Life Extension Program for the Modular Caustic Side Solvent Extraction Unit at Savannah River Site – 13179

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ABSTRACT

Caustic Side Solvent Extraction (CSSX) is currently used at the U.S. Department of Energy (DOE) Savannah River Site (SRS) for removal of cesium from the high-level salt-wastes stored in underground tanks. Currently, the Actinide Removal Process (ARP) and the CSSX process are deployed in the (ARP)/Modular CSSX Unit (MCU), to process salt waste for permanent disposition. The CSSX technology utilizes a multi-component organic solvent and annular centrifugal contactors to extract cesium from alkaline salt waste. The original plant was permitted for a three year design life; however, given the successful operation of the plant, a life extension program was completed to continue operations. The program included detailed engineering analyses of the life-expectancy of passive and active components, resulting in component replacement and/or maintenance and monitoring program improvements. The program also included a review of the operations and resulted in a series of operational improvements. Since the improvements have been made, an accelerated processing rate has been demonstrated. In addition, plans for instituting a next-generation solvent are in place and will enhance the decontamination factors.

INRODUCTION

The Savannah River Site liquid waste system consists of several facilities to safely receive and store radioactive waste, treat, and permanently dispose waste. The large underground storage tanks and associated equipment, such as evaporators to minimize liquid storage and maximize tank space, are known as the F-Area and H-Area Tank Farms. The tank farms include a complex interconnected transfer system which includes underground transfer pipelines, diversion boxes, and pump pits to direct the flow of waste. The waste in the tanks may be classified into two broad general categories, high heat waste (HHW) and low heat waste (LHW), which are defined by their rate of heat generation. The majority of the HHW are direct byproducts of the primary separations processes which were historically used. The majority of the LHW are also byproducts of these processes, however, other processes and facilities such as resin regeneration, decontamination, and laboratories also contribute significant quantities of LHW. Both HHW and LHW are present in three forms: supernatant, sludge, and salt. The supernatant is a multicomponent aqueous mixture, while sludge is a gel-like substance which consists of insoluble solids and entrapped supernatant. Salt contains salt crystals which form after the evaporated supernatant is cooled, and entrapped interstitial supernatants. The waste from these tanks is retrieved and treated as sludge or salt. Once the waste is retrieved and processed, the tanks are prepared for closure. Closure consists of removing the bulk of the waste, chemical cleaning, heel removal, stabilizing remaining residuals with tailored grout formulations and severing/sealing external penetrations.

SALT PROCESSING

Salt waste at the SRS is currently processed through the Actinide Removal Process in concert with the Modular Caustic Side Solvent Extraction Processing (ARP/MCU) [1]. A summary of the process is shown in Figure 1.

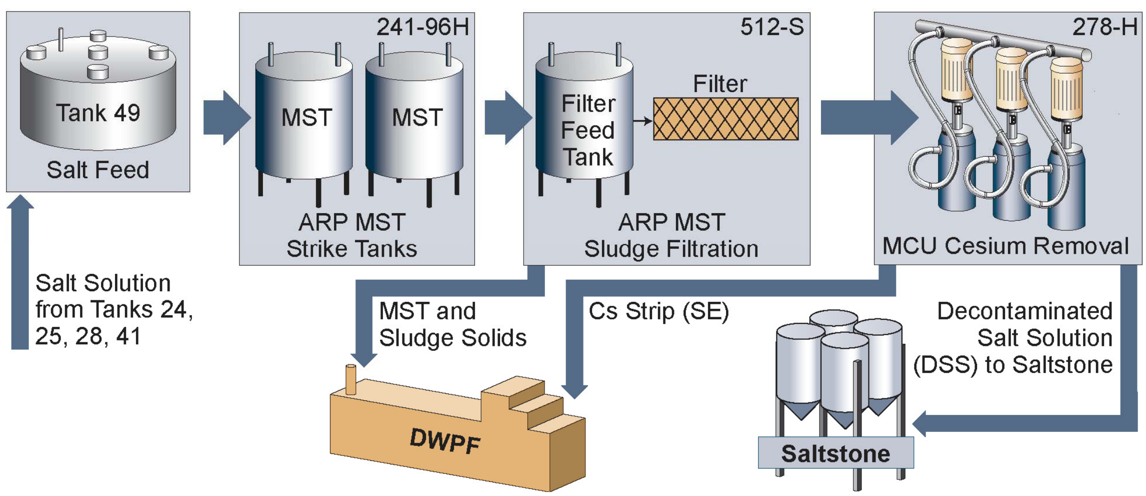

Figure 1: Schematic of the ARP/MCU Process

The ARP decontaminates low-curie salt solution via adsorption of strontium-90 (Sr-90), actinide radionuclides, and entrained sludge solids in the salt solution onto MonoSodium Titanate (MST) followed by filtration through a cross-flow filter. The actinide, Sr-90, and MST laden sludge waste streams are transferred to Defense Waste Processing Facility (DWPF) for vitrification and the remaining clarified salt solution is transferred to the MCU process. The MCU process extracts Cs from the clarified salt solution using caustic side solvent extraction chemistry. The low Cs-137/low actinide Decontaminated Salt Solution (DSS) is subsequently grouted through the saltstone production facility (SPF), and the strip effluent (SE) solution of cesium nitrate from the CSSX process is transferred to DWPF for vitrification.

DEMONSTRATED PERFORMANCE

The ARP/MCU process was validated during the initial first months of operation. As a first-of-a-kind nuclear operation, ARP/MCU was started up after a series of reviews to validate readiness. Operations proceeded very deliberately to ensure initial performance per the safety basis and operational requirements. “Sample and hold” requirements were enforced to ensure that all acceptance criteria would be met until confidence was established in facility baseline performance and to ensure collection of key operational data. The salt processing proceeds via ‘salt batches’ that are ‘qualified’ for the process as well as for immobilization and disposition.

Thus far, the ARP/MCU has processed four salt batches and is currently processing salt batch 5. ARP/MCU processed an average of 87,000 liters per month of Salt Batch 1 with a peak performance of 245,000 liters per month yielding a total of 538,000 liters of salt waste processed for Salt Batch 1. Improvements based upon experience and lessons learned from Salt Batch 1 reduced risk and increased productivity with Salt Batch 2 processing. Enhancements and improvements in process chemistry, process efficiency, and overall attainment/throughput resulted in record processing rates (on two occasions) of more than 150,000 liters within one week during 2009/2010 Salt Batch 2 processing. In addition, the ARP/MCU process has performed better than expectations for decontamination factor requirements for Cesium 137 and solvent carryover performance. These factors are summarized in Table 1 for the first three sludge batches processed.

Actions taken since startup of ARP/MCU in 2008 have demonstrated an increased processing rate from the original design of approximately 3,790,000 liters per year to approximately 5,300,000 liters per year. Enhancements and improvements include in-tank chemistry adjustments, reduced cycle-times, redesign and replacement of the secondary filter at the cross-flow filtration facility. A 12hr strike time was implemented at ARP, along with a 23 L/min processing rate increase at MCU which reduced the overall processing cycle-time.

Table 1: Summary of Select ARP/MCU Performance Parameters

	Expected	Actual Salt Batch 1 (SB-1) 4/2008 – 9/2008	Actual Salt Batch 2 (SB-2) 1/2009 - 5/2010	Actual Salt Batch 3 (SB-3) 6/2010 – 9/2011	Total
Cs-137 DF	>12	100-350	100-450	100-400	
Sr-90 DF	54.4	>190	~205	N/A	
Pu DF	14.4	>100	~139	N/A	
Liters Processed		~538,000	~2,760,000	~2,590,000	~5.9M Liters
Curies Processed		~40000	~180000	~180000	~400K curies

Although the overall attainment has significantly improved from SB-1 to SB-2, equipment reliability issues limited the overall attainment hence overall throughput. The majority of the affected equipment has been repaired, replaced, and or redesigned commensurate with the failure causes identified. Efforts continue to improve equipment reliability, reducing unexpected downtime to improve overall attainment. In addition to equipment and processing upgrades, alternative system planning is being done to more efficiently qualify subsequent salt batches to reduce downtime between batches.

PROCESS IMPROVEMENTS

The ARP/MCU process is carefully monitored and process parameters are continuously monitored to ensure reliable operations. In addition, the process parameters were used to identify opportunities to increase the throughput of the facility. The data revealed specific improvements to increase throughput and reliable operations of the facility. The improvements included:

- Improvements to the ARP filtration cycle to support a MCU processing rate of 7.6 M liters per year, such as:
    - Installation of an improved ARP secondary filter with increased solids handling capacity.
    - Improved temperature controls increasing overall ARP filtration rate.
    - Improved process shutdown controls to minimize solids formation.
- Implementation of an ARP/MCU blend tank capability reducing the time for salt batch makeup outages by 5 weeks. This configuration has been deployed and proven for Salt Batch 3.
- Upgrades to shielded sampler system, eliminating the potential for sample cross contamination and thereby reducing costly re-sampling evolutions.
- Improvements to pre-filter installation and coalescer cleaning reducing the accumulation of solids, reducing coalescer pressure differential, and extending media life.
- Improvements to solvent quality controls ensuring process performance and eliminating process interruptions resulting from degraded solvent.
- Automation of control systems to be installed, improving MCU startup consistency and reducing shutdown conditions.

- Improvements to the durability of the pumps.

The combination of the completed improvements, planned improvements, and the increasing process confidence commensurate with more experience provide the basis for achieving a throughput of 7.6 M liters /year.

LIFE-EXTENSION

Processing approximately 370 million liters of salt solution from the Tank Farms is planned over the life of the program. The ARP/MCU process was constructed and permitted initially for a three-year service period, bridging the crucial period before the startup of the Salt Waste Processing Facility (SWPF). However, given the delays to SWPF startup, extension of the ARP/MCU facility is necessary to continue processing salt waste. The life extension program was comprehensive review of the systems, structures, and components culminating in the implementation of recommendations for the review that ensured the facility could safely continue operations. The originally planned service life of ARP / MCU, which was three years of operation, was reached in 2012. A detailed engineering analysis identified the key parameters necessary for life extension. Most of the individual components were determined to be capable of a longer operating life than three years. An evaluation concluded that ARP/MCU can operate until 2015 before major equipment failure is expected. The passive process components such as piping and structures were determined to have service life in excess of 2025.

Actions to address ARP/MCU life extension beyond the three-year period included:

- Implement process and equipment upgrades and improvements.
- Evaluate and procure spare parts.
- Adjust preventive maintenance.
- Increase equipment performance monitoring.
- Obtain appropriate regulatory approvals.

ENGINEERING EVALUATION

The detailed engineering evaluation included the structures, systems, and components (SSC's) that comprise the operations of ARP/MCU. The evaluation considered active and passive components in the distinct facilities that make up the ARP/MCU process. Active components are consumable and degrade in performance over time. They include motors, pump rotating assemblies and seals, actuators, and instruments in that nature. Passive components are not considered consumable; however, they will degrade over time. Passive components include cell coatings, building structures, tanks and vessels, piping, pump casings, heat exchangers, and etc. The evaluation considered the degradation of these components as a function of degradation mechanisms, e.g. radiation exposures. The distinct facilities considered were: (1) ARP; (2) Filter; and (3) MCU.

ARP

The ARP consists primarily of the MST strike tanks and the associated equipment, as shown in

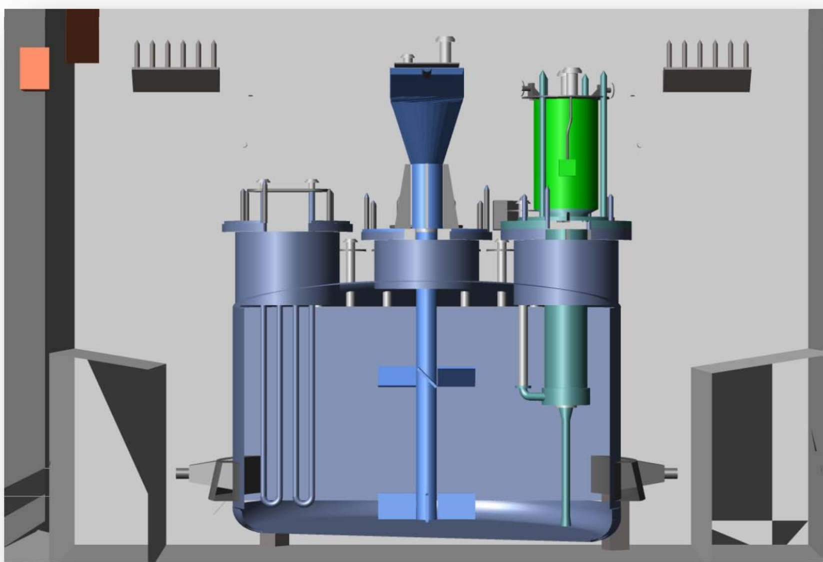

Figure 2: Typical MST STrike Tank

Figure 2. The process components of the ARP were evaluated against the expected radiation exposure. Over 25 years, estimated absorbed dose for the equipment (non-wetted) would be 0.0496 MGray (MGy) in the process cells, outside of the tanks. The agitator motors, agitator lubricant, wiring and other equipment would remain operational, unless a mechanical failure occurred. Equipment located inside the 241-96H process cells (wetted) will receive a maximum total dose of 0.0731 MGy. Motor Operated Valves are only wetted during waste transfers. The estimated dose of 0.0731 MGy is derived by assuming the Motor Operated Valves are wetted 5 percent of their operational life. Equipment inside the tanks will receive a total dose of 0.52 MGy

over 25 years. It is expected tank, pump, and agitator materials of construction can absorb this dose without degradation. The tank agitators' blades have an in-tank radiological exposure which is estimated to be 0.52 MGy [2]. However, these metal components are nearly immune to this level of radiological exposure. There may be erosion of these blades over many years of service; however the slow rotational speed of the agitator blades mitigates this potential. As such, the current maintenance practices were to be continued to ensure reliable operations. In addition, the systems at ARP are included in a system performance monitoring/system health monitoring program. These programs monitor system degradation and viability. Reductions in system performance will be tracked and recommendations will be made on restoring the system's viability.

Cross-Flow Filter

Chemical exposure of the cross-flow filter system wetted parts is also similar to that of ARP. Over 25 years, equipment in similar locations (in tank, out tank wetted and outside tank non-wetted) would receive radiation doses similar to that of equipment in the ARP. The cross-flow filter facility was designed with a 25 year life. The existing maintenance strategy was sufficient to maintain the 25 year life of the facility. However, improvements to the filtration process were outlined previously. The active systems at 512-S are included in a system performance monitoring/system health monitoring program. System engineers will monitor system degradation and viability. Reductions in system performance will continue to be tracked and recommendations will be made on restoring the system's viability.

MCU

A summary of the MCU process and the associated vessels are shown in Figure 3 [2].

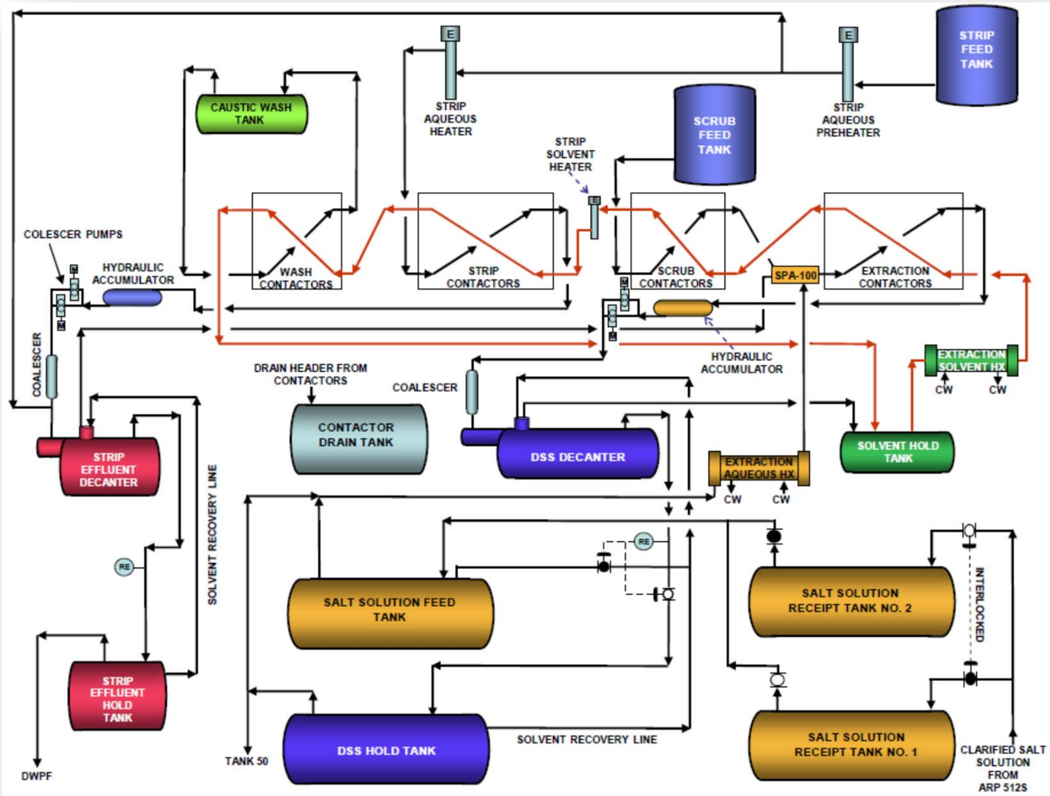

Figure 3: MCU Process Schematic

The clarified salt solution from ARP is received in the MCU Salt Solution Receipt Tanks and these supply the Salt Solution Feed Tank that feeds the Extraction Contactors. CSSX Solvent is supplied to the Extraction Contactors from the Solvent Hold Tank. Cesium is extracted from the clarified salt solution by the CSSX solvent in the Extraction Contactor Bank resulting in decontaminated salt solution (DSS) and highly contaminated solvent. Since the solvent used for the extraction process is combustible and can give off combustible vapors under certain conditions, it cannot be sent to downstream processing facilities without serious safety concerns. Consequently, the DSS from the Extraction Contactor Bank is decanted in the DSS Decanter to remove as much residual solvent as possible before it is placed in the DSS Hold Tank. The DSS Hold Tank is the staging tank for transfers in preparation for grouting. The decanted solvent is returned to the Solvent Hold Tank for reuse.

The remainder of the CSSX process is primarily the generation of the strip effluent for DWPF processing and solvent recovery. The cesium (Cs+) bearing solvent from the extraction contactors is first scrubbed in the Scrub Contactor Bank with dilute nitric acid (0.05M) to remove impurities that could impact the Cs+ stripping process. The acidic solution containing the contaminants from the scrub contactors is recycled back to the Extraction Contactor Bank inlet line to mix with the incoming salt solution while the solvent is discharged to the Strip Contactor Bank. In the Strip Contactor Bank, the Cs+ is removed from the solvent and becomes part of the strip acid solution called strip effluent (SE).

The SE is sent to the SE Coalescer and Decanter, via an SE Hydraulic Accumulator and SE Coalescer Pump, to remove residual solvent and then the acidic SE is discharged to the SE Hold Tank. The SE is eventually transferred to DWPF where it is combined with sludge and vitrified. The residual solvent captured in the SE Decanter is recycled to the Extraction Contactor Bank via a special piping assembly (SPA-100). The decontaminated solvent from the Strip Contactor Bank is washed with caustic in the Wash Contactor Bank to neutralize the solvent pH and removes solvent degradation products before being recycled to the Solvent Hold Tank to start the process all over again.

In both process effluent (DSS and SE) hold tanks there is a potential for residual solvent to come out of solution and form a solvent layer in the hold tanks while the solution is stored. For this reason, both the DSS and SE Hold Tanks have solvent recovery lines that enable the recycle of carried-over solvent back to the process.

Materials Exposures and Evaluations

Based upon the process flow paths and the materials fed, the numerous vessels are exposed to radiation and chemicals, including internal and external tank components. Internal tank components include submersible pumps and their power cables. Other components include flow elements, radiation sensors, electrical power and instrument cables, air operated valves, and vertical shaft pumps. The MCU process also includes contactors.

Motors, Pumps, and Valves

The motors on these pumps are located outside the tanks but inside the process cells. The dose rates for these components can vary, depending on the activity level of material being handled in the process vessel; e.g., salt solution activity is 0.29 Ci/L; strip solution activity is 4.35 Ci/L. The Decontaminated Salt Solution (DSS) and Caustic Wash Streams have low radiation exposure rates. Due to the low rates for these process streams, it is possible for equipment in or near this process stream to not experience any radiological or chemical related failures. The Contactor Drain Tank (CDT), Solvent Hold Tank (SHT), Salt Solution Receipt Tank (SSRT), Salt Solution Feed Tank (SSFT) streams have moderate to high radiation exposure rates. There may be radiation-related failures to equipment located in, or near, these streams. The Strip Effluent (SE) stream would be exposed to high radiation rates, due to waste concentration potentially as high as

4.35 Ci/L. Motors that are designed to operate in a harsh radiation environment, such as that found in nuclear power plants, are qualified for total exposures in the range of 0.01 to 2 MGy. Since not all pumps were procured with a radiation hardened motor, a dose of 0.37 MGy was assumed to result in motor failure. SEHT motors and wiring, near the SE Hold Tank, would receive this dose by 2015, SSRT motors and wiring in the SSRT by 2021, and CDT, DSS Decanter, DSSHT, SSFT and SHT motors and wiring would not reach 0.37 MGy through 2025. SSRT wiring outside the SSRT is not expected to exceed 0.37 MGy through 2025.

Motors installed at MCU were supplied with high quality poly-urea grease in sealed bearings. Most bearings were expected to last 5-6 years of continuous service (2012-2013). Taking into account two pumps being employed one in use at a time, bearings may last until 2017 to 2019 before radiological effects are taken into account. This type of bearing has a radiation resistance of 0.10 MGy to 1 MGy. The Strip Effluent Hold Tank Pump Motor bearings would receive a 0.50 MGy dose by 2015 (Intermittent Operation). The Strip Effluent Coalescer Pump Motor bearings would receive a 0.50 MGy dose by 2015 (Continuous operation). The Decontaminated Salt Solution Hold Tank pumps are required approximately 50 percent of the time. Other process pumps are limited in use to approximately 1 to 2 hours per day.

It was estimated that between 2015 and 2017, The Strip Effluent Hold Tank Pump Motor, Strip Effluent coalescer Pump Motor, and Contactor bearings would have failed.

Contactor bearings would have been at risk of failures beginning 2014 as they have no alternate and run when the process operates. Bearings in the Strip Contactors would have been at increased risk of even shorter life due to their higher radiological exposure. Bearing grease would have oxidized faster in these areas resulting in poor lubrication. Metal fatigue failures in motor bearings are not likely in intermittent use pumps and motors. However, bearing metal fatigue failure is possible in those motor bearings running continually.

The Salt Solution Feed and Receipt process streams experience moderate radiation exposure. The pair of submersible pump motors that is located inside each Salt Solution Receipt Tank (SSRT) is potentially susceptible to radiation damage. Each motor receives a gamma dose of 3.89 Gy per hour based on an activity level of 0.29 Ci/L in the salt solution. This equates to a dose of 0.0341 MGy per year. Exposure of components in wetted contact with the feed solution would reach 0.23 MGy through 2017. Non-wetted components will experience approximately 0.12 MGy through 2025. It is possible that the SSRT pump elastomer seal might fail by 2013 due to radiation damage, and subsequently at 3 year intervals with 0.29 Ci/L feed material. There is also the risk of SSRT motor bearing failure as early as 2013, due to operating time and radiation dose. Salt Solution Feed Tank B Pump experienced problems in January 2009 and failed in May 2009.

The Contactor Drain Tank and Solvent Hold Tank wetted equipment will experience moderate to high dose rates. These areas would have received a cumulative dose of 0.10 MGy through 2010 and 1 MGy through 2019. SHT pump B and CDT Pump B experienced failures in February 2009. These failures were not attributed to process chemistry. Solvent Hold Tanks flow elements are also at risk from radiological damage.

The Strip Effluent process stream will see the high radiological exposures. The Strip Effluent process stream may experience up to 4.35 Ci/L at the maximum feed value of 0.29 Ci/L. However, the design maximum feed value of up to 0.29 Ci/L may not be realized. Pumps and equipment in contact with the Strip Effluent stream would have begun to experience radiation induced failures as early as 2012. During 2013, valves and equipment in intermittent contact with the Strip Effluent stream could experience radiation doses exceeding 1 MGy. Motors and equipment (i.e. Strip Effluent Hold Tanks flow elements and Strip Effluent Decanter radiation sensors) near the SEHT stream could reach 1 MGy exposure in 2022.

Most valves in the process cell are expected to not fail by 2025 based on assumed radiation rates. The failure threshold for MCU valves was assumed to be 1 MGy. Most valves are self -draining and only in contact with waste intermittently. CDT valves that are constantly immersed may reach the 1 MGy threshold by 2020. SHT valves may be exposed to 1 MGy by 2019. All other valves are not expected to reach the 1 MGy threshold through 2025.

Contactors

The contactor system is subdivided into several system segments: extraction contactors, scrub contactors, strip contactors, caustic wash contactors, and the caustic wash tank. The contactor skid was determined by the vendor to be capable of 5 years of operation without any maintenance being performed.

Each set of contactors make up a system segment that includes component groups such as drive motors with speed and vibration monitoring equipment. Components in the contactor package consist of those placed inside the contactor enclosure that are exposed to high radiation and components located outside the contactor enclosure that are exposed to normal background radiation. The highest component dose rates are found in locations inside the enclosure building where solution with an activity as high as 4.35 Ci/L flows through the strip contactor. A dose rate calculation for seals on the connector ends of the flexible hoses shows that seals in contact with this solution would receive 0.10 MGy, which is the nominal service life of Kalrez gasket material, by 2021. This is a conservative assumption.

Contactor bearing was a limiting factor as well. Assuming 75 percent utility, contactor bearings should have lasted 5-6 years, not having taken radiological degradation into account. However, radiological exposure would have shortened the bearing life further. Strip contactors were especially at increased risk of failure due to the elevated radiation rates in these contactors. It was estimated that the SEHT motors would receive a cumulative dose of 0.37 MGy, the assumed motor failure dose, by 2014.

LIFE EXTENSION MODIFICATIONS

Based upon the evaluations as outlined above, many of the components were replaced. These included Solvent Hold Tank Pumps, Aqueous DSS Decanter Pumps, DSS Hold Tank Pumps, DSS Coalescer Pumps, and Process Enclosure Sump Pump. Contactor bearings have been replaced as well. As ARP/MCU continues to operate, equipment monitoring will increase the ability to predict impending equipment failure. This will allow repairs or replacements to be scheduled during planned outages, reducing system downtime.

CONCLUSIONS

The ARP/MCU plant was permitted for a three year design life, however, given the successful operation of the plant, a life extension program was completed to continue operations. The program included detailed engineering analyses of the life-expectancy of passive and active components, resulting in component replacement and/or maintenance and monitoring program improvements. The program also included a review of the operations and resulted in a series of operational improvements. Since the improvements have been made, an accelerated processing rate has been demonstrated. In addition, plans for instituting a next-generation solvent are in place and will enhance the decontamination factors.

REFERENCES

[1] R. A. LEONARD, 2002, *Caustic-Side Solvent Extraction Flowsheet for Optimized Solvent*, ANL.
[2] P.E. Carroll, 2009, *ARP/MCU Life Extension Evaluation*, U-ESR-H-00084, Rev. 0.