained elevated concentrations of HF. The hole was covered by an aluminum patch, and core samples of the salt underlying the hole were taken through two ports in the patch for chemical analyses. The cylinder was weighed for accountability and then valved into the cascade. Evacuation was begun in the same manner as for the cylinder with the smaller hole and was continuing at the time of this report.

DESCRIPTION OF FAILURES

The larger of the holes found in the two breached cylinders at Portsmouth is pictured in Fig. 1. The area of salt exposed by the breach was approximately 9 in. along the axis of the cylinder and 13 in. from top to bottom. Viewed from the valve end, the hole was centered at the 9 o'clock position and extended under the nearest stiffening ring. The upper corner of the lifting lug of the adjacent cylinder was positioned at the approximate center of the hole. A green-colored, jagged, fluoride salt layer covered the exposed hole area slightly below the original solid UF_6 surface, and a similarly colored layer of salt covered areas below the hole and on the underlying cylinder. Some loose, green salt deposits were also found on the top of the underlying cylinder and on the concrete pad directly under it. Removal of the salt deposit from the underlying cylinder revealed a series of vein-like grooves etched into the cylinder surface. The grooves were relatively widely spaced and shallow near the top of the cylinder but converged into a much deeper groove over the right-hand quadrant of the cylinder, where it reached a maximum depth of .13 in. None of the other neighboring cylinders showed any evidence of corrosion.

The smaller of the two holes is pictured in Fig. 2. The hole lies at the 3 o'clock position when the cylinder is viewed from the valve end. Like the larger hole, it was positioned at a point close to the front stiffening ring and again was located directly under the top corner of the lifting lug of an adjacent cylinder. The upper circular section of the hole (Fig. 2) was approximately 2 in. in diameter and was concentric with the juxtaposing lifting lug corner. A lower tear drop section extended approximately 2 in. below the

Fig. 1. Appearance of large hole. Width is 9 in. and length is 13 in.

Fig. 2. Appearance of smaller hole. Width is 2 in. and length is 4 in.

circular section. Green salt deposits covered the lifting lug and completely obscured the hole until the lug was pulled away. (Because the green salt deposit closely matched the color of the paint covering the cylinder, very careful inspection was required to spot the hole when viewed from the front of the cylinder). There were no salt deposits on the underside of this cylinder and only a small green spot (about the size of a dime) on the concrete pad directly under the hole. Although examinations of neighboring cylinders showed no effects directly resulting from this failure, several of the neighboring cylinders did show minor mechanical damage attributable to improper spacing (overcrowding) and misalignment of lifting lugs.

PROBABLE FAILURE CAUSES

The team's analysis indicated that the smaller of the two holes had been initiated by a through-wall crack that occurred at the time of stacking. The crack resulted from the impingement of the upper corner of the lifting lug of an adjacent cylinder, which deformed the shell of the subject cylinder in the immediate vicinity of the front stiffening ring. Because of the restraint afforded by the 1-in.-thick stiffening ring, this deformation led to tearing of the shell along the fusion line of the weld joining the shell to the stiffening ring. Conditions contributing to the puncture were: (1) an insufficient separation distance between the cylinders, which resulted from too tight a spacing of the wooden saddles under the cylinders, and (2) the offending lifting lug being placed at a position of 7 o'clock rather than the desired 9 o'clock position. Although the original breach had been largely obliterated by the extensive wastage of the shell associated with the exposure of UF_6 to air and moisture (Fig. 2), evidence for the initial failure cause was afforded by examinations of neighboring cylinders, which indicated a relatively high incidence of dents caused by the lifting lugs of adjacent cylinders. In most cases, these dents were sufficiently far away from an unyielding stiffening ring that they could be accommodated by dimpling of the shell rather than tearing. The small-hole cylinder sustained such a dent at the end opposite from where the hole occurred. Although the degree of indentation here appeared to be of the same order as that which occurred at the failed end, the indent was not restrained by the stiffening ring and there was no indication of cracking in this location. In the few instances where lugs were found to have impinged much closer to the stiffening ring, the degree of indentation was insufficient to have caused failure. However, in these cases the indents in fact appeared as small tears.

Other support for the stated failure mode comes from drop tests performed on similarly designed cylinders at Paducah. A consistent observation in these tests has been the susceptibility of the cylinder shell to tearing when it was impacted next to the stiffening ring (4). The tests also showed that the 5/16-in. A516 steel plate used in the manufacture of the small-hole cylinder was particularly prone to lamellar tearing along the fusion line of the stiffening ring weld. Failures by this mode were already on record for at least two other cylinders made from this particular grade of steel. The first occurred in March 1978 when a cylinder was accidentally dropped onto a wooden saddle and the second, in March 1982, by an indeterminate cause during shipping on a railroad flatcar. It was shown by Blue (4) that sulfide inclusions in this steel adversely affected its impact properties transverse to the rolling direction — a problem that is exacerbated by welding, which concentrates the inclusions along the weld fusion line. Based on these findings, the allowable sulfur content was reduced in the specifications for all cylinders subsequently built with A516 steel components.

In common with the smaller hole, the larger hole is located in the region of a stiffening ring with the corner of the lifting lug of the adjacent cylinder again positioned near the center of the hole. Again, the spacing between the failed and adjacent cylinder was less than the desired minimum. However, unlike the small-hole cylinder, physical measurements at the outer circumference of the large hole showed that a small gap had actually existed between the corner of the opposing lifting lug and the original outer circumference of this cylinder. Thus, while the positions of the two failures, with respect to the stiffening ring and lifting lugs, are quite similar and point to a common failure mechanism, any impingement of the lifting lug on the large-hole cylinder must have occurred before, not after the cylinder reached its rest position. This caveat seems reasonable given the stacking situation for this cylinder. Unlike the small-hole cylinder, which was at ground level and supported by wooden saddles, the large-hole cylinder was in the second tier and was therefore positioned in a crease formed by two underlying cylinders. If either this cylinder or the one opposing it had been lowered and released into a position that was slightly away from the lowest point of the crease, a rolling motion would be set up as the cylinder moved to its rest point. Given the tight spacing, only a slight movement past the rest point would be required to make contact with the adjacent cylinder and, because of the relatively close position of the lifting lug to the stiffening ring, to initiate the type of tearing fracture discussed above. Operators at Portsmouth, who have experience in stacking these cylinders, confirm that difficulties in visual perception and in steadying the cylinders make this sequence of events entirely plausible.

The shell of the larger-hole cylinder was fabricated of a different grade of steel than the smaller-hole cylinder. The former was made of A285 and the latter of grade A516. A change from A285 to A516 occurred shortly before the smaller-hole cylinder was built because the fracture toughness properties of the latter grade were nominally better at low temperatures (≤0°C). However, given the probable temperature of the cylinder and its contents at the time of stacking, the A285 cylinder should not have been any more vulnerable to fracture than one constructed of A516. In fact, based on drop tests of cylinders built with the A285 grade, this material has even better toughness properties than the particular A516 heat used in constructing the small-hole cylinder. Nevertheless, experience during shipping and handling of these cylinders has shown that tearing near the stiffening rings can occur with even the best of these materials.

As discussed in a companion paper (2), corrosion played a major role in these failures once the cylinders had been breached. However, the investigation team could find no evidence that corrosion might have contributed to the initial break in the wall. Wall thickness measurements were performed at numerous points around both failed cylinders, and there were no indications of wall thinning at any point other than in the immediate vicinity of the holes. Even at crevice regions, which are the most susceptible to atmospheric corrosion, there were no indications of localized attack or pitting.

ENVIRONMENTAL IMPACT

Analysis of an air sample taken near the large hole showed approximately 1 ppm HF. Contamination surveys of the concrete pad in the vicinity of the large-hole cylinder showed negligible contamination, except in the area directly under the hole and along a short track where water runoff had deposited traces of UF_4. This area was successfully decontaminated without removing any of the pad surface. Contamination of cylinder surfaces in the immediate vicinity of the leaking cylinder was minor except where there were visible traces of UF_4. Surfaces of four neighboring cylinders (in addition to the failed cylinder) required decontamination, which again was completed using conventional swabbing techniques. Before decontaminating the area around the cylinder, approximately 15 lbs of UF_4-containing salt were collected from the pad under the cylinder and as loose deposits on the cylinder directly under the failed one. In the case of the small-hole cylinder, contamination surveys showed negligible contamination, except in the immediate vicinity of the hole, and smears of the pad under the cylinder showed negligible alpha/beta readings.

One of the soil samples checked when preparing the roadway to access the large-hole cylinder indicated contamination above background levels. Several samples were then surveyed from soil debris left by road construction. One sample had a maximum activity of 12 picocuries/gram, and the rest were below 8 picocuries/gram. (A level of 30 picocuries/gram is permitted for landfill disposal.) The source of this activity appeared to be water runoff from the cylinder, which crossed the concrete pad and entered the ground at the edge of the pad where the new road attached.

An analysis of the chemical processes accompanying the formation and growth of the cylinder holes was used to estimate the mass of UF_6 that could have escaped the cylinders as unrecovered material. Lower bounds were determined from comparisons of the pre- and post-failure cylinder weights together with recovered solid and gaseous products. Upper bounds were estimated from a corrosion model (2) that predicted the reaction paths for UF_6 and the masses of uranium reaction products not necessarily accounted for in the weight measurements. The unrecovered mass of UF_6 from the longer-term failure (13 years) was estimated to be between 17 and 109 lbs. In the case of the shorter-term failure (4 years), the unrecovered UF_6 was below measurable limits but possibly amounted to 4 lbs.

IMPLICATIONS OF FAILURES ON PRESENT STORAGE POLICIES

Cylinder Design Faults - Based on the overall failure statistics for the 14-ton, depleted UF_6 cylinders, the mechanical design has proved generally reliable and well suited to the required filling and handling operations. Nevertheless, an obvious design change that could reduce the probability for the type of failures observed here is to round the upper corner of the lifting lug. Other modifications that would divert impacts away from the area of the stiffening ring weld include welding a band around the outside diameter of the stiffening ring or devising removable shields (bumpers) that can be placed on the lifting lugs during stacking. These failures also point to a need for qualification testing that would assess mechanical properties in the context of storage requirements, where external pressures acting on the cylinders need to be addressed as well as internal pressure. Paducah personnel responsible for qualification testing of the cylinders have indicated (5) that mechanical tests could be included in their test program to evaluate the effects of external loads typical of lifting lug impacts, particularly forces required to penetrate the cylinders. Without such tests, there is currently no methodology for recommending design changes to the shell that would make it more resistant to handling mishaps, such a as sharp impacts, or to movements induced by ground tremors or shifting between cylinders.

An issue related to cylinder design is the selection of the material to be used as the pressure boundary. The selection process for the steel for these cylinders has been very exacting, with input from a relatively long history of acceptance tests and a large service data base. The A516 grade currently specified for the 14-ton cylinders was adopted because minimum toughness properties could be included in the acceptance test requirements for this grade. It replaced the A285 grade, which had no toughness requirements as part of the purchase specification. Although both grades are considered acceptable in terms of their toughness properties at the expected cylinder service temperatures, A516 is obviously preferred for quality assurance reasons. Metallurgical tests at Paducah (5) have indicated that the heat of A516 steel used to fabricate the smaller-hole cylinder probably contained segregated sulfide inclusions, which may have increased the risk for the type of damage (tearing) that ultimately occurred. However, current specifications for this steel call for a lower sulfur level to avoid the sulfur segregation problems, so this issue is not relevant to cylinders being manufactured today. Furthermore, the failure in the larger-hole cylinder, which is made of A285 steel, indicates that, even without sulfide inclusions, tearing can result from an impact near the stiffening ring. Thus, the failures observed here appear endemic to the cylinder design and cannot be tied to insufficiencies in the types of steels specified.

Integrity of Neighboring Cylinders - The heat of A516 steel used in the manufacture of the smaller-hole cylinder has been shown to be more vulnerable to a tearing failure along the stiffening ring weld than steels used before or since. It is particularly important that other cylinders made from this same heat of A516 be fully inspected and kept under surveillance if evidence of mechanical damage near the stiffening ring is found. Furthermore, in the unstacking of cylinders to gain access to the subject failed cylinders at Portsmouth, cylinders were identified that have dents near the stiffening ring weld. Although subsequent non-destructive examinations of these cylinders showed no detectable cracks and they, therefore, were restacked, the affected cylinders should be kept under surveillance and periodically inspected. The same requirement should be extended to cylinders for which mechanical defects have been noted or which are obviously vulnerable to impingement by a neighboring cylinder.

The cylinder directly below the large-hole cylinder was corroded by

HF-containing reaction products to a maximum depth of 0.13 in. Although the resultant loss of wall thickness posed no problems in moving the cylinder, the thickness was below that allowed for liquid feeding back to the diffusion plant, and warm feeding will have to be used to empty this cylinder. Based on the experience here, it must be accepted that failure of a neighboring cylinder could be induced by breached cylinders, provided that the neighboring cylinder is in the line of movement of HF-containing reaction products. Such an induced failure could have potentially serious consequences if: (1) the damaged area was so extensive that it precluded patching or moving the cylinder or (2) it formed near the top of the cylinder where there was very little solid UF_6 available to act as a plug to prevent the spread of contamination. The latter scenario reinforces the importance of early detection of failures of the type incurred here and emphasizes the need for inventorying and maintaining surveillance on any damaged cylinders currently in storage.

Potential Accident Scenarios and Remedial Actions - Until the present failures, the primary concern with the existing thin-wall cylinder design for long-term storage was corrosion. This stemmed from the assumption that, once the cylinder had been transported to the cylinder yard and stored, corrosion should be the only factor to reduce mechanical integrity. These failures indicate that mechanical factors can have an influence on cylinder lifetime even after stacking. One problem stems from mechanical damage induced at the tail end of the stacking sequence, which has remained undetected by prevailing inspection procedures. Another problem stems from damage that can occur even after the cylinders are stacked ,due to one cylinder resting on another or the shifting of one cylinder relative to another. These failures also emphasize the vulnerability of the cylinders to impact damage by projectiles and jostling from earthquakes. Inspections of the cylinder storage yards at all three diffusion plants, conducted in the course of this failure analysis, indicated that the sequence of events leading to these two failures has a relatively low probability of occurrence and does not reflect a generic problem. Nevertheless, some of the circumstances contributing to the failures, such as insufficient spacing between adjacent cylinders and improperly oriented lifting lugs, are common among several of the storage sites. Thus, the possibility that additional cylinders may have sustained damage by the same means, while considered quite small, cannot be completely ruled out because not all cylinders can currently be viewed in their entirety. In some yard locations at all three sites, cylinder rows are stacked too closely for a thorough visual inspection. Because of the effective plugging by the reaction products of the two exposed areas, the contamination from any unidentified cylinders damaged from the same cause should be minimal as was the case with these two failures. Nevertheless, a concerted effort is needed to identify and empty such cylinders.

The information gained from the present failures provides significant insight into the effects of cylinder failures that occur below the solid-vapor interface of the cylinder. The environmental effects associated with the smaller-hole cylinder failure (which went undetected for 4 years) were minimal, and the cylinder was moved and emptied with little difficulty. Even in the case of the larger hole, formed over a period of 13 years, the environmental insult was minimal as it existed on the site at the time of discovery. Unfortunately, the present failures do not allow a full assessment of breaches which occur nearer the top of the cylinder, i.e., in the vapor space. Scenarios have been proposed in which extreme conditions could lead to a steam-driven expulsion of contaminated liquid and HF from the cylinder, although this situation appears unlikely if the breaches are small, i.e., detected in a reasonable time span(≤1 to 2 years). In any case, an experimental effort should be directed at examining the long-term effects of a hole nearer the top of the cylinder with water introduced under controlled conditions.

To counter the causes and effects of the present failures, the most important remedical action involves improved handling and inspection procedures. Routine inspections of all stockpiled cylinders can effectively limit the impact of potential future failures by a similar cause. Inspections of the cylinders at the time of stacking should be instituted to verify that no mechanical defects exist and that the spacing between cylinders is adequate. Where inspections reveal that mechanical defects exist, or that determinations of such defects are precluded because of poor access to the suspect areas, such situations should be noted and covered by surveillance procedures.

CONCLUSIONS AND RECOMMENDATIONS

An examination of the two failed cylinders, together with inspections of neighboring cylinders, indicated that the failures most probably resulted from mechanical damage that occurred at the time that the cylinders had been placed in the storage yard. In both cylinders evidence pointed to the impact of a lifting lug of an adjacent cylinder near the front stiffening ring, where deflection of the cylinder could occur only by tearing the cylinder (the exact force to fracture a thin-wall cylinder is unknown). The impacts appear to have punctured the cylinders and thereby set up corrosion processes that greatly extended the openings in the wall and obliterated the original crack. Fortunately, the reaction products formed by this process proved to be relatively protective and prevented any large-scale environmental insult or loss of uranium.

Factors that facilitated the failures were inadequate spacing between cylinders and deviations in the orientations of lifting lugs from their intended horizontal position. In both failure cases, neighboring cylinders were crowded and lugs were misaligned to the point where dents due to impinging lugs could be found. Thus, although the current cylinder design is particularly vulnerable to punctures near the stiffening rings, the present failures stem more from handling and stacking procedures than from design inadequacies. However, design changes that could be beneficial involve rounding the lifting lug profile and modifying the present paint scheme to a color that contrasts with that of UF_4. An assessment of the failures experienced here indicates that the associated environmental impact can be greatly reduced by a more rigorous visual inspection during the stacking operation and routine inspections thereafter. In the four years that elapsed in forming the smaller hole, there was no measurable quantity of unrecoverable uranium lost. With adequate inspection and monitoring, both of these incidents most likely would have been detected early enough to have avoided any significant impact on the environment or loss of inventory. However, the inherent difficulty of visually inspecting stacked cylinders for dents and possible cracks will require a high level of awareness training of the personnel responsible for inspecting the cylinders.

The team's survey of the cylinder yards at the three diffusion plants showed that the factors that led to the failures were not unique to the Portsmouth site. The frequency of these factors at other sites appeared less. Yet, the possibility that a similar accident sequence could have occurred for other cylinders now in storage cannot be completely ruled out, and further efforts will be required at all three sites to examine those cylinders whose spacing has prevented a complete visual inspection.

In view of the size of the holes produced and the length of time that corrosion processes operated (13 years in one case), these failures approach the "worst-case" scenario of what can accrue when a storage cylinder is breached in an area backed up by solid UF_6. Fortunately, the effects appear to have been minimal in terms of the inventory of uranium and fluorine lost to the environment and the overall physical damage to the cylinders. However, it is not known whether a failure closer to the top of the cylinder (i.e., in the vapor space) would have resulted in the same environmental effects, since the reaction products formed may not be as protective as those formed in the present failures. Accordingly, a study of the effects of a break into the gas space of the cylinder will be required before the maximum credible incident associated with the mechanical failure during storage can be absolutely defined.

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