

SECTION 1 BACKGROUND AND PRIOR WORK

Designs for advanced integrated-gasification-combined-cycle (IGCC) power systems call for desulfurization of coal gasifier gas at high-temperature, high-pressure (HTHP) conditions using highly efficient, regenerable metal oxides such as zinc titanate. Regeneration of the sulfided sorbent using an oxygen-containing gas stream results in a sulfur dioxide (SO₂)-containing offgas at HTHP conditions. The patented Direct Sulfur Recovery Process (DSRP) (USP 5,366,717 and USP 5,798,088) developed by the Research Triangle Institute (RTI) with support from the National Energy Technology Laboratory (NETL) and its precursor organizations (Morgantown Energy Technology Center [METC] and Federal Energy Technology Center [FETC]), is an attractive option for treating this regeneration offgas (ROG). Using a slipstream of coal gas as a reducing agent, it efficiently converts the SO₂ to elemental sulfur, an essential industrial commodity that is easily stored and transported. Figure 1 is a schematic diagram showing a proposed commercial embodiment of DSRP.

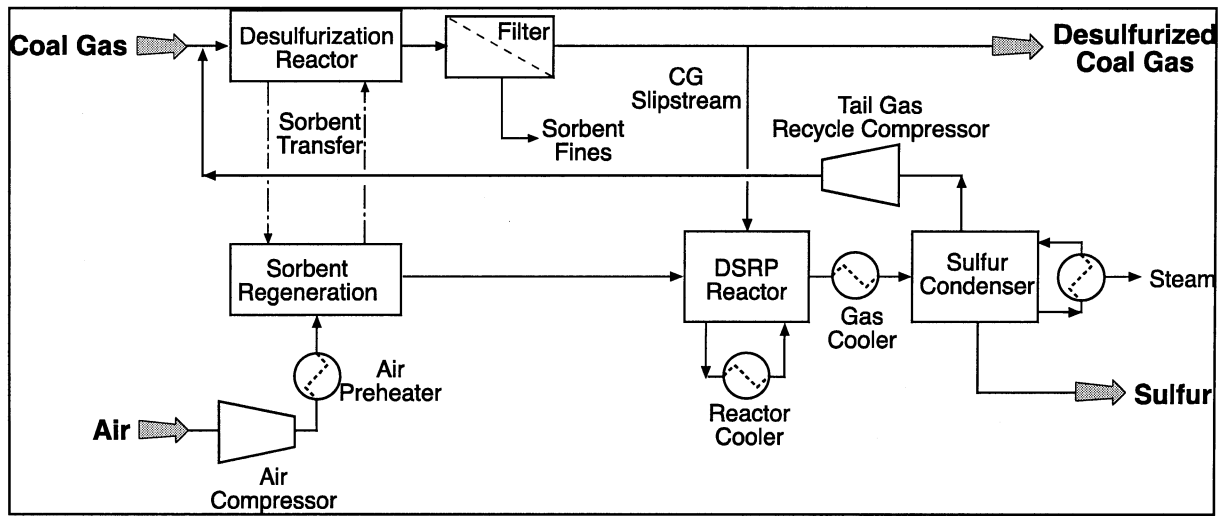


Figure 1. Hot-Gas Desulfurization/DSRP Integration

Prior to the current contract, the DSRP was developed in a laboratory setting, using synthetic gas mixtures to simulate the ROG and coal gas feeds (See McMichael & Gangwal, 1991; Gangwal & Chen, 1994). Work done in the first five tasks for this contract is described in detail in Appendices A and B. Under these tasks, the DSRP was tested using actual coal gas and actual ROG. One of the main objectives was to test the integrated system over an extended period with actual coal gas from an operating gasifier to quantify the degradative effect, if any, of the trace contaminants present in coal gas.

In order to test with actual coal gas, RTI designed and fabricated a mobile laboratory containing a bench-scale, integrated hot-gas desulfurization/DSRP unit. The 75-mm (3-in) fluidized-bed desulfurization reactor was used to test the U.S. Department of Energy (DOE)/RTI-patented zinc titanate-based fluidizable sorbent formulation, ZT-4L, and to produce an “actual” ROG stream. The mobile lab was installed at the NETL-Morgantown site and testing was conducted with a slipstream of coal gas from the pilot-scale gasifier

located there. Three separate slipstream test campaigns plus an additional exposure test took place over a period of two years:

- September 1994: Initial shakedown test of mobile laboratory at Morgantown with integrated desulfurization reactor and two-stage DSRP
- October 1994: Test run of integrated desulfurization reactor and two-stage DSRP at Morgantown
- July 1995: Long-duration run (160 h) slipstream test of single-stage DSRP at Morgantown
- March-April 1996: DSRP catalyst exposure to pure coal gas at the General Electric (GE) pilot plant in Schenectady, New York
- April-May 1996: Operation of RTI laboratory DSRP unit to test the exposed DSRP catalyst.

The 1994 slipstream testing included testing of the ZT-4L sorbent. During a run of 4 days' duration in October 1994, the ZT-4L was subjected to three sulfidations and two regenerations. The ZT-4L consistently removed H₂S from coal gas down to <20 ppmv at 873 K (1110 °F) and 21.89 MPa (260 psig). The DSRP was very effective in converting SO₂ in actual or in synthetic ROG to elemental sulfur, achieving 95% to 99% conversion after the first stage of the two-stage bench unit DSRP test rig. The overall conversion of the two-stage unit was less than that achieved in the first stage alone; the undesirable "reverse Claus" reaction was believed to be the problem. The results of the initial 1994 tests were encouraging and led to the decision to refit the mobile laboratory with a single-stage DSRP unit (Figure 2) and with new control hardware and software to improve the stoichiometric flow control of the coal gas stream.

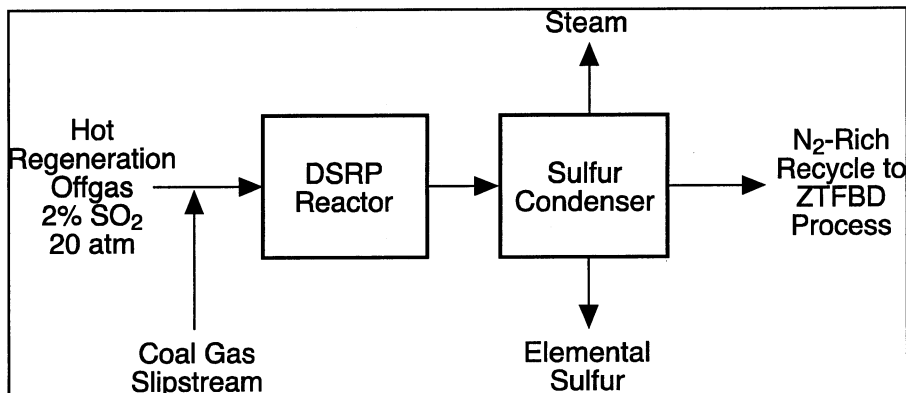


Figure 2. Single-Stage DSRP

In the 1995 slipstream test campaign, the single-stage unit produced 98% conversion of SO₂ to elemental sulfur at both the beginning and end of the run. Thus, 160 h of exposure of the catalyst to coal gas resulted in no detrimental effect. The apparatus used in that field test included a "first generation" automatic coal gas flow control system, designed to maintain the coal gas at the desired stoichiometric ratio to the SO₂ in the ROG. Though

insufficiently rugged for long-term use, that scheme greatly enhanced the operators' ability to attain and maintain steady-state operation of the DSRP reaction.

To accelerate the exposure of the catalyst to the trace contaminants present in actual coal gas, the "used" catalyst was removed from the Mobile Laboratory (then at Morgantown) and shipped to the GE pilot plant for placement in a coal gas line throughout a 10-day pilot plant run. This action resulted in additional exposure of the catalyst during March-April, 1996 to about 200 h of coal gas, roughly equivalent to 1,330 h of exposure at the DSRP conditions used at the Morgantown site. Thus, total exposure of the catalyst including the Morgantown testing was approximately 1,500 h.

The exposed catalyst was tested in April-May, 1996 in a 75-mm (3-in) bench-scale reactor using synthetic mixtures of feed gases and simulated coal gas. During the laboratory testing, an "induction period" was observed, as the conversion steadily improved with increasing run time. It appeared that a surface-cleaning phenomenon was occurring, leading to removal of impurities and improved activity. The conversion to sulfur was 96% after approximately 22 h of testing, compared to 98% during the slipstream testing. It was hypothesized that the induction period was due to removal of tar and soot buildup on the catalyst as received from GE. Subsequent testing of the catalyst showed that the bench-unit test program had indeed reduced the carbon content. The overall conclusion is that the DSRP catalyst is quite rugged in the presence of tar-laden actual coal gas, even after 1,330 equivalent hours of exposure.

Task 6 of this contract involved the design and construction of a DSRP test unit that had six times the capacity of the bench-scale unit. Designated the 6X DSRP, this unit was initially designed for use at Enviropower's European test site, and the design was strongly influenced by the specific site requirements. Subsequent to the start of construction, the partner's cooperative research and development agreement (CRADA) with DOE dissolved; a search for an alternate test site commenced.

As originally conceived, the 6X DSRP was skid-mounted and sized to accommodate shipping in a standard overseas shipping container. When it became clear that the DOE/NETL Power Systems Development Facility (PSDF) in Wilsonville, Alabama, would be the chosen test site and that over-the-road shipping would be possible, design changes had to be incorporated. The major impact was that the Mobile Laboratory (described above) would be remodeled to be used as a control and analytical space and the 6X DSRP would be positioned adjacently, but outdoors. With these design changes, the nomenclature for the apparatus also changed, from 6X DSRP to "skid-mounted DSRP field-test unit."

This report describes the completion of fabrication of the skid-mounted DSRP field-test unit and its subsequent commissioning at PSDF.