Closing the Circle on One Problematic Nitrate Waste Stream at Los Alamos National Laboratory's NMT-2

Abstract:

The Actinide Process Chemistry Group (NMT-2), Los Alamos National Laboratory have closed the circle on one of the most problematic waste streams in the DOE complex—plutonium-contaminated nitric acid. The Nitric Acid Recovery System (NARS) at the LANL Plutonium Processing and Handling Facility (TA-55) is a distillation process that recycles acid used for plutonium dissolution and recovery. NARS virtually eliminates this waste stream. NARS allows LANL to avoid discharges of TA-55-generated nitrates to the environment. NARS also recycles 100% of radioactivity back into the system, generating activity-free product water.

Return on investment: 128% on a $2,000,000 capital cost.

Nomination Description:

INTRODUCTION

The Actinide Process Chemistry Group (NMT-2), Los Alamos National Laboratory (LANL), has closed the circle on one of the most problematic waste streams in the DOE complex—nitric acid (HNO3) contaminated with plutonium (Pu).

The Nitric Acid Recovery System (NARS), brought online in April 2001 at the LANL Plutonium Processing and Handling Facility (TA-55), is a distillation process that recycles acid used for plutonium dissolution and recovery. NARS has in eight months of operation achieved a 93% source reduction in Pu-contaminated HNO3 effluent. Long-term source reductions attributable to NARS will virtually eliminate this waste stream, with efficiencies projected to approach 99.98% by April 2002. Not only will NARS allow LANL to avoid further discharges of TA-55-generated nitrates at its industrial outfall (an increasingly sensitive environmental issue), but NARS will also recycle 100% of radioactivity back into the system, generating activity-free product water.

Aqueous processing—often by HNO3, using the acid for dissolution and anion exchange—is the chemical workhorse of TA-55, where since 1978 Pu operations at LANL have been centered. With the advent of the 1996 Comprehensive Test Ban Treaty, TA-55 operations have become pivotal to national security because 1) production of Pu pits, the core of nuclear weapons in the US arsenal, is now being implemented at TA-55, and 2) of all facilities in the DOE complex, only TA-55 is equipped to carry out key research to support the Stockpile Stewardship Program. These developments translate into increasing volumes of acid processing at TA-55, compared to a mid-1990s baseline.

NMT-2 recognized this trend as early as 1995 and concurrently recognized a trend among the regulatory community to view with mounting concern heretofore routine, acceptable discharges of nitrates into the environment—concern occasioned by a growing awareness of the negative impact on human health and the ecosystem associated with nitrates. The NMT response to an evolving regulatory climate was proactive promotion of an integrated waste management approach to acidic effluent, with an emphasis on source reduction. Such foresight was rewarded when in 1999 the New Mexico Environment Department (NMED) promulgated a stringent new groundwater discharge permit limit of 44 ppm.

BACKGROUND

Pu is critical to American defense posture. Operations carried out at the 75,000-square-foot Plutonium Processing and Handling Facility, situated at LANL Technical Area-55, are unique and indispensable. At this facility, 239-Pu metal is formed into samples for experiments, cast and/or machined into weapons pits, and recovered from decommissioned weapons for reuse or disposal.

Because Pu decays radioactively and over time becomes contaminated with trace amounts of decay products like uranium, neptunium, and americium, the metal must be purified, after which, it can be reused. Purification is accomplished mainly by means of aqueous processing but also by pyrochemical processing.

In aqueous processing by the HNO3 anion exchange method, Pu-bearing residues, e.g., impure oxides, crucibles, casting molds, and pyrochemical salts, are dissolved in, or leached with, acid. The acid used in dissolution and leaching is strong—at least 12 molar. The HNO3 solution passes through a column packed with thousands of beads made of an anion-exchange resin, which captures the Pu by sorption. When the resin beads are fully “loaded” with Pu, beads are washed with a weak HNO3 eluting solution—about 0.5 molar. The Pu is recovered from the weak acid solution by means of precipitation and calcination.

For the last 20 years at TA-55, effluent streams from dissolution and elution, contaminated with trace amounts of residual Pu and other actinides, were processed in an evaporator. Evaporator bottoms were stabilized with cement in drums and stored for eventual shipment to the Waste Isolation Pilot Plant (WIPP) in Carlsbad, New Mexico. The distilled liquid from the evaporator was condensed and sent via the Process Acid Waste Line to the LANL Radioactive Liquid Waste Treatment Facility (RLWTF) at Technical Area-50, where the acid was neutralized and the actinides were precipitated out. The neutralized effluent was discharged to the environment through an industrial outfall, and the precipitate was stabilized with cement in drums and stored for shipment to WIPP.

After 20 years, TA-55 has changed its way of doing business. Evaporator distillate is now sent to a newly installed recycle distillation unit—the heart of NARS—that recovers HNO3 through fractional distillation. Fractional distillation is a well established process in the chemical and petroleum industries. It is used to separate constituents with different boiling points. Water boils at a higher temperature than does HNO3, allowing purified water to be removed from the top of the distillation column and discharged to the environment without further treatment. The reconcentrated acid is removed from the bottom and reused. See Figure 1.

BENEFITS

LANL has derived major benefits from NARS, which reduces by 99.98% the source of TA-55-generated nitrate waste going to RLWTF. Given increasing levels of Pu processing at TA-55 since the mid 1990s, future nitrate releases to the environment from RLWTF would have increased in magnitude as well, absent NARS. Further, reducing releases of radioactivity by reducing a source virtually to zero is an environmentally responsible course of action. Before NARS came on line, up to 60 microcuries per liter of liquid was sent down the Process Acid Waste Line to RLWTF.

Information on cost savings is presented in the SIGNIFICANCE and RETURN ON INVESTMENT sections below.

SIGNIFICANCE

The impact of NARS is dramatic:

- a 99.98% annual reduction in TA-55-generated HNO3 going to RLWTF
- a 100% annual reduction in radioactive releases to the environment resulting from treatment at RLWTF of TA-55-generated HNO3
- an 80% annual reduction in high-molarity HNO3 procurements at TA-55
- a significant reduction in the number of drums of transuranic waste destined for WIPP
- a 128% return on investment on capital costs of $2 million.

Disposing of a drum of conforming newly-generated transuranic waste at WIPP involves packaging, certifying, and shipping costs at the generator end (about $14,500) and lifecycle costs at the receiving end ($9950). The true dollar costs of nitrate releases are not directly calculable by standard accounting practices, but the stringent new NMED standards reflect a firm consensus in the regulatory community that they are high.

The return-on-investment calculation is based on comparisons of two alternate strategies: one to install new denitrification processes downstream at RLWTF, the other to install NARS upstream at TA-55 (see the RETURN ON INVESTMENT section below). Not surprisingly, the upstream solution prevailed.

Apart from cost savings, source reduction of HNO3 waste by 99.98% is a significant pollution prevention achievement. While HNO3 is a familiar reagent in technological-industrial settings, it is by no means a benign substance. The acid, when concentrated in the 12-15 molar range, is highly corrosive. When contaminated with actinides characterized by long half-lives (24,100 years in the case of Pu), the resulting waste is particularly hazardous.

ORIGINALITY

Implementing NARS required innovative engineering solutions to solve problems unique to the TA-55 workplace. Industrial-scale fractional distillation columns commonly stand more than 100 feet tall and are normally designed to operate under the open sky. Further, they are designed for continuous operation over months at a time. To bring NARS on line, in an enclosed facility where space was restricted, the technology essentially had to be miniaturized. The column is 10 inches in diameter and stands 13 feet tall (see Figure 2). Security requirements at TA-55 limit operation time to 10 hours a day, which incurs novel safe-startup and -shutdown issues.

The engineering problems were solved through close collaboration between NMT-2 staff and the Atlanta Technology Group, of Tucker, Georgia, a firm with extensive experience in designing specialty chemical process equipment.

POTENTIAL FOR BROADER APPLICATION

NARS technology can be applied in any application requiring acid concentration where space is at a premium.

More importantly, NARS has been instrumental in promoting the pollution prevention ethic at TA-55. Its success has prompted other organizations dealing with special nuclear materials to participate in a top-down reexamination of operations, with a view to implementing source reduction. This commitment is systematized in the TA-55 Waste Minimization Plan (1996), NMT Waste Management Program Plan (1995), and the Transuranic Waste Minimization Plan (in press). Specific targets announced to date in connection with these plans call for a 90% reduction in liquid waste and the virtual elimination of combustible waste, with additional targets currently under study.

RETURN ON INVESTMENT

The project is estimated to post a return on investment of 128% on a $2 million capital cost. The calculation was made by comparing two alternate strategies for dealing with TA-55-generated HNO3: one to install new denitrification processes downstream at RLWTF, the other to install NARS upstream at TA-55. Operational costs were annualized on the basis of 10 years of useful life, though actual life expectancy may extend to 20 years. A detailed discussion of the calculation is inappropriate here. Further information is available in Boerigter, Stephen T., "Cost/Benefit Analysis for Selected Waste Minimization Technologies at TA-55," Los Alamos National Laboratory LA-UR-95-1977 (1995).

The importance of eliminating the actinide-contaminated HNO3 waste stream has been discussed in the SIGNIFICANCE section above. The team members nominated for the award are: Matthew W. Bailey, NMT-2, Ronald Chavez, NMT-2, Anna D. Flores, NMT-2, Willie Maestas, NMT-2, Benjie T. Martinez, NMT-2, Dennis P. Montoya, NMT-2, Mel J. Montoya, NMT-2, Joey L. Moya, NMT-2, Don W. Mullins, NMT-2, Howard L. Nekimken, NMT-2, Stephen B. Schreiber, NMT-2, Wayne D. Smyth, NMT-2, Aquilino D. Valdez, NMT-2, and Stephen L. Yarbro, NMT-DO. For more information, contact Tom Starke, Environmental Stewardship Office, (505) 667-6639, tps@lanl.gov.

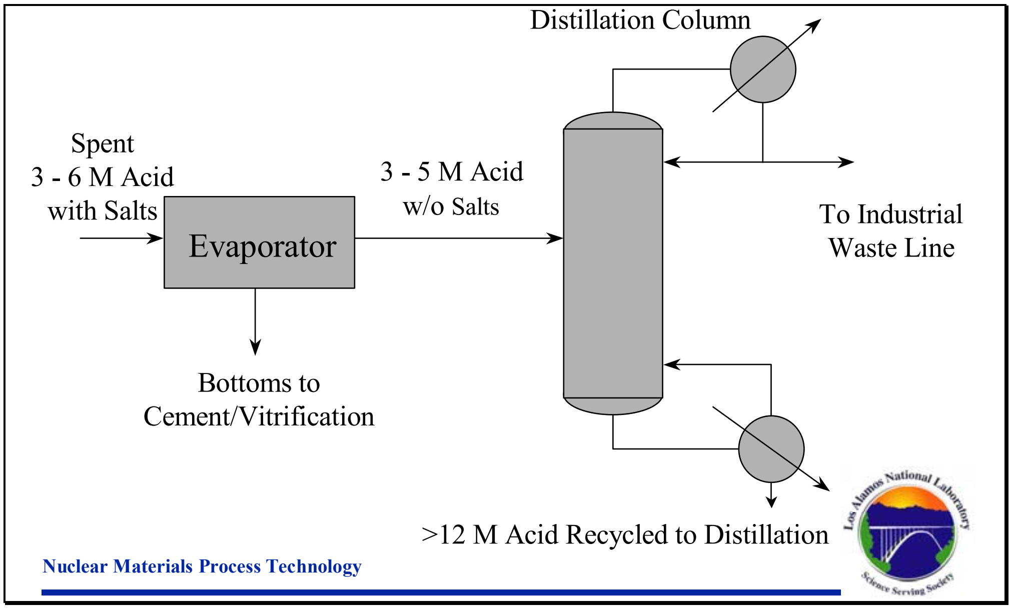

Figure 1. NARS process flowsheet.

Figure 2. Photograph of the NARS unit. The 13-foot fractional distillation column is the vertical member just left of center.