Cesium Removal at Fukushima Nuclear Plant – 13215

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ABSTRACT

The Great East Japan Earthquake that took place on March 11, 2011 created a number of technical challenges at the Fukushima Daiichi Nuclear Plant. One of the primary challenges involved the treatment of highly contaminated radioactive wastewater. Avantech Inc. developed a unique patent pending treatment system that addressed the numerous technical issues in an efficient and safe manner.

Our paper will address the development of the process from concept through detailed design, identify the lessons learned, and provide the updated results of the project. Specific design and operational parameters/benefits discussed in the paper include:

- Selection of equipment to address radionuclide issues
- Unique method of solving the additional technical issues associated with Hydrogen Generation and Residual Heat
- Operational results, including chemistry, offsite discharges and waste generation

Results show that the customized process has enabled the utility to recycle the wastewater for cooling and reuse. This technology had a direct benefit to nuclear facilities worldwide.

THE CHALLENGE

The Great East Japan Earthquake that took place on March 11, 2011 created a number of technical challenges at the Fukushima Daiichi Nuclear Plant. One of the primary challenges involved the treatment of highly contaminated radioactive wastewater. Avantech Inc., in cooperation with Toshiba and Shaw Global Services, LLC developed a unique patent pending treatment system that addressed the numerous technical issues in an efficient and safe manner. The goals of this system were to safely remove the cesium (Cs), minimize waste generation, and ensure operations within the short time frame.

Radionuclide Removal – Challenge #1

As U-235 fissions during normal power operations, it produces heat *(used for power production)* and a variety of radioisotopes know as fission products. Of these fission products Cs-134/137 have the most notoriety due to their strong gamma-ray. Ordinarily these fissions products are maintained within the fuel pellet or fuel rod, but during a meltdown such as that experienced at Fukushima, they are released into the water and/or air around the fuel. Emergency measures during the accident included the use of seawater (45,000 µS/cm) for cooling the reactor following the earthquake and tsunami. The mixing of seawater and fission products produced a particularly challenging waste stream due to its high salinity and extraordinarily high radioactivity. Due to its high Cesium (5×10^6 Bq/cc) content, this mixture known as "accumulated water", was stored underground in site buildings and facilities. Removing the Cesium was critical to ensure the safety of plant personnel, minimize further off-site radiation exposure and to expand upon Tokyo Electric Power Company (TEPCO's) options for water storage. The removal of Cesium in a seawater environment while minimizing waste generation and personnel exposure became the goal.

The removal of cesium in a high salinity water has been studied using differing techniques including co-precipitation, ion exchange, solvent extraction, electrochemical and membrane processes. Ion exchange and natural zeolites are an obvious choice but they do not have a high selectivity for radionuclides, especially Cs. Crystalline silicotitanates (CSTs) are a new class of ion exchangers that were jointly invented by researchers at Sandia National Laboratories and Texas A&M University. One particular CST, known as TAM-5, is remarkable for its ability to separate parts-per-million concentrations of cesium from highly alkaline solutions (pH>14) containing high sodium concentrations (>5M). It is also effective for removing cesium from neutral and acidic solutions, and for removing strontium from basic and neutral solutions. Cesium isotopes are of particular concern due to gamma radiation exposure and volatility at the high temperatures experienced during some waste stabilization techniques such as vitrification. Tests performed at numerous locations with early lab-scale TAM-5 samples established the material as a leading candidate for treating radioactive waste volumes such as those found at the Hanford site in Washington.

Thus Sandia developed a Cooperative Research and Development Agreement (CRADA) partnership with UOP, a world leader in developing, commercializing, and supplying adsorbents. CSTs are now commercially available from UOP. These materials exhibit a high capacity for cesium in a wide variety of solutions of interest to the Department of Energy, and they are chemically, thermally, and radiolytically stable.

Avantech chose the UOP media because of the stability and distribution coefficient (i.e. – sorption capacity) of the CST. With a distribution coefficient greater than 20,000 L/kg, the UOP IONSIV™ media had more than enough capacity for the removal of cesium in a once through process. Avantech and UOP worked extensively to ensure the ion exchanger design provided the optimal design conditions for effective removal of cesium. Furthermore, because the media had not been deployed on a full scale basis, preparation of the media became paramount.

Shielding – Challenge #2

In developing the overall process it was quickly understood that the capacity of the CST media far exceeded our ability to safely shield and cool the ion exchanger. Therefore, we limited the ion exchanger to less than 200,000 curies of activity. The goal was to enable the working staff to be able to maintain the equipment in a work environment of less than 200 millirem per hour (2 millisievert per hour) or less. As a result, we integrated the ion exchanger into a permanently shielded container much like a transportation cask. The integral shielding was designed for a six inch lead equivalent and resulted in the overall vessel weighing approximately twenty three metric tons or fifty thousand pounds. Lead shot was utilized because it expedited the product development and eliminated several transportation issues associated with shipping significant weights between the United States and Japan.

Provided below is a picture of the end view of the Shielded Ion Exchange Module (SIXM) with the shielding annulus shown in Avantech's manufacturing facility.

Heat Generation – Challenge #3

A variety of factors including dose rate, weight and decay heat lead to an accumulated radioactivity limit of 200,000 Ci (7.5E+15 Bq). With two hundred thousand curies of radioactive material in a SIXM, heat generation became a problem during long term storage. Providing a design that dissipated decay heat was critical to preventing a phase change in the ion exchange media and potentially melting lead in the SIXM walls. Avantech and Shaw evaluated numerous passive cooling approaches to resolving this issue. The end result was a cooling annulus between the ion exchanger and the shielded wall with a serpentine of cooling pipes to allow air to passively cool the ion exchanger.

Hydrogen Gas – Challenge #4

Hydrogen gas was a significant concern during both operations and storage of the radioactive material. Accumulation of hydrogen in the process was eliminated through the use of automatic vent valves.

Once an ion exchanger was removed from service it was placed on a dry storage pad. Once in dry storage the ion exchanger would build up decay heat and ultimately evaporate the remaining moisture within the vessel. A concern was raised that hydrogen could accumulate as a result of water radiolysis during the initial storage of the vessels. To alleviate the concern, a battery operated blower was connected to the SIXM vessel during the first forty-eight hours of storage to ensure adequate dissipation of any potential hydrogen.

The Integrated Process

The integrated process was designed to ensure filtration and two stages of ion exchange in two parallel trains. This was accomplished utilizing seven vessels for each train; two filtration, three primary ion exchange, and two polishing ion exchange vessels. The ion exchange vessels were operated in a "lead-lag" carousel scenario to optimize the media usage and allow for a managed distribution of radionuclides.

Each media vessel was designed to look and operate exactly the same to minimize potential operator errors and ensure consistent material handling. The vessels were designed with trunnions that allowed for easy crane handling.

The picture above shows the equipment lay-out.

The customer had a strong desire to have a simple contact handled operation that minimized the potential for operator error and the reliance on automation. Automation was utilized to monitor the process, trend key data, and perform the key flushing activities.

Operating Results

System operation has proven to be very successful because of the simplicity of the design and the effectiveness of the operation. The system has operated at a decontamination factor in excess of 2 million since starting up while achieving throughputs of over 1800 bed volumes of wastewater per bed volume of secondary waste (BV). Due to these high throughputs the system has experienced minimal down time and TEPCO has been able to reduce the size of their waste storage facility. The system to date has processed over eighty (80) million gallons (300,000 cubic meters) of water.

System operations have produced minimal waste generation. The system has produced only approximately 55 canisters which is over 35 times less than the competing process.

Acknowledgements

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