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Attachment A

ADVANCED OFF-GAS CONTROL SYSTEM DESIGN FOR RADIOACTIVE AND MIXED WASTE TREATMENT Nick Soelberg, Idaho National Laboratory September 2005 INL/CON-05-00658, **INL/CON-05-00658**

"Off-gas control technologies and system designs that were satisfactorily proven in mixed waste operation prior to the implementation of new regulatory standards are in some cases no longer suitable in new mixed waste treatment system designs. Some mixed waste treatment facilities have been shut down rather than have excessively restrictive feed rate limits or facility upgrades to comply with the new standards.
New mixed waste treatment facilities in the U. S. are being designed to operate in compliance with the HWC MACT standards. Activities have been underway for the past 10 years at the INL and elsewhere to identify, develop, demonstrate, and design technologies for enabling HWC MACT compliance for mixed waste treatment facilities. Some specific off-gas control technologies and system designs have been identified and tested to show that even the stringent HWC MACT standards can be met, while minimizing treatment facility size and cost. [abstract]

"Mixed waste incineration has declined in the U.S. for several related reasons. Public opposition to incineration in general, and regulation of mixed waste thermal treatment under the new National Emission Standards for Hazardous Air Pollutants (NESHAPS) for hazardous waste combustion [Hazardous Waste Combustor (HWC) Maximum Achievable Control Technology (MACT) standards], contributed to the reassessment of existing and planned U.S. Department of Energy (DOE) mixed and radioactive waste incinerators.

"Regardless of whether incineration or other thermal treatment technologies were used, mixed waste treatment would generally be regulated under the HWC MACT standards unless other regulations applied, such as the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA). Even under CERCLA remediations, requirements such as the MACT standards could be applied as CERCLA Applicable or Relevant and Appropriate Requirements (ARARs).Off-gas control technologies and system designs that have been satisfactorily proven in mixed waste operation prior to the implementation of new regulatory standards are in some cases no longer suitable. New mixed waste treatment off-gas system designs or retrofits of existing facilities, that need to comply with new regulations such as the HWC MACT standards, should consider technologies or features that can improve regulatory compliance and lower costs.
[pg. 1 and 2]

"THE HWC MACT STANDARDS
The U.S. Environmental Protection Agency (EPA) has regulated air pollutant emissions from hazardous waste combustors based on maximum achievable control technology (MACT). The full regulation is under National Emission Standards for Hazardous Air Pollutants (NESHAP): Final Standards for Hazardous Air Pollutants for Hazardous Waste Combustors, [U. S. Code of Federal Regulations (CFR), Title 40, Part 63, Subpart EEE (Part 63 Sections 1200 through 1214)], most recently revised July 1, 2004 (EPA 2004a). The Hazardous Waste Combustor (HWC) MACT standards were promulgated in a joint effort of Resource Conservation and Recovery Act (RCRA) and the Clean Air Act (CAA) regulations, intended to consolidate and revise air emission and operational requirements previously regulated by RCRA.

"The EPA has included mixed waste thermal treatment facilities among facilities regulated under the HWC MACT standards because of the hazardous waste component of mixed waste. Shortly after the HWC MACT standards were proposed, the U. S. Department of Energy (DOE) Mixed Waste Focus Area provided commented to EPA that it was not appropriate or practical to regulate mixed waste treatment facilities under the HWC MACT standards (Eaton 1996, INEEL 1996, Pelletier 1997), because the radiological hazards of mixed waste, in addition to the chemical and toxic hazards associated with the hazardous waste component, make mixed wastes and mixed waste treatment facilities sufficiently unique to require other regulations than those that focus just on the hazardous waste component. The EPA considered excluding mixed wastes from the HWC MACT standards, but eventually included mixed wastes in the MACT standards promulgation.

"These tank wastes are aqueous solutions that contain practically no organic content. During vitrification, they would produce essentially no combustion gas, even though organic reductants added to the melter feed to react with nitrates and nitrites in the feed will produce combustion gas (CO_2 and H_2O). The 7% O_2 correction is not possible when the off-gas, as in the case of Hanford's melters, is primarily purge and cooling air (Oh 2000).

"NEW MIXED WASTE THERMAL TREATMENT AND OFF-GAS CONTROL SYSTEMS IN THE U.S. Some of these, such as the LLW and HLW melter systems for the Hanford River Protection Project, are under construction. Others, such as the In-Container Vitrification (ICV) melter process for supplemental LAW waste treatment at Hanford, and the fluidized bed steam reformer system at INL, are in design and demonstration phases. Some other systems such as the proposed SBW Vitrification Facility, and the proposed NWCF upgrade for HWC MACT compliance, were conceptually designed, complete with equipment and facility sizing and mass and energy balances, but were eventually not selected for further design and construction.

"These new or proposed mixed waste treatment facilities indicate how specific mixed wastes are being treated to meet storage and disposal requirements and how compliance to the HWC MACT standards is being accomplished for mixed waste treatment facilities. Pre-existing facilities that are continuing operation with HWC MACT compliance are doing so with feed limits or by limited modifications to enable compliance.
Regardless of primary mixed waste thermal treatment technology, MACT-compliant off-gas systems generally have these unit operations:
Comply with As Low As Reasonably Achievable (ALARA) objectives by minimizing the exposure of workers, the public, and the environment to radiological and other hazards. [pg 3]

"Table III. Example mixed/radioactive waste treatment and off-gas control systems currently planned, proposed, or under construction in the U.S.

Facility	Treatment system	Off-gas system	Status and comments
Proposed SBW Waste Vitrification Facility at the INL	Refractory-lined joule-heated melter	Film cooler, acid quench, venturi, HEME, reheater, prefilter, HEPA, staged NO_x and organics destruction, quench, ME, reheater, carbon bed Hg sorption, HEPA, ID fan	Vitrifying the SBW was one of several alternatives evaluated for treating the SBW (Quigley 2000, Bates 2001, Taylor 2001, Barnes 2004). The off-gas system was designed to be HWC MACT-compliant (Wood 2001). SBW vitrification was eventually not selected as the preferred SBW treatment option.
Proposed SBW steam reforming facility at the INL	Fluidized bed steam reformer system	Cyclone, oxidizing unit, partial quench, prefilter, HEPAs, carbon bed Hg sorption, ID fan	SBW steam reforming was one of several alternatives evaluated for treating the SBW (Williams 2002, Barnes 2004, Cowan 2005). Steam reforming has been selected as the preferred SBW treatment option.

"OFF-GAS TECHNOLOGIES AND CONCEPTS FOR FUTURE OFF-GAS SYSTEM DESIGNS

"Each of these treatment systems has included offgas control systems. In the past decade, the INL has tested, developed, and designed advanced treatment technologies including high temperature melters, thermal desorption, and fluidized bed calcination and steam reforming technologies.

"These projects have included off-gas control technology development and demonstrations in the five most challenging areas, or areas of greatest need and technical uncertainty, for mixed waste off-gas control: high temperature filtration, NOx control, organics oxidation, Hg control, and off-gas system design concepts. A few recommendations can now be made for future mixed waste off-gas system designs based on work at the INL and advances elsewhere that can provide more confidence in certain new off-gas control technologies or new applications. [pg 6]

"During the late 1990's, the DOE Mixed Waste Focus Area funded high temperature filtration demonstration projects. Most recently, high temperature filtration was included for the past 4 years of periodic demonstration tests performed by the INL for fluidized bed steam reforming (Olson 2004).

"These successful operations and demonstrations of high temperature filtration provide operating data showing that high temperature filtration can be used more widely in mixed waste off-gas systems. Both sintered metal and ceramic filters have been used with success, and each have specific advantages.

"Sintered metal filters, such as were used in the INL steam reforming tests, are less susceptible to physical or thermal shock. Removal efficiencies for the INL filters ranged between 99.5% to over 99.9%. While ceramic filters are susceptible to breakage from physical or thermal shock, these are used successfully in the Studsvik radioactive waste steam reforming facility with removal efficiencies ranging up to 99.9%. The filters are replaced during every shutdown, by allowing the old filters to fall into the filter hopper after which they are broken up and combined with the filter ash product. [emphasis added]

"NO_x Control and Organics Oxidation

"Several NO_x control technologies including selective catalytic reduction (SCR), non-selective non-catalytic reduction (NSNCR), and steam reforming have been studied for mixed waste off-gas systems for many years.

"However, concerns about SCR catalyst poisoning, SCR reagent handling, process control during upset conditions, and formation of potentially explosive ammonium nitrate, have limited SCR applications and have increased process cost and complexity.

"The INL has discarded SCR NO_x control in favor of NSNCR, also called staged combustion. Test results and modeling (MSE 2001, Boardman 2004, Olson 2004) have provided data and confidence in the ability of NSNCR to achieve high efficiency NO_x destruction (exceeding 99% under some conditions) and high efficiency destruction (exceeding 99.99% for some conditions) of residual organics in off-gas streams from melters, calciners, and steam reformers. Properly operated NSNCR systems can achieve not only highly efficient destruction of off-gas NO_x resulting from processing nitrate and nitrite-bearing mixed wastes, but also can replace any other offgas organics control technology. This combination eliminates any concerns related to SCR NO_x control and can meet applicable regulatory limits for both NO_x and hydrocarbon emissions and for POHC destruction efficiency.

"NSNCR systems tested to date have used added fossil fuel (natural gas, propane, or fuel oil) to provide heat needed to heat the off-gas to the desired operating temperatures of 800- 1,000$_o$C, and to adjust the off-gas toichiometry in the first (deNO_x) stage. The added stage 1 fuel (and air, if needed) needed to heat the off-gas can cause the total off-gas flowrate to increase by 1.5 to 3 times. This increase can be eliminated by using electrical or indirect heating to heat the off-gas to the stage 1 temperature. A demonstration-scale prototype of an electrically-heated NSNCR process for destroying NO_x and residual hydrocarbons from a liquid-fed cold crucible induction melter (CCIM) is shown in Figure 1.

"Mercury Control

"Mercury was used in fuel reprocessing, and so is present in liquid mixed wastes from nuclear fuel reprocessing activities. Mercury control efficiencies exceeding 99.9% are required for thermally treating these wastes compliant to the HWC MACT standards. The INL has been studying and developing technologies to remove Hg from the liquid wastes, and to remove Hg from mixed waste treatment off-gas, for over a decade (Chambers 1998, Soelberg 2003b). Results show that (a) even if waste pretreatment is used to remove much of the Hg prior to thermal treatment, efficient off-gas Hg control will still be necessary for Hg-laden fuel reprocessing wastes, and (b) the only reliable and efficient technology presently available for Hg control in mixed waste off-gas systems is sulfurimpregnated activated carbon beds. Wet scrubbing, used in some non-nuclear applications, is not reliable enough or efficient enough for removing off-gas Hg regardless of speciation. Innovations such as oxidizing systems to oxidize elemental Hg to less volatile or more water-soluble species, that would enable more efficient and reliable Hg wet scrubbing, are promising, but not sufficiently demonstrated for mixed waste processes. Carbon injection, used worldwide for Hg and dioxin/furan control, is not generally efficient enough, and it generally produces up to 10 times more spent carbon waste than fixed carbon beds do. [pg. 6 & 7]

"Test results and modeling over several years provide performance data and confidence in the ability of NSNCR to achieve high efficiency NOx destruction (exceeding 99% under some conditions) and also high efficiency destruction (exceeding 99.99% for some conditions) of residual organics in off-gas streams from melters, calciners, and steam reformers.

"Using electrical or indirect heating to heat the off-gas to the stage 1 temperature can reduce the total off-gas flowrate. Mercury is ubiquitous in liquid mixed wastes from nuclear fuel reprocessing activities. Mercury control efficiencies exceeding 99.9% are required for thermally treating these wastes compliant to the HWC MACT standards. Fixed beds of sulfur-impregnated activated carbon are still the best technology presently available for achieving this level of Hg control in mixed waste off-gas systems.

" Using the off-gas technologies described above, some innovative mixed waste off-gas systems can be configured that are simpler, and might be more reliable, have lower technical risk, and lower costs than some current designs." [pg8]