

**Title: MULTISTAGE COMBUSTION SYSTEM FOR HIGH-LEVEL NO<sub>x</sub> REDUCTION AT MSE TECHNOLOGY APPLICATIONS, INC.'S OFFGAS TEST UNIT**

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## BACKGROUND

For more than 3 decades, the Idaho National Engineering and Environmental Laboratory (INEEL) has used a calciner plant to treat radioactive and hazardous liquids, converting millions of gallons of potentially troublesome liquids into a more stable form.

The New Waste Calcination Facility (NWCF) at the INEEL was built in 1982, prior to the enactment of many of today's hazardous waste and air-quality laws. This facility now faces an uncertain future given the proposed Environmental Protection Agency's (EPA) new Maximum Achievable Control Technology (MACT) requirements.

As a result of thermal processing of high nitric acid-laden waste, offgas from the NWCF contains approximately 35,000 parts per million volume (ppmv) of oxides of nitrogen (NO<sub>x</sub>). This NO<sub>x</sub> level needs to be reduced for two reasons. First, this high level of NO<sub>x</sub> makes it difficult, if not impossible, to obtain quality standard EPA isokinetic sampling as required to verify compliance with the new MACT rule. This inability to obtain quality samples is due mainly to excessive acid condensation in the sampling train. Also, an orange cloud is sometimes present in the NWCF area, increasing public pressure to reduce NO<sub>x</sub> emission levels. After evaluating available technologies, including selective catalytic reactors, a multistage combustion NO<sub>x</sub> removal system was selected by the NWCF as the technology most feasible to control NO<sub>x</sub> emissions.

MSE Technology Applications, Inc.'s (MSE) Controlled Emissions Demonstration (CED) Project focuses on installation, testing, and evaluation of state-of-the-art offgas treatment technologies and monitoring equipment. The CED Project is sponsored by the U.S. Department of Energy's (DOE) Mixed Waste Focus Area (EM-50). One project in the Fiscal Years 1999 and 2000 project work scope is to install and test a pilot-scale multistage combustion NO<sub>x</sub> removal system. The objectives of this project are to install a pilot-scale multistage

combustion NO<sub>x</sub> removal system in MSE's offgas test bed, set up process conditions similar to the NWCF's exhaust gas and NO<sub>x</sub> emission level, test the performance of the NO<sub>x</sub> removal system, report the test results, and perform a scale-up study based on installation at the NWCF.

## HYPOTHESIS, TEST OBJECTIVES, AND EXPECTED RESULTS

Staged combustion incinerators have been used in many industrial applications to reduce NO<sub>x</sub> emissions from waste gas streams having relatively low NO<sub>x</sub> concentrations. Data is needed on the use of staged combustion on waste gas streams having high-level NO<sub>x</sub> concentrations and relatively low organic concentrations prior to installing a full-scale NO<sub>x</sub> removal system at the NWCF. By employing a three-stage combustion process where the high NO<sub>x</sub> surrogate offgas is first converted under reducing conditions, then quenched to a lower temperature, and finally reoxidized under a moderate temperature and excess oxygen (O<sub>2</sub>), a 95% reduction is expected for inlet concentrations of up to 40,000 ppmv of NO<sub>x</sub>.

MSE will test the NO<sub>x</sub> reduction capabilities of a multistage NO<sub>x</sub> reduction system on a waste gas stream containing up to 40,000 ppmv of NO<sub>x</sub> at varying (up to 4:1) nitrogen dioxide/nitrogen oxide (NO<sub>2</sub>/NO) ratios and perform scale-up projections based on installation at the NWCF.

## MSE's TEST-BED DESCRIPTION

The NO<sub>x</sub> treatment test-bed section will be installed in the MSE slipstream test bed (SSTB). Upstream of the SSTB, the primary thermal driver can be selected from two options: a 500-kilowatt electric plasma arc torch or a 1.6-million British thermal units per hour (Btu/hr) O<sub>2</sub>-enriched natural gas-fired burner. Both of these thermal drivers have been shown to be capable of generating high NO<sub>x</sub> concentration levels, i.e., greater than 3,500 ppmv. Downstream of the primary thermal drivers is a 540,000-Btu/hr secondary combustion chamber. Approximately 2.4 pounds per minute mass flow of offgas will be drawn from the main offgas system to supply approximately 3,500 ppmv of initial NO<sub>x</sub>-laden gas to the NO<sub>x</sub> removal test system. In the upcoming testing, it is anticipated that the O<sub>2</sub>-enriched natural gas-fired burner will be used as the primary thermal driver. Typically 8% carbon dioxide (CO<sub>2</sub>) levels will be present in this offgas. Additionally, if required, spikes of organics and metals could be added to the supply offgas.

In order to test the multistage NO<sub>x</sub> removal system at up to 40,000 ppmv, a NO<sub>x</sub>-formation subsystem will be used to produce additional NO<sub>x</sub> in the offgas to be tested. The additional NO<sub>x</sub> will be generated by injecting 6 molar nitric acid into an electrically-heated flameless oxidizer.

An in-line electric heater will be installed upstream of the flameless oxidizer to preheat the temperature of the offgas to approximately 900 to 1,000 EF. The preheating enhances nitric acid vaporization and subsequent conversion to NO<sub>x</sub> within the electric oxidizer.

The nitric acid will be injected using a metering pump and atomizing nozzle injector into the offgas piping through ports located immediately upstream of the flameless oxidizer. The flameless oxidizer unit will control the offgas temperature to approximately 1,000 EF at the outlet. This temperature, combined with a gas residence time of 6 to 8 seconds in the heating unit, will convert the nitric acid into the target NO<sub>2</sub>/NO speciation ratio of 4:1. The stream composition analyzer will be configured downstream of the electric flameless oxidizer unit to monitor offgas conditions. The NO<sub>x</sub>-laden offgas will then be piped to the experimental multistage NO<sub>x</sub> removal unit.

## MULTISTAGE COMBUSTION NO<sub>x</sub> REMOVAL SYSTEM PROCESS DESCRIPTION

The staged combustion approach involves initial combustion under O<sub>2</sub>-deficient conditions, followed by offgas cooling and final combustion under excess O<sub>2</sub> conditions. The approach takes advantage of the basic thermodynamics of NO<sub>x</sub> formation, which is favored by excess O<sub>2</sub> and temperatures above 2,400 EF. Based on thermodynamic and kinetic modeling and previous operating experience at lower concentrations, stage combustion is expected to provide a 95% reduction of NO<sub>x</sub> at levels up to 40,000 ppmv.

The NO<sub>x</sub> removal system to be tested is a three-stage process. The first stage (offgas supplemented with natural gas) will burn fuel rich with an expected flame temperature near 2,600 EF and a residence time of approximately 2 seconds. Both existing test data and theoretical models indicate that under these conditions, NO<sub>x</sub> and the other offgas constituents will react to form nitrogen (N<sub>2</sub>), CO<sub>2</sub>, water, carbon monoxide (CO), and hydrogen (H<sub>2</sub>), that is, the NO<sub>x</sub> will convert to N<sub>2</sub>. The offgas at this point has residual fuel value in the form of CO and H<sub>2</sub>, and may also have fuel value in the form of partially burned hydrocarbons.

In the second stage, the offgas will be water quenched to approximately 1,400 EF to limit reformation of NO<sub>x</sub> in the subsequent oxidizing stage.

In the third stage, O<sub>2</sub> will be added to burn the residual CO, H<sub>2</sub>, and any unburned hydrocarbons. Sufficient O<sub>2</sub> will be added to maintain the stack at 1.5 to 2% excess O<sub>2</sub>. The temperature at this stage will be maintained at 1,800 to 2,000 EF by addition of quench water at the prior stage. This temperature permits conversion of residual fuel value to CO<sub>2</sub> without reforming NO<sub>x</sub>. The residence time in this stage will be approximately 1 second (chosen primarily to permit conversion of CO to CO<sub>2</sub>).

NO<sub>x</sub>-speciation and stream-composition continuous emissions monitoring system analyzers will be installed upstream and downstream of the NO<sub>x</sub> removal system to determine the NO<sub>x</sub> reduction capability of the multistage combustion process.

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