DEVELOPMENT AND TESTING OF A WATER-GAS SHIFT REACTION CATALYTIC FILTER ELEMENT IN GASIFICATION PROCESS

Xiaofeng Guan, Pannalal Vimalchand, Alexander Bonsu, Subhash Datta, and Wanwang Peng
Southern Company
Power Systems Development Facility
31800 Highway 25 North, Wilsonville, AL 35186, USA

ABSTRACT

A catalytic filter element has been developed at the Power Systems Development Facility combining a water-gas-shift reaction process with particulate removal within a filter element. A prototype of such catalytic filter element has been evaluated in a proof-of-concept test in a recent gasification test run. The catalytic filter element was tested under a wide range of operating conditions in a particulate free syngas. The preliminary test showed promising results. A CO conversion rate of 14%-84% was achieved at 533-728 K (500-850°F) temperature range. This paper serves as a progress report for the development and initial testing. Further modifications have been made and more detailed testing is underway.

INTRODUCTION

The Power Systems Development Facility (PSDF) is an engineering scale test facility of advanced coal-fired power systems, high-temperature and high pressure gas filtration systems, and gas cleanup systems. The PSDF was designed at sufficient scale so that advanced power systems and components can be tested in an integrated fashion to provide representative data for commercial scale-up. The PSDF is funded by the U.S. Department of Energy, the Electric Power Research Institute, Southern Company, Kellogg Brown & Root, Inc. (KBR), Siemens Power Generation, Peabody Energy, Burlington Northern and Santa Fe Railroad, and the Lignite Energy Council. The transport reactor has been operated successfully on sub-bituminous, bituminous, and lignite coals as a pressurized combustor and as a gasifier in both oxygen- and air-blown modes and has exceeded its primary purpose of generating gases for downstream testing. Latest test results are summarized in References [1], [2], and [3]. Test run reports and published papers can be found on the PSDF website [4]. With a continuous interest in coal utilization and growing sense of urgency in carbon dioxide (CO_2) control, the PSDF has shifted its focus to CO_2 capture technology. The primary objective is to develop, evaluate, and facilitate the commercialization of emerging carbon control technologies. The PSDF has set up a flexible and cost-effective process system to test gas cleanup and CO_2 capture technologies.

One of the newly-developed technologies at the PSDF incorporates a water-gas shift (WGS) reaction catalyst into a particulate filter element to form a catalytic filter element. In a carbon constrained scenario, coal gasification for power generation will be achieved by a process that incorporates WGS. In the scenario, after syngas is generated in the gasifier and filtered to remove particulate matter, the syngas undergoes WGS reaction whereby carbon monoxide (CO) in the syngas reacts with steam to produce hydrogen (H_2) and CO_2 as follows:

$$CO + H_2O \Leftrightarrow CO_2 + H_2 \tag{1}$$

The CO_2 would be separated from the main gas stream and pressurized for storage or sequestration. The main gas stream, rich in H_2, would then be sent to the gas turbine for power production.

In our catalytic filter element arrangement, particulate filtration and the WGS process are combined to eliminate or reduce capital and operation and maintenance (O&M) costs. The catalytic filter elements can be used as a stand alone or polishing process for the WGS reaction. The main benefit, however, is to fully utilize the significantly large surface area of the filter elements in a filter system. With a properly installed catalyst layer inside the filter element, the syngas is uniformly distributed through the catalyst layer with a slow face velocity. This arrangement

ensures a high efficiency WGS reaction with the near-perfect flow pattern and effective gas-catalyst contact. Conventional WGS process is conducted in fixed bed reactors. A typical plant requires a number of sizable WGS reactors [5]. Compared to a fixed bed WGS reactor, the catalytic filter offers a lower pressure drop and catalyst-to-gas ratio. In addition, the operating conditions (temperature, pressure, etc.) of a particulate filter system are favorable for the WGS reaction, thereby allowing the WGS catalyst to be readily incorporated into the filter element without significant modification to the filter system.

CATALYTIC FILTER DEVELOPMENT AND TEST SETUP

Catalytic filter elements have been used for tar cracking at 600-900°C in a model biomass gasification gas [6] and NOx removal at about 300°C in a flue gas cleaning system [7]. In these cases, active catalyst is incorporated in the ceramic filter element either by impregnating the catalyst to the thick filter wall or installing a particle layer catalyst inside the filter cavity. In an Integrated Gasification Combined Cycle (IGCC) plant, the syngas filtration process is conducted in a particulate control device (PCD) usually at a warm temperature (about 315°C). At this warm temperature, there are a variety of metal alloy materials that can be used to make filter elements. In general, metal filter elements are relatively more robust and durable than ceramic materials particularly under thermal shocking conditions. Also, metal filter elements have a thinner wall compared to ceramic filter elements. The thin wall makes the inside cavity larger, which in turn allows for installing larger volume of catalysts inside the filter cavity. Based on these considerations, our development is focused on incorporating catalyst powder layer inside metal filter elements.

The current development is in its prototype stage. As a first step, a section of a Pall iron aluminide metal filter element with a built-in failsafe fuse has been modified to accept catalyst material. The failsafe serves as an inner porous tube so that the catalyst powder can be used to fill the gap between the filter wall and the inner tube. In a real filtration process system, the inner tube would still act as a failsafe to block particulate leak passage in case of a filter element failure. The first “proof-of-concept” test of the prototype catalytic filter element was conducted during a recent coal gasification test run at the PSDF. A commercially available sour WGS catalyst was installed after being ground and sieved to 180-500 micron sizes.

The filter element prototype was installed in a vessel in the syngas cleanup unit (SCU), which is a slipstream test unit downstream of the main gasification process system. The SCU is located downstream of the particulate control device (PCD) in the main process. Therefore, the syngas is particulate free. For this proof-of-concept test, the particulate free syngas was used as a first step approach. The PSDF gasification process and the SCU system are shown schematically in Figure 1.

The catalytic filter element was connected to outside via a 1.27 cm (½ inch) tubing. The installation was arranged in such a way that the syngas and steam flowed from the bottom of the vessel and entered the filter element from outside surface. The WGS reaction occurred in the catalyst layer. The product gas collected inside the filter and exited vessel through the tubing. A schematic process flow diagram and a picture for the catalytic filter installation are shown in Figures 2 and 3.

The inlet gas compositions were analyzed by Applied Automation Optichrome Advance process GC, and the outlet gas compositions were analyzed by Siemens Maxum I process GC. The typical syngas compositions during an operating period with Powder River Basin (PRB) coal are summarized in Table 1.

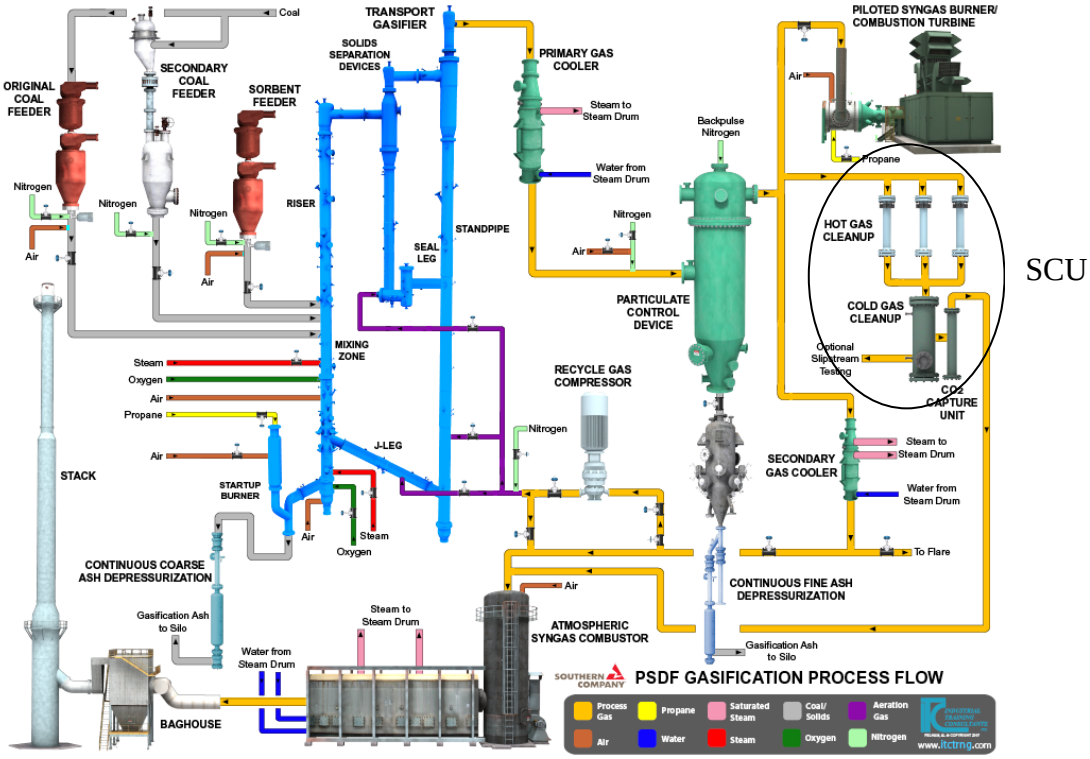

Figure 1. PSDF gasification and syngas cleanup unit.

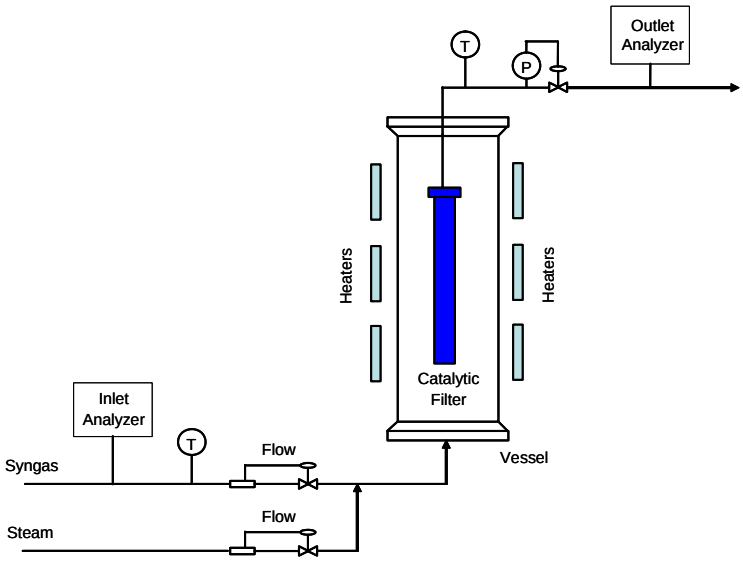

Figure 2. Simplified process flow diagram

Figure 3. Picture of the catalytic filter element.

Table 1. Typical syngas compositions

	Composition (vol %)
H_2O	9
CO	9
CO_2	8
H_2	7
CH_4	1.5
N_2 and Others	Balance

The test was conducted at 1076-1517 kilopascal (156-220 psig) pressure and 533-728 K (500-850°F) inlet temperature. The syngas flow rate and extra steam flow rates were controlled at 2.7-13.6 kg/h (6-30 lb/h) and 0-2.7 kg/h (0-6 lb/h), respectively. The face velocity on the filter element was 0.32-1.85 cm/s (0.63-3.65 ft/min). These operating conditions were determined by the following criteria: (1) the face velocity on the catalytic filter element should be in an approximate range of the actual face velocity on the filter elements in a typical particulate filtration system, and (2) a relatively wide range of operating parameters were explored in this proof-of-concept test to evaluate the performance for further development and improvement. The inlet temperature variation was achieved by external heaters on the vessel, which can heat inlet gas to a higher temperature. The WGS is an exothermal reaction resulting in higher temperature in the product gas. Temperature measurements were also made at the outlet to help evaluate CO conversion.

RESULTS AND DISCUSSION

The CO conversion rate is plotted in Figure 4 as a function of average residence time in the catalyst layer. At 533 K (500°F), the CO conversion rate was 14%-34% at 0.5-2.0 seconds of residence time (excluding an outlier of 56%). When the temperature was raised to 700-728 K (800-850°F), the CO conversion rate was 54-84% in the range of 0.3-0.7 seconds of residence time.

It should be noted that the catalyst tested in this proof-of-concept test was not pre-sulfided. As a result, the activity of the catalyst was lower than expected. A higher temperature of 700-728 K (800-850°F) was used to increase the CO conversion rate. It is desired that the inlet temperature is at 550F for the WGS reaction in a typical plant [5]. With the pre-sulfidation treatment for the catalyst, the CO conversion rate would be increased to a high level at a relatively lower temperature. This issue will be addressed in the next test run.

In general, the CO conversion rate correlated well with the residence time. Since the catalyst layer inside the filter element is thin, the residence time is critical for the reaction. However, a good

conversion rate was obtained during this short residence time, which is corresponding to a typical face velocity on the filter element in a particulate filtration system.

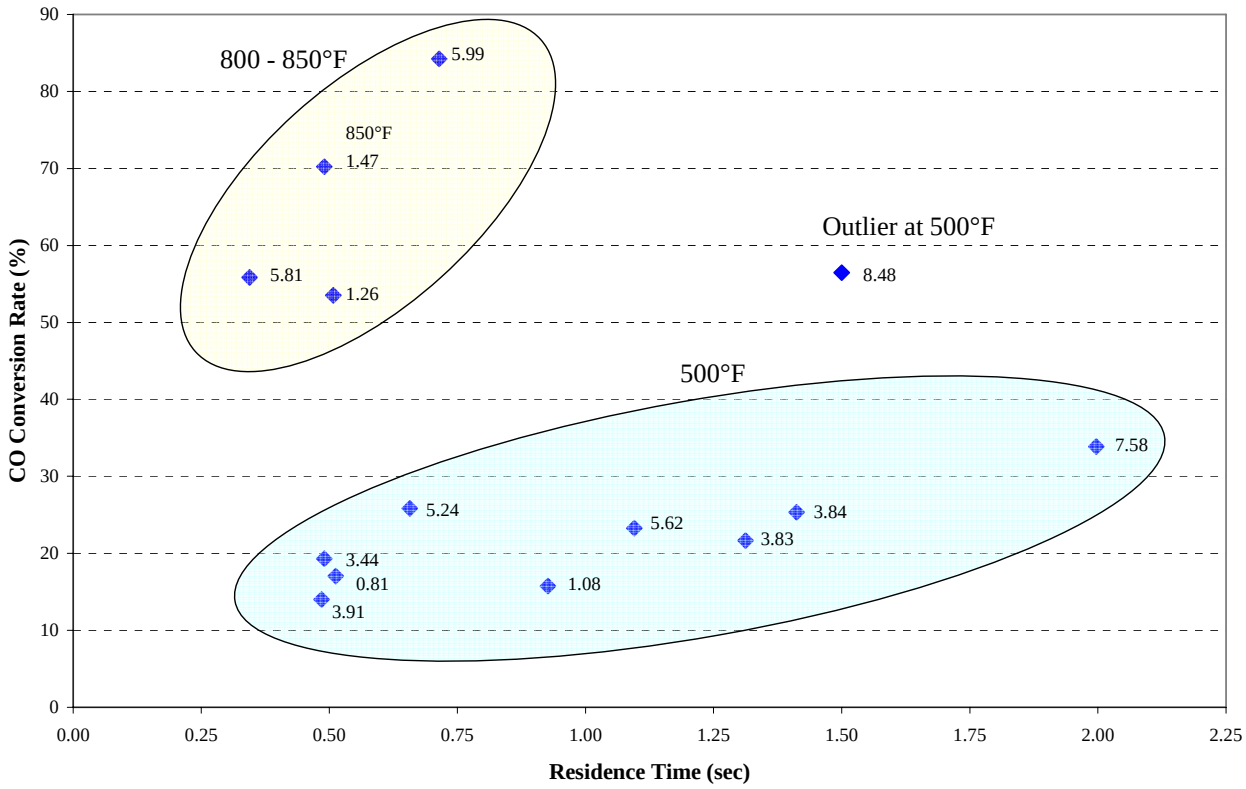

Figure 4. CO conversion rate. (The number near the data point indicates H_2O to CO molar ratio in the inlet gas mixture.)

In this test, the moisture content in the wet syngas was about 9%, while the CO content was 8-12%. In some cases, the H_2O/CO ratio in the wet syngas was already high enough for the WGS reaction without injection of extra steam although extra steam injection is in favor for conversion towards right direction in Equation (1). For the syngas compositions in this test, the concentration ratio $([CO_2]\cdot[H_2])/([CO]\cdot[H_2O])$ is significantly lower than the equilibrium concentration ratio in this relatively low temperature range, which enabled the fairly high CO conversion rate. However, a wide range of H_2O/CO ratio was used to evaluate the catalytic filter's performance. In this particular configuration, the influence of the H_2O/CO ratio was not as strong as temperature.

MODIFICATIONS AND FUTURE TEST PLAN

In an ongoing test run scheduled in Spring 2008, two modified catalytic filter elements have been installed with finer catalyst particle size (106-212 microns) and a thicker catalyst layer in a new filter internal configuration. Two commercially available sour WGS catalysts were installed in these two catalytic filter elements separately. The catalysts were pre-sulfided per manufacturers' recommendation. Parametric testing will be conducted in this test run to further optimize the design and performance of the prototype. The development goal is to achieve the necessary CO conversion with a face velocity well within the operating range for particulate filtration. The filter element will eventually be tested with particulate removal having online back-pulse cleaning to determine the combined effect of gas filtration and WGS reaction. The PSDF has also proposed to

apply this concept to multiple-functional catalytic filter elements to achieve other gas cleanup and gas separation processes within a particulate filter element.

SUMMARY

A WGS catalytic filter element has been developed at the PSDF. The proof-of-concept test was conducted with a real particulate free syngas from a gasification process. A wide range of operating conditions was used to evaluate the performance of the catalytic filter and to prove the concept. The test showed promising results as the initial design achieved fairly good CO conversion rate. Based on the preliminary testing results, modifications have been made to further improve the design and performance. More testing is underway with this new configuration. The detailed results will be reported in the near future.

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