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A Pilot-Scale Process Development Unit for Transport and Fluid-Bed Hot-Gas Desulfurization

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Introduction

The Morgantown Energy Technology Center (METC) has designed and is currently constructing an on-site, hot gas desulfurization (HGD) Process Development Unit (PDU). The PDU is designed to use regenerable solid metal oxide sorbents that absorb hydrogen sulfide from high-temperature, high-pressure simulated coal-gasification fuel gas that is generated by a METC designed syngas generator. The simulated coal gas is a mixture of partially combusted natural gas, water, carbon dioxide, and hydrogen sulfide. PDU process conditions will be representative of anticipated commercial applications in terms of temperatures, pressures, compositions, velocities, and sorbent cycling. The PDU supports the Integrated Gasification Combined Cycle (IGCC) mission at METC by providing a test bed for development of IGCC cleanup systems that offer low capital cost, operating costs, and costs of electricity. METC intends to develop additional industrial involvement opportunities as the project progresses towards operations.

Objectives

The primary objectives of the PDU are to (1) fill the gap between small-scale testing and large-scale demonstration projects by providing a cost effective test site for transport and fluid-bed desulfurization reactor and sorbent development, (2) demonstrate sorbent suitability over a wide range of parameters, and (3) generate significant information on process control for transport and fluidized bed based desulfurization. PDU data is expected to be used to optimize process performance by expanding the experience for larger scale demonstration projects such as Sierra Pacific Power Company's Clean Coal Technology project.
Background

During the PDU’s early conception[1,2], an IGCC-system economic study showed minimal cost and performance differences when using low velocity HGD fluid beds versus fixed or moving beds. However, installation and operating costs could be lowered by using higher fluidizing velocities[3]. The study also revealed economic advantages for a system that uses a minimal amount of undiluted regeneration air. Given this information and the encouraging results from small-scale transport reactor testing, METC designed the PDU to explore the advantages of higher velocity regimes and alternate contacting modes. Transport reactor provisions were then incorporated into the conceptual design on both the sulfidation and regeneration sides of the PDU[4]. Because the PDU does not require a coal feed or gasifier system, it should cost 25 to 75 percent less per test-hour than would otherwise be required.

At that time, METC decided to draw upon existing gas-solid processing technology. In particular the similarity between the continuous, integrated cracking and catalyst regeneration operations used in fluid and transport catalytic cracking (FCC) units and the PDU concept was viewed as a potential link to success. METC took advantage of much of the existing industry expertise by teaming with the M.W. Kellogg Co. (MWK) for preliminary and detailed design activities. MWK designed the PDU reactor system while METC designed the balance of plant facilities. With METC acting as the general construction contractor, activities were split between several contractors for underground and above ground utilities, civil construction and vessel fabrication. METC personnel are doing the instrumentation and control, piping field fits and similar activities. In addition, METC will conduct all operations and maintenance.

PDU Description

Design Features

The PDU operates at 400 psia (2,750 kPa) pressure and at temperatures up to 1,200 °F (650 °C) on the sulfidation (fuel gas) side and up to 1,400 °F (760 °C) on the regeneration (air) side. The unit continuously circulates sorbent material between the sulfidation and regeneration sides of the desulfurization system. The PDU has provisions for fluid bed and transport contacting. When operating in the fluid bed contacting mode, fuel gas is fed into an 18-inch (.457-meter) inside diameter (i.d.) reactor and sorbent is circulated with steam or nitrogen to the 10-inch (.254-meter) i.d. regenerator reactor. When operating in the transport mode, a 5.2-inch (.132-meter) i.d. absorber riser reactor is used along with a 1.7-inch (.043-meter) i.d. regenerator reactor. Density difference is the primary driving potential for circulation. The following are a few of the primary design features of the METC PDU.

- Fuel gas flow: 60,000 to 120,000 scfh typical
- H₂S concentration: 0.5 to 1 volume % typical
- Absorption temperature: 1,000 to 1,200 °F design
- Regeneration temperature: 1,100 to 1,400 °F design
- Absorber-regenerator differential temp.: 400 °F maximum
- Operating pressure: 400 psia maximum
- Fluid-bed absorber: 18-inch i.d., 10-ft bed maximum
- Transport absorber: 5.2-inch i.d., 50-ft length
• Fluid-bed regenerator: 10-inch i.d., 12-ft bed maximum
• Transport regenerator: 1.7-inch i.d., 50-ft length
• Underflow standpipes: 1.7 to 6.8 inch i.d., 20-ft length (approx.)
• Fluid-bed superficial velocities: 1 to 3 ft/s typical
• Riser superficial velocities: 15 to 20 ft/s typical
• Sorbent inventory: 1,000 to 2,000 lb typical
• Sorbent cycles per day: 50 to 100 typical
• Circulation rate: 2,000 to 5,000 lb/hr typical
• Transport absorber recirculation rate: 5,000 to 55,000 lb/hr typical
• Transport regenerator recirculation rate: 0 to 2,000 lb/hr typical
• Riser bulk densities: 2 to 12 lb/ft³ design
• Sorbent size: 50 to 300 micrometers typical
• Sorbent flux: 100 lb/sec-ft² design
• Reactor Vessels: Refractory lined, carbon steel
• Major Piping: Hot-walled, Incoloy 800H alloy

**Operation**

Simulated low-Btu coal gasification gas will be supplied to the PDU by a natural gas-fired fuel gas (syngas) generator. This precludes the ability to test the effects of trace contaminants on sorbent performance. This approach is more cost-effective and presents fewer site environmental issues. A previous description of the fuel gas generator remains generally accurate[5]. Notable changes include the decision to use sulfuric acid rather than sulfur dioxide as the source of hydrogen sulfide for the fuel gas, and the use of a direct water quench instead of an indirect heat exchanger for final fuel gas temperature trim. In the absorber, a sorbent, such as zinc oxide, becomes sulfided by absorbing sulfur species from the fuel-gas stream. In the regenerator, the captured sulfur in the sulfided sorbent is reacted with air, which restores or “regenerates” the activity of the sorbent. The sorbent is then recirculated back to the absorber, thus providing continuous operation. Inert gases (steam and/or nitrogen) are used to fluidize the sorbent in the standpipes above the valves and to prevent fuel gas and air intermixing.

The METC PDU is designed for operation in four distinct modes, which are combinations of either fluid bed and transport contacting on either or both the absorber and regenerator sides. Initial METC efforts will focus on transport absorption and regeneration (Mode 4) because of the similarities to the Sierra Pacific Power Company's clean coal technology project and because transport contacting offers potential economic benefits in both capital investment and operating simplicity.

The general operating strategy for the PDU will be to maintain constant flow rates, temperatures, pressures, and compositions during a specific test run. Values for independent variables will be specified prior to a run, and dependent variables will then be calculated to provide initial control setpoints. In order not to exceed design temperatures, operations will start with both the absorption and regeneration temperatures below the targeted values, and then feed temperatures will be increased. Process changes will be made gradually and the unit will be allowed to stabilize. As a general rule, the sorbent inventory will be cycled a minimum of three times between process state changes.
A stable pressure balance must be maintained for smooth, uninterrupted sorbent circulation. The absorber-regenerator differential pressure will be set to a constant to balance the hydrostatic pressure buildups in the fluid beds and standpipes versus the pressure drops across the circulation slide valves and the risers. The normal differential pressure between the absorber and regenerator freeboards is expected to be in the range of +/- 2 psi (13.8 kPa), depending upon the operating mode. The pressure drop across the circulation control slide-valves should be about 5 to 10 psi (35 - 70 kPa). The absorber-regenerator differential pressure will be maintained by modulating the regenerator backpressure valve in response to the differential pressure. To keep the differential pressure as constant as possible, this controller will be tuned for a fast response relative the controller for the absorber backpressure valve.

Sorbent flow rates cannot be measured directly. They will be inferred from pressure drops across the standpipe slide valves, which must be calibrated during startup activities. During operations, the circulation rate will be set at a targeted test value by positioning the slide valve in the absorber circulation standpipe (see Figure 1). This will establish a constant flow rate of sorbent from the absorber to the regenerator. The return flow rate of sorbent from the regenerator to the absorber will be automatically controlled by modulating the slide valve in the regenerator circulation standpipe to maintain a preset sorbent bed level in the absorber. The regenerator bed level "floats" in this scheme, but a constant sorbent feed rate is maintained to the regeneration side of the process where temperature concerns are greatest and the need for uniform sorbent flow is more critical. Recirculation rates to the transport reactors will be set by slide valve position in the recirculation standpipes.

Figure 1. PDU Reactor Schematic
The entire output of the syngas generator will be fed to the PDU during normal operations. A slipstream approach is not possible due to air permit restrictions. Delivery pressure is set by a backpressure valve on the feed line to the PDU. The fuel-gas flow rate is established and controlled by input rates to the gas generator. The major composition will be established by firing stoichiometry and the proportional amounts of injected water and carbon dioxide. The hydrogen sulfide content will be controlled by the injection rate of sulfuric acid. Nearly complete conversion of the sulfuric acid to hydrogen sulfide is expected. This air-blown, partial combustion process is projected to produce a nominal 100 to 130 Btu/scf (3,726 to 4,844 kJ/m³) gas. Although the unit will be monitored for soot generation, any soot remaining in the gas that is fed to the PDU is not expected to be a significant problem due to the relatively high moisture content of the gas (minimum about 15 mole %), the hot refractory wall temperatures (minimum 1,250 °F or 677 °C), and the long piping length from the syngas unit to the PDU absorber (approximately 200 feet or 61 meters). These three features combined are expected to promote the conversion of soot to gaseous compounds.

PDU Project Status and Schedule

Structural steel fabrication and steel erection were 100 percent complete in June 1996. The incinerator, barrier filter vessels, fines lock hoppers, sorbent feed vessel, filter blowback accumulator, and air and inert gas preheaters were mounted in the structure. As of June 1, 1996, project construction was 60 percent complete, up from 32 percent a year ago. The project is on schedule to desulfurize coal-gas by October 1997.

Future Activities

In addition to demonstrating fully integrated operations, process and control scale-up data, performance data, and addressing other actual engineering challenges, future tests will concentrate on key operational and sorbent durability issues such as: Which is more optimum from a sorbent as well as an overall process economic viewpoint; operating with small changes in sulfur loading on the circulating sorbent and thus at high circulation rate, versus running "deep" cycles with larger changes in sulfur loading but at comparatively lower circulation rates? Is bubbling, turbulent, or transport flow regime best for chemical reaction and/or sorbent life? These and other data will be generated at METC during four nominal 10-day test periods per year beginning after PDU shakedown in the fall of 1997. Initial testing will involve more numerous but shorter duration test periods. Test planning, operations, data reduction and reporting will be performed by DOE/METC personnel in partnership with industry.

Summary

The PDU offers for coupled system operation of various reactor configurations to optimize gas/solids contacting and prove system safety and control aspects. With its dedicated purpose, operating flexibility, and high degree of instrumentation; the PDU will provide design data on this highly complex HGD process over a broad operating window for about a dozen major operating variables.

At this point, the PDU is not linked to a single developer. Therefore intellectual property provided by the PDU will be available for application to other IGCC systems including those using oxygen-blown gasifiers. The expected cost savings can thereby be
realized by a much broader market than if the technology were limited in use to only one gasifier supplier. METC is interested in pursuing industrial cost sharing of PDU activities through arrangements such as cooperative research and development agreements, whereby intellectual property rights can be obtained by the industrial partner.

References


