Sodium Separation from Remote-Handled Transuranic Waste via Distillation – 14479

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ABSTRACT

Sodium distillation has been selected as the principal method to remove the sodium from the RH-TRU non-combustible waste destined for WIPP disposal. Previous remote-handled waste sodium distillation systems have been installed and operated within an inert gas hot cell. The hot cell available for this system is an atmospheric hot cell with high surface contamination levels and limited floor space. A suitable siting was located in the basement of the Idaho Nuclear Technology and Engineering Center (INTEC) Fluorinel Dissolution Process (FDP) facility in a shielded room below the hot cell. This room has a 56-cm-diameter tube running vertically from the floor of the hot cell where waste is repackaged, down three stories, and exiting through the ceiling into a shielded room outside the boundary of the hot cell. This room provides a non-radiologically-contaminated area to place the equipment, and has sufficient shielding to protect the surrounding areas from high radiation fields. However, because the shielded room has no manipulators a remote closure device had to be developed to seal the vessel to achieve sufficient vacuum to perform distillation. Previous hot cell distillation systems used manipulators to bolt a gasketed blind flange closure on the distillation vessel. The basic design of the system and the numerous lessons learned and design improvements developed during off-Site development of the system is discussed.

INTRODUCTION

At the INL Site in Idaho, USA, elemental sodium was used as a coolant in experimental reactor test loops. Subsequently, the waste generated by disassembling these highly radioactive reactor test loops is contaminated with residual and bulk sodium. The disposition of sodium-contaminated remote-handled radiological hazardous waste presents a unique and challenging problem. The sodium is distributed within heat exchangers, pipes, pumps, and other debris waste forms. The waste has been assigned RCRA waste numbers of D001 (ignitable) and D003 (reactive) due to the sodium within the debris. The waste targeted for this system is RH-TRU and destined for WIPP, which will not accept waste with these hazardous waste numbers. In order to remove the D001 and D003 hazardous waste numbers from the debris, the sodium must be removed or treated utilizing a permitted treatment process.

DESCRIPTION

The sodium distillation system (SDS) is a vacuum distillation system designed to remove sodium contamination from highly radioactive, solid, non-combustible RH-TRU waste (see Fig. 1). The system consists of a distillation vessel, condenser, collection vessel, cold trap, vacuum pump, transfer vessel, and associated instrumentation, controls, valves, and fittings. The distillation process is monitored via pressure and temperature sensors. System process pressures are measured in two locations, upstream of the condenser, between the distillation vessel and condenser, and downstream of the condenser, near the vacuum pump. These two pressures are used to measure the pressure drop across the condenser, which occurs when sodium vapor enters the condenser and condenses from a vapor to a liquid. The valve body is filled with an inert gas (argon) purge to prevent any in-leakage of air past the primary seal into the distillation vessel during distillation. This will prevent any formation of sodium oxides in the distillation system that was initially present during early commissioning activities.

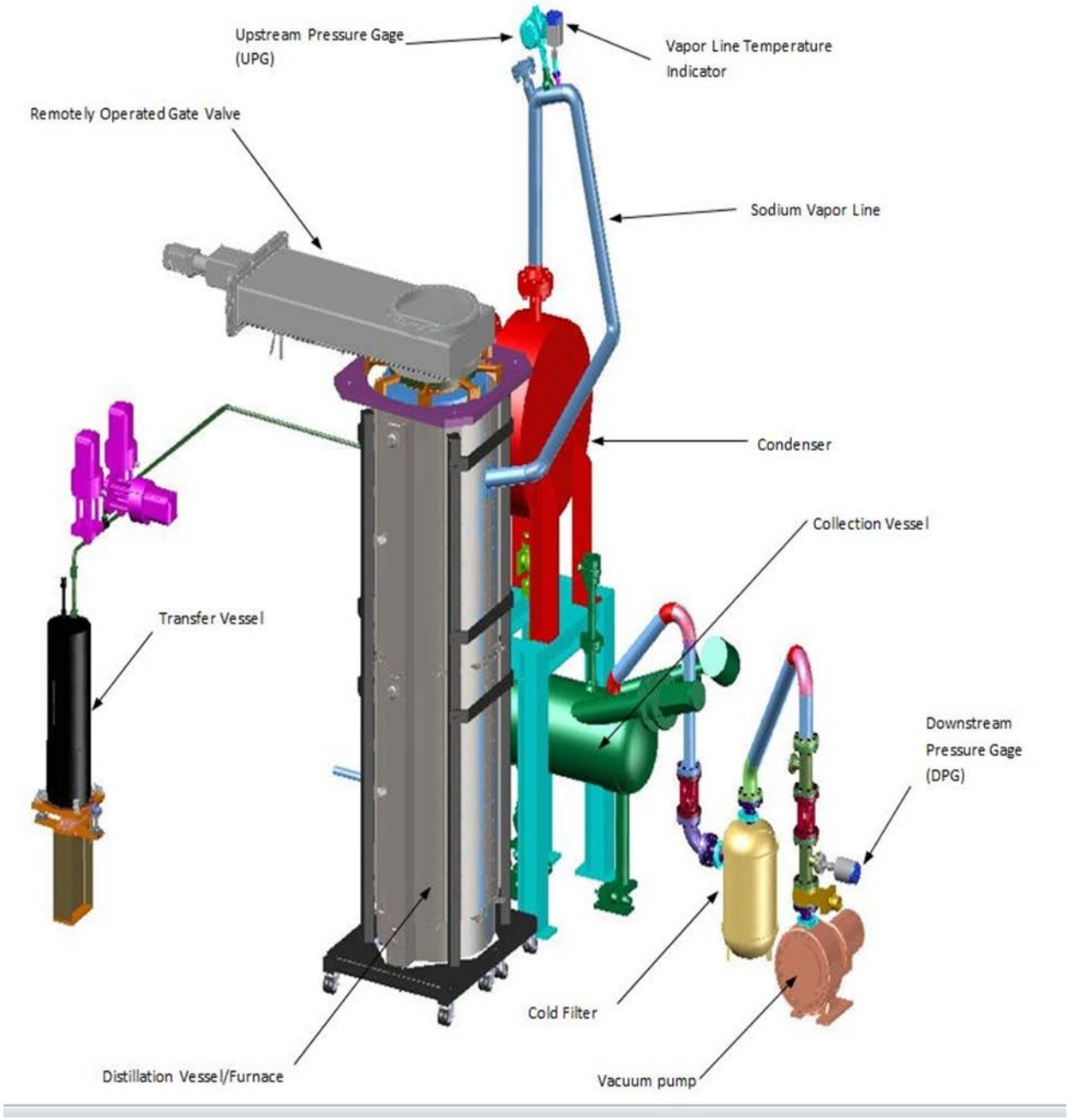

Fig. 1. Idaho Cleanup Project sodium distillation system.

To measure the vapor temperature, a thermal well with a temperature probe was installed at the top of the 5-cm vapor line. The vapor temperature provides an additional means to monitor the presence of sodium vapor in the line when boiling is occurring in the distillation vessel. The system has been proven to distill sodium across a broad range of conditions, from 379°C at 27 Pa to 538°C at 1,067 Pa Dual process data monitoring (pressure and temperature) provides clear indications of when sodium distillation is in progress and the completion of the distillation process across this range of operating parameters (see Fig. 2). These process parameters can be used together for confirmation but are a reliable indication of end state on their own.

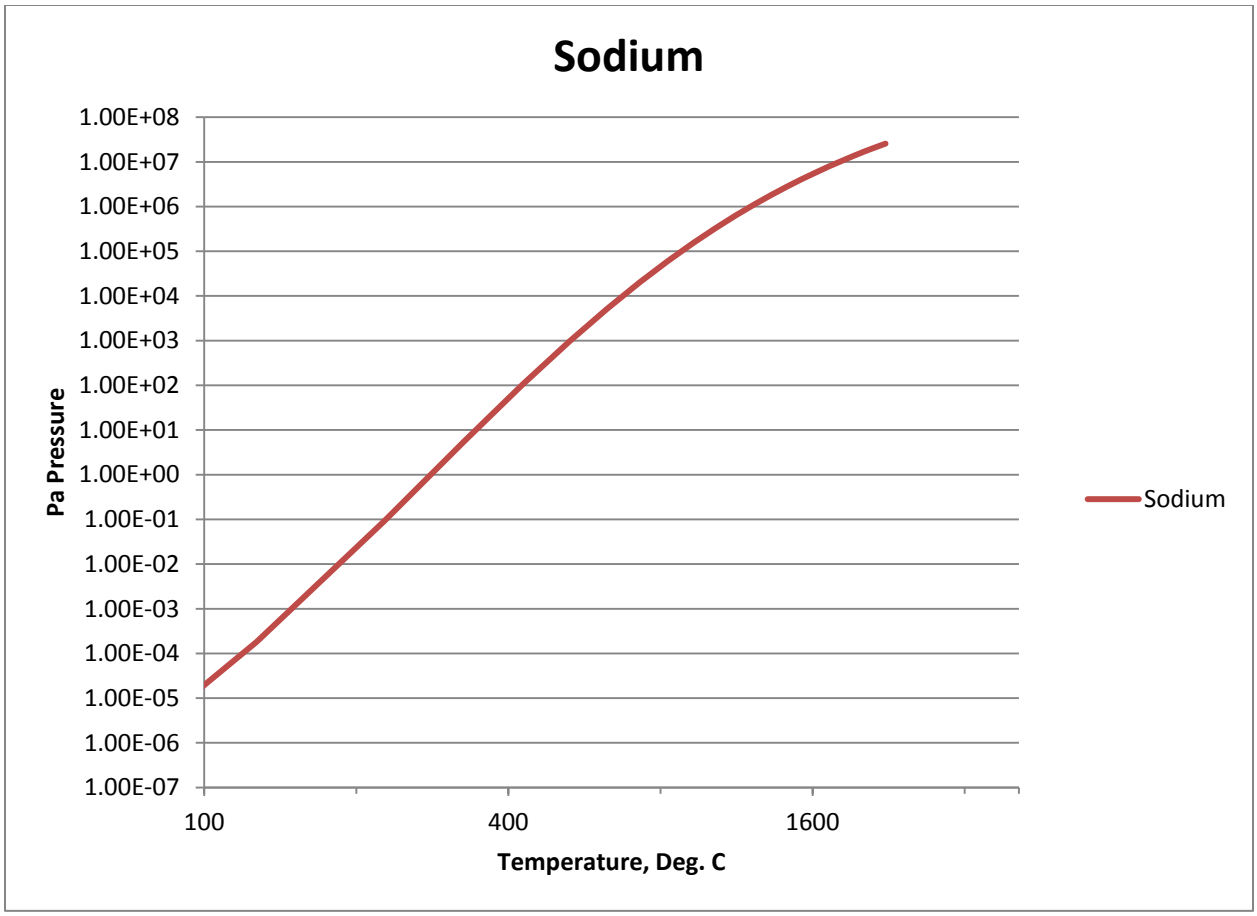

Fig. 2. [Sodium Vapor Pressure Curve].

The remote-handled sodium-contaminated waste is unpackaged from storage containers within a shielded enclosure (hot cell), examined to ensure that sodium vapor has an exit path, and placed in a specially fabricated distillation waste container. This waste container is then lowered through a tube from the hot cell and directly into the distillation vessel in another shielded room in which the SDS has been installed.

To start the distillation system, the distillation vessel's remote closure top is sealed and the vessel's furnace top is lowered onto the top of the distillation vessel. The vacuum pump is then started, and all heat trace and vessel furnace elements are started. When the sodium in the system starts to boil the sodium vapor generated in the distillation vessel generates enough pressure to drive itself through the system piping to the condenser, which, at 149°C, cools the sodium vapor sufficiently to condense it into a liquid but not enough to allow it to freeze into a solid. The sodium vapor pressure generated in the distillation vessel by boiling the sodium in the high temperatures of the furnace and low vapor pressures in the condenser generated by cooling the vapor into a liquid provide the motive force to move the sodium from the waste and into the sodium condenser. The condenser temperature is controlled at 149°C by a thermal fluid system using silicone fluid as the heat transfer medium. Silicone oil was chosen as the coolant fluid for this application because it is non-reactive with sodium and non-toxic in the event of an accidental leak or release.

In the condenser, sodium vapor condenses into a liquid and flows down into the collection vessel. The collection vessel is a small horizontal tank placed below the sodium condenser, which provides volume

for the system to collect and store sodium during and between distillation cycles. The collection vessel is fitted with band heaters capable of melting solid sodium that has been accumulated during distillation operations to allow the sodium to be transferred to the transfer vessel whenever necessary. Any vapors generated during the distillation process, which escape the condenser, will be driven downstream and captured by the cooler temperatures and lower vapor pressures in the filter/vacuum trap. The filter, or vacuum trap, is located downstream of the collection vessel and upstream of the vacuum pump. The filter is an uninsulated steel vessel that contains filter elements where any remaining condensable vapors are filtered out and collected. Because the filter is un-insulated, it remains cooler than any upstream portion of the distillation system. The filter will also filter out any remaining sodium vapors in the off-gas stream that get past the condenser and collection vessel. The filter elements are replaceable and are expected to be replaced as necessary.

The parameters monitored during the process provide considerable insight as to the state of the distillation process throughout the system. These parameters are recorded and graphed on a screen during distillation and the data has proven very accurate at providing the system operators with the information they need to determine when sodium distillation starts, is progressing, and is complete. Process parameters that are monitored are the pressure in the vapor line (upstream of the condenser, upstream pressure gauge [PG]), the pressure in the collection vessel outlet (downstream of the condenser, downstream pressure gauge [DPG]), and temperature in the vapor line. In order to understand why the data appear with all of the various peaks and fluctuations, it is beneficial to understand what is occurring within the system as it is under vacuum, heating up.

Typical Distillation Pressure Trend

Organic materials, such as oils, residual cleaning fluids, small quantities of residual tape, and silicon, are anticipated to be present in the waste in addition to the sodium contamination. During initial pump-down of the system to establish a vacuum, some liquid compounds may evaporate, and, during heat up, the remaining items will volatize and will pass through the system without condensing in the condenser due to the relatively high temperature within the condenser. These events will be observed as parallel pulses of pressure on both the upstream pressure gauge (UPG) and downstream pressure gauge (DPG). These are considered non-condensable compounds and, since they do not change state, no pressure drop is observed across the condenser.

During a sodium distillation event, sodium vapor expands in the distillation vessel and the sodium vapor line and generates an increase in pressure in these locations which is observed in the UPG. Because sodium condenses in the condenser, a corresponding pressure increase is not then observed in the DPG. This increase in the pressure differential between the pre-condenser, UPG, and the post condenser DPG is a primary indicator that sodium distillation is in progress. In addition, as sodium vapor passes past the vapor line thermo well, a temperature increase is observed in the sodium vapor line.

During sodium distillation, other vapor state compounds are transported with the sodium vapor through the condenser. Because these compounds do not condense in the system, both the UPG and DPG record pressure pulses which fluctuate as the compounds pass through the system. This is apparent in the data when pressure pulses appear across both the UPG and the DPG (see Fig. 3). Within these fluctuations, an increase in the pressure differential between the UPG and the DPG indicates that sodium is condensing in the condenser, resulting in an increase in the differential pressure between the two gauges.

It is worth noting that previous SDS designs relied primarily on temperature indications in the vapor line for distillation process monitoring. The design team devised an innovative approach to monitoring the pressure within the vapor line. A small tube (1.3-cm diameter) was branched off of the top of the 5-cm vapor line between the distillation vessel and the condenser. This line was extended upward at a 45-

degree angle for a length of 1.5-m. This line was heat-traced to 149°C to ensure that any vapor entering the line would condense to liquid sodium, but not freeze into a solid, and run back into the condenser. In this manner, the face of the pressure detectors could be kept from coming into contact with sodium vapor while still monitoring the pressure in the vapor line.

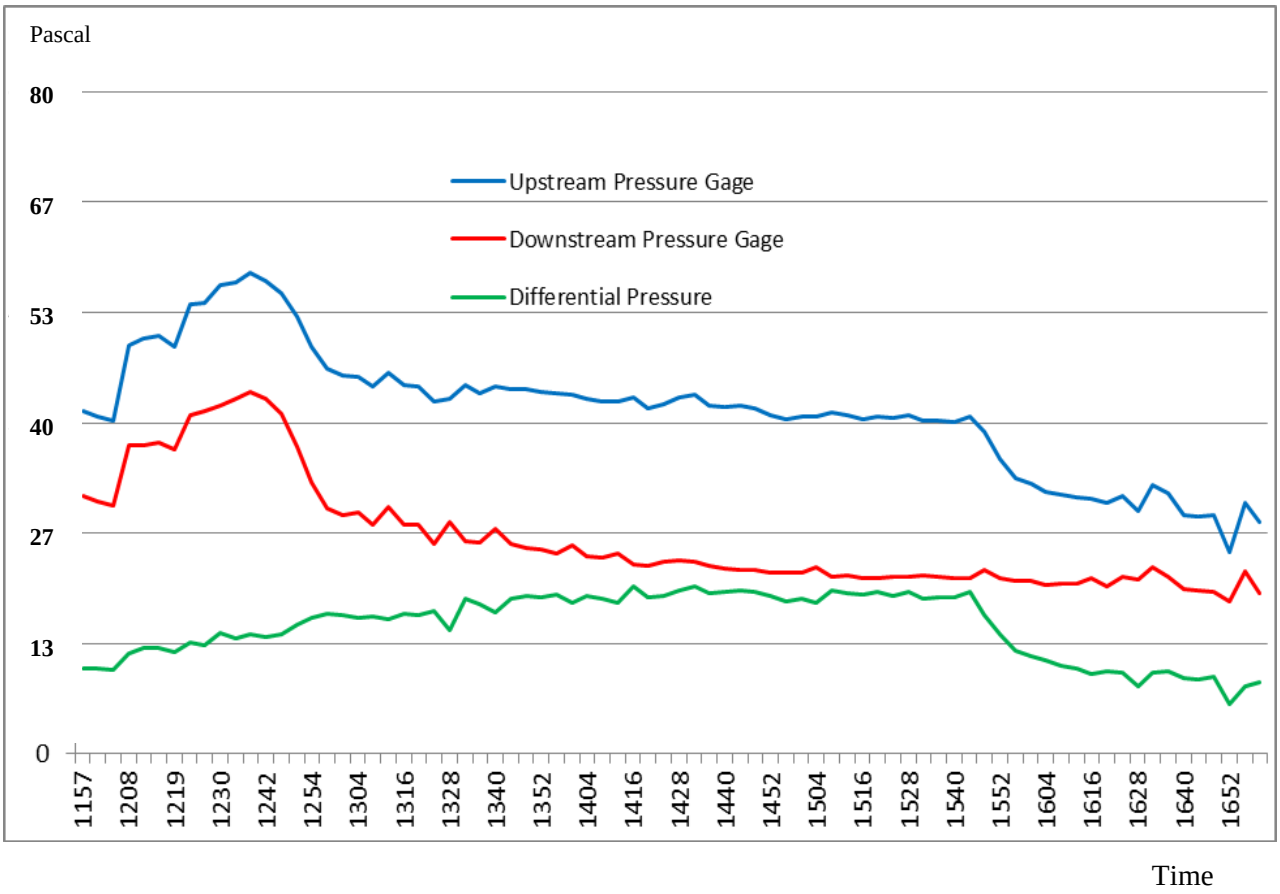

Fig. 3. [Typical Distillation Data Trend].

Directly determining the amount differential pressure that occurs during sodium distillation can be difficult to observe by simply comparing the difference between the pressures of the UPG and DPG on the pressure gauge data trend, due to fluctuations on the pressure gauges caused by vapor state compounds; however, by plotting the differential pressure between the two pressure gauges (ΔP), the increase in differential pressure between the two gauges can be clearly observed. When the pressure differential between the two gauges and the temperature in the vapor line thermo well drop to steady values, sodium distillation has ceased and the process is shut down.

DISCUSSION

During the design and testing phases of this project many lessons were learned pertaining to the distillation and handling of sodium. Portions of the design were more successful than others. During initial testing, design flaws were discovered and the system modified to correct original design deficiencies. The remote-handled waste SDS's design, testing results, and lessons learned are discussed in more detail below.

Keeping the distillation system outside of the hot cell boundary is a key design feature of the system. Previous distillation systems for remote-handled waste have been placed in inert gas hot cells, which must

be operated, maintained, and repaired remotely. By placing the system outside of the hot cell, the shielded room can be accessed with minimal radiological controls for maintenance and repair when remote-handled waste is not in the system. However, due to the high radiation fields of the waste, the system needed to operate remotely. Several challenging design features, such as a remote closure device (a blind flange closure, typical of previous in cell designs is not practical from an exposure perspective) and providing an inert environment to prevent the formation of sodium oxides inside the system, have been incorporated into the design.

As previously stated, the primary design challenge was to operate the system remotely outside of an inert hot cell. This presented two distinct challenges, although only one was recognized at the inception of the design, the remote closure device. It was not recognized that the small amount of leakage past the valve seal while under vacuum and in a distillation state would present a challenge in keeping the system clean. The design team considered and investigated a multitude of remotely operated closure devices, from a variety of valves and autoclave devices. It was discovered during this investigation that the combination of maintaining a vacuum sufficient to achieve distillation at the maximum design temperature was not a product that was commercially available. The design team eventually selected a sliding gate valve that was rated for full vacuum at 649°C that incorporated a silicon O-ring that needed to be cooled through cooling channels in the knife and under the seating surface. This SDS design included a thermal fluid system that would be operated at 149°C to heat the condenser. The decision was made to select this valve and provide the required cooling fluid from the thermal fluid system. Some evidence from the valve manufacturer indicated this valve had been successfully used in a smelting application. While this valve presented some project risk, due to the compressed schedule and long lead time of some of the components, the design team decided to move ahead with this valve.

The design was finalized in December 2013 and fabrication began at Premier Technology, Inc., in Blackfoot, Idaho, a short distance from the INL Site. Component factory acceptance testing was completed in April 2013. Optimization testing began in May 2013 using test fixtures designed to simulate the waste and elemental sodium was provided in individually wrapped 28-g ingots (see Figs. 4 and 5). Calculations predicted that sodium could be distilled in this system at a rate of ~1.4 kg/hour when not restricted by waste forms.

The first distillation cycle was performed on May 14, 2013, using a test fixture loaded with 454 g of sodium.

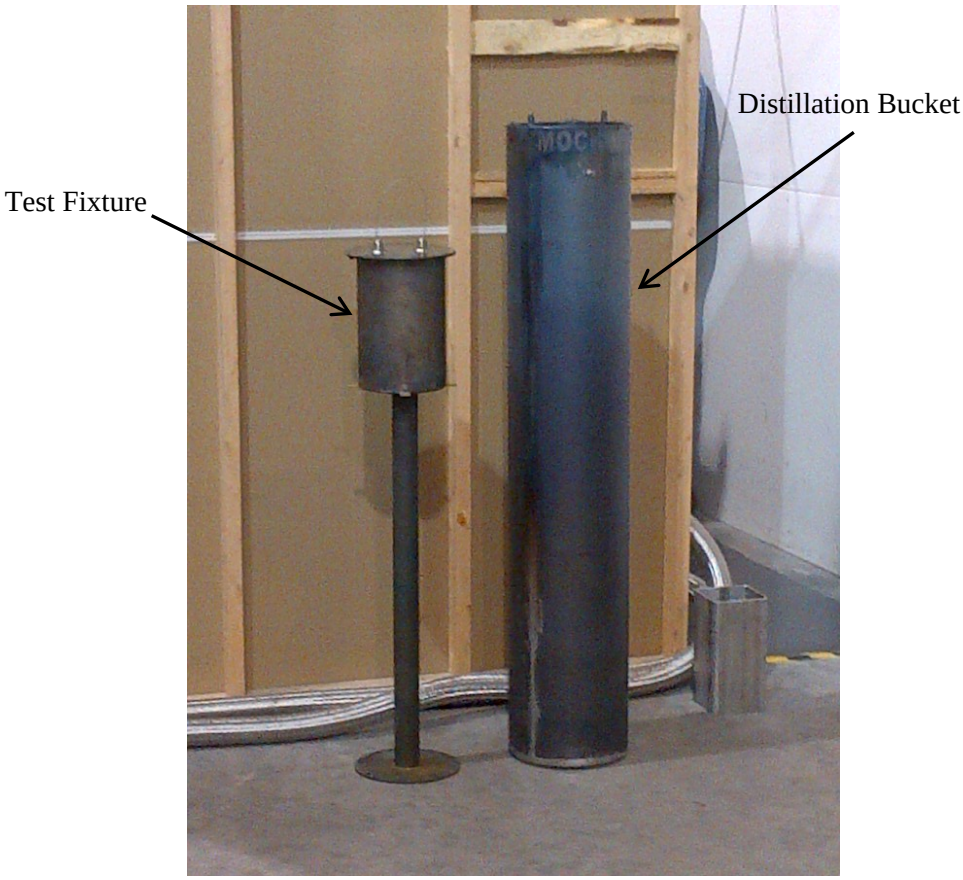

Fig. 4. Test fixture and distillation bucket.

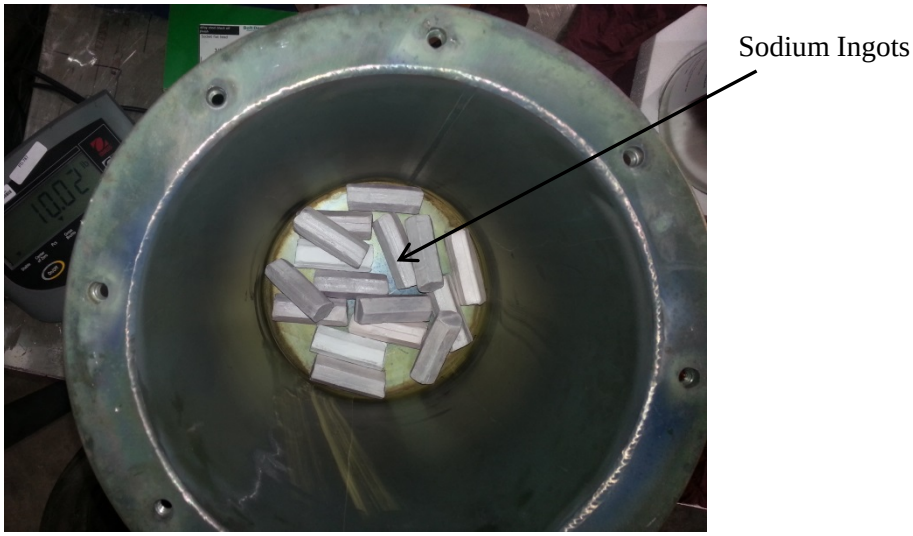

Fig. 5. Sixteen 28-g ingots of sodium loaded in the fixture.

On the morning of May 14, 2013, a test fixture loaded with 454 g of sodium was placed inside the distillation bucket and the bucket was loaded into the SDS. The system was sealed and a vacuum was

established. Heaters and heat trace were ramped up to their maximum settings and the pressure and temperature indications were monitored. After 1 hour of maintaining maximum temperatures it was noted that the UPG pressure was rising, indicating that sodium was boiling. In addition, it was noted that the temperature in the vapor line was increasing, a further indication of sodium vapor flowing past the vapor line thermowell. After 1 hour and 15 minutes of boiling/distillation it was noted that the DPG was raising in pressure, indicating a leak in the system. It was suspected that the silicon O-ring may have failed. Temperatures were lowered to cease any further sodium boiling; however, the system was unable to achieve the vacuum that was expected (less than 27 Pa). The system was secured and allowed to cool overnight.

On the morning of May 15, 2013, the system was opened for inspection. Figure 6 is a photograph of the interior top of the vessel. Note that the O-ring appears intact but, at the vessel/valve flange, there is an indication of inleakage. The white coating is sodium oxide formed when air entered the system, either through the failed metal gasket at the vessel/valve body flange ot past the O-ring interface, or a combination of both. There was evidence of unreacted sodium beneath the surface of the oxide layer. Some larger deposits of oxide were bisected and a silver surface was revealed, which was unreacted sodium.

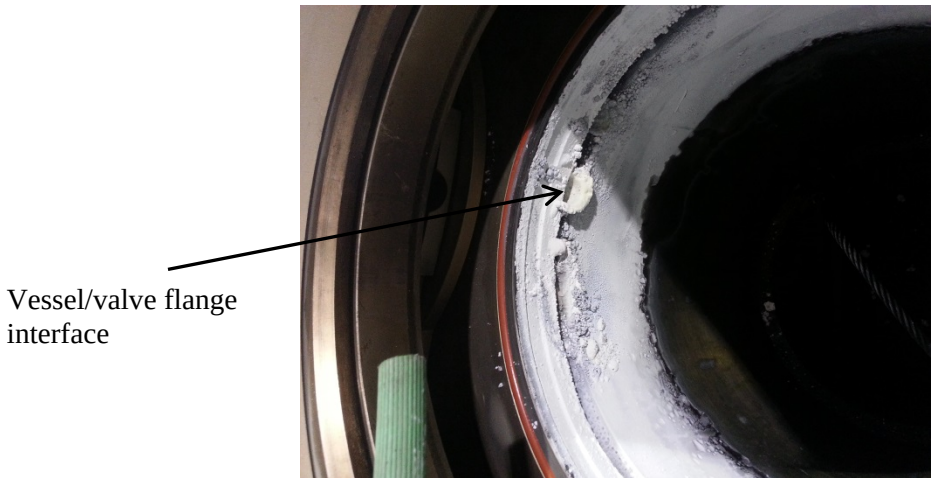

Fig. 6. Interior top of the vessel indicating leakage at the vessel/valve flange.

The distillation bucket was unloaded from the SDS and the test fixture was removed. The test fixture was weighed and opened to invesitigate if any sodium remained in the fixture. Based on the weight, it appeared that all the sodium had successfully been distilled from the test fixture. The fixture was opened; it was visually verified to be free of sodium and further verified through water immersion.

The sodium oxides were also present on the underside of the knife. The valve was dissassembled and all of the residual sodium, sodium oxide, and sodium hydroxides were cleaned. The flange interface was re-torqued to the maximum allowable value for the fasteners.

The next distillation cycle occurred on June 6, 2003. This run produced similar results in that all of the sodium was removed from the surrogate waste but significant quantities of sodium and sodium oxides remained in the distillation bucket and vessel body, as well as deposits on the underside of the valve knife gate. In addition, the metallic seal between the valve body and vessel had failed.

The test provided evidence that the following conditions existed and needed to be addressed.

1. The condition at the top three to four inches of the vessel was too cool to prevent the sodium vapor from condensing at that location.

   To mitigate this condition the knife gate with cooling channels across its surface was replaced with a knife gate with a cooling channel that only provided cooling directly over the O-ring seating area. Additional heating in the form of a 3" wide band heater was added just under the valve/vessel flange to add additional heat at the top of the vessel.

2. The repeated failure of the metallic valve/vessel flange was allowing oxygen to enter the system resulting in the formation of sodium oxides.

   To mitigate this in leakage the gate valve body/vessel flange was seal welded to eliminate any further in leakage at this location.

3. The location of the sodium oxide formation under the gate knife indicated that there was in leakage at the O-ring location.

   To mitigate this condition, argon gas was flooded into the valve body so that any in leakage would minimize oxygen and be non-reactive with the sodium vapor.

These improvements were tested with positive results in preventing sodium oxides from forming at the top of the vessel. However, elemental sodium continued to condense at the top of the vessel directly under the knife gate which would eventually lead to failure of the silicone O-ring. The cooling channel in the knife gate continued to provide a heat sink that facilitated condensation of the sodium vapor in this region. See Fig. 7.

Fig. 7, improved results but still excessive sodium buildup near the top of the vessel

The team evaluated this condition and developed two further modifications; a small diameter metal O-ring was placed directly in front of the silicon O-ring to attempt to protect the silicon O-ring by -acting as a thermal barrier, and a thick walled carbon steel pipe (heat plug) was placed inside the vessel chamber that extended up into the flange region to provide additional heating in the interior cold zone through heat conductance along the length of the pipe. Additional distillation cycles were performed and while these

additions improved the results, there was still sodium condensate in the O-ring area that caused eventual failure of the O-ring and left unacceptable quantities of residual sodium in the seating area. See Fig. 8.

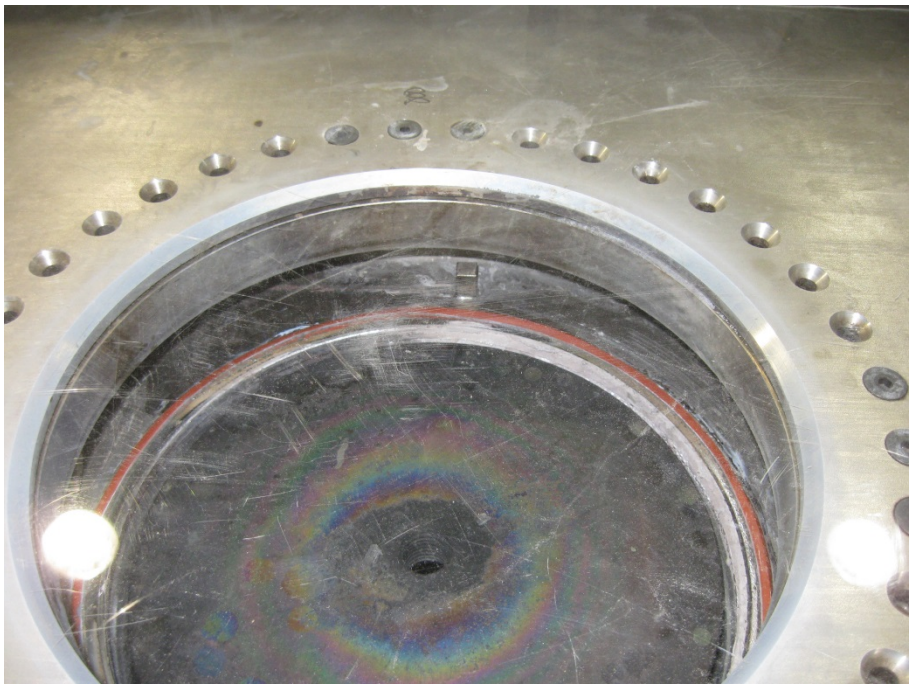

Fig. 8, Sodium Condensate with the Heat Plug Installed

The team had concluded that all cooling in the O-ring seating area had to be secured to reach the desired temperature to achieve complete vapor removal from the vessel. A metal seal was the obvious answer but previous attempts to find a metal seal suitable for this application had failed to secure a source.

In parallel with trying to develop compensatory measures to completely remove all sodium from the vessel, the design team was working with a metal seal manufacturer to develop a metal seal that would work with the sliding gate valve design. A typical metal seal in a vacuum application can take as much as 500 pounds of force per linear inch of seating surface. The valve we selected was designed for elastomer seals and only provides 50 pounds per linear inch of seating surface. A custom thin wall metal seal was fabricated for this application that required $1/10^{th}$ of the seating force normally required for a metal seal.

The team procured there custom seals and installed them in the valve. The cooling flow was secured from the seal seating area and distillation runs were conducted. Subsequent sodium distillation cycles demonstrated that the seating area reached sufficient temperature to eliminate all condensation from the interior of the vessel. See Fig. 9.

Fig. 9, seating area after securing all cooling and installation of a metallic seal

CONCLUSIONS

The Sodium Distillation System for Remote Handled wasted demonstrated that it is successful in treating non-combustible waste that is contaminated with elemental sodium and provides a treatment protocol that has been accepted by the regulatory agencies and the Department of Energy, to remove RCRA waste numbers of D001 (ignitable) and D003 (reactive) from the waste. This is a necessary and crucial step to prepare RH-TRU waste for WIPP disposal.

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