

4.4.2 Equipment Description

The reactor system used to test the ammonia decomposition catalyst is part of a skid-mounted pilot plant in RTI's mobile laboratory. Operation of this pilot plant depends on an external source of coal gas. The 10-in. fluidized-bed coal gasifier located at DOE/FETC's Morgantown facility currently provides the gas source.

The corrosive nature of sulfur gases particularly in the presence of steam requires the use of either 316 or 304 SS components. Corrosion resistance of critical parts is enhanced by Alon processing (a high-temperature aluminum vapor treatment). Figure 11 is a simplified schematic of the reactor system. This reactor system contains the following features: (1) gas conditioning, (2) a fixed-bed reaction chamber, (3) process control, (4) data acquisition, (5) gas and condensate sampling, and (6) reactor off-gas venting. Each of these features is discussed briefly below.

Temperature of the coal gas feed is maintained with Temptrace[®] metal-sheathed mineral-insulated heat tracing and Omega temperature controllers. In addition to heating the coal gas, coal or ash fines being conveyed with the coal gas are removed with a Mott GasShield[™] filter. Specifications for this filter claim it removes >99.999999 percent of all particles down to 0.01 μm .

The reactor is constructed of 4-in. schedule 160 316 SS pipe and is identical to the bench-scale reactor at RTI (Figure 2). A unique feature of this reactor is the removable cage allowing easy loading and unloading of the catalyst. The reactor can accommodate either a 7.62 cm (3 in.) or a 5.1 cm (2 in.) dia cage. A removable α -alumina distributor plate is positioned at the bottom of each cage to introduce hot coal gas into the reactor. The reactor is housed inside a three-zone furnace equipped with a separate temperature controller for each zone.

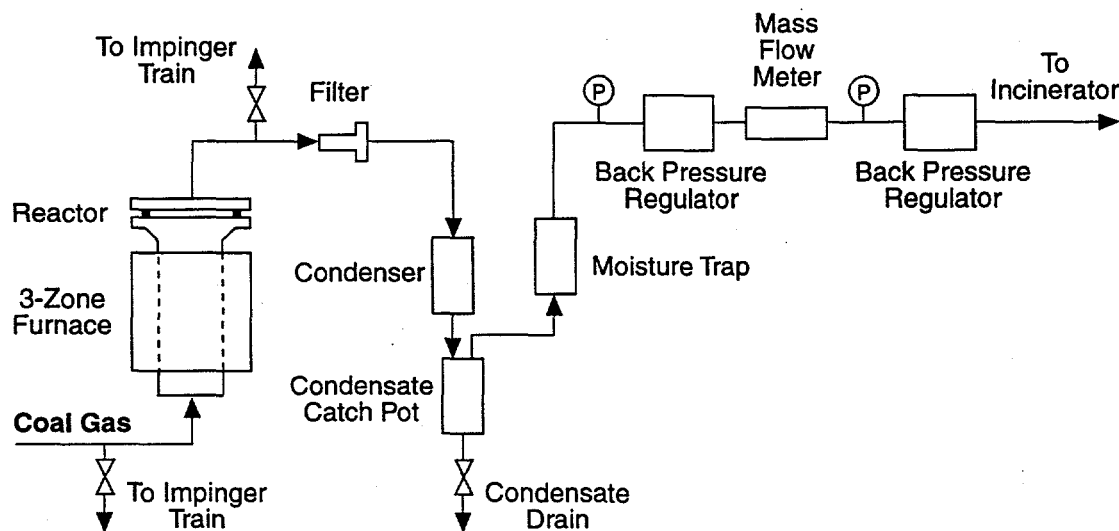


Figure 11. Schematic of skid-mounted reactor system.

Reactor pressure is controlled by two BPRs connected in series. One is upstream of an electronic mass flow meter (MFM), and the other is downstream. This configuration maintains reactor pressure at 150 psig and limits the pressure drop across the MFM to within the specified operating range. The signal from the MFM serves as an input for a PID controller used to maintain the coal gas flow rate at 68 slpm with a Badger valve

An Omega OM480 data logger and LabTech Notebook software record temperature, flow rates, and pressures for the entire mobile laboratory. For this reactor system, stored data include flow rates, reactor temperatures, and stream or skin temperatures needed to monitor or evaluate system performance.

Small slipstreams of coal gas are periodically removed before and after the reactor. The ammonia contained in these streams is captured in an acid impinger train. Each impinger train consisted of three impingers and a dry gas test meter. The first two impingers are filled with 15 mL of 0.1 N hydrochloric acid. The third impinger is filled with 75 to 80 mL of silica gel. The dry gas meter measures the volume of coal gas tested. Slipstreams pass through the acid impinger trains for approximately 1 h at 1 slpm at 6-h intervals.

A ceramic filter is used to capture particles elutriated from the reactor. After filtration, the coal gas is cooled in a double-pipe heat exchanger with cold process water in the outer shell. Condensate formed during this cooling stage is periodically drained from a catch pot into a measuring flask. The cold reactor effluent is dried in a moisture trap containing Drierite[®]. After passing through the BPRs and MFM, the cold effluent is vented to an incinerator.

4.4.3 Test Conditions and Sampling Procedures

- Reactor Loading

For the 100-h test of the ammonia decomposition catalyst, HSTR-1, a 7.62 cm (3 in.) cage was used. The reactor cage was initially loaded with beryl saddles to a depth of 25.4 cm (10 in.). Approximately 820 cm³ or 1,539 g of HSTR-1 catalyst was loaded on top of the beryl saddle bed. The beryl saddle bed ensured the coal gas was at temperature before reaching the catalyst. The height-to-diameter ratio for the catalyst bed was 2.3. The gas hourly space velocity for the catalyst bed, assuming a coal gas flow rate of 68 slpm, is about 5,000 h⁻¹. The temperature in the catalyst bed was measured by a thermocouple with its tip 10.16 cm (4 in.) below the catalyst surface, roughly the middle of the catalyst bed.

- Ammonia Sampling

After the acid impinger train had been exposed to a slipstream for the required period, the content of each acid impinger was emptied into its own sample bottle. Any residual acid solution containing ammonia in the impinger or connecting tubing was rinsed out and into the appropriate sample bottle with distilled water. The content of the sample bottle was diluted to form 30 mL of solution with distilled water. Each sample bottle was carefully labeled to identify data, time, and impinger from which the sample was removed. These were the standard procedures for preparing samples for ammonia testing. If the silica gel had been

Table 8. Average Wet Coal Gas Compositions During 100-h Test

	Wet coal gas composition	
	Wt%	Vol%
CH ₄	1.86	2.87
H ₂ S	0.18	0.13
H ₂	1.14	14.14
Ar	0.78	0.48
CO ₂	19.92	11.21
CO	13.42	11.87
N ₂	54.86	48.50
H ₂ O	7.61	10.47
NH ₃	0.23	0.33
Total	100	100

During the first 40 h of operation the results obtained from ammonia in the acid impinger trains were extremely low. This initiated the collection of condensate samples to provide an alternative measurement of ammonia concentration in the gas stream. Removal of the filters used to condition the slipstreams generated acid samples in which ammonia concentrations were in the anticipated range and reasonably close to values determined from condensate samples. Ammonia concentrations for both impinger and condensate sampling are provided in Table 9. Some of the difference observed between the two methods results because only one condensate sample for the raw coal gas was collected each day by FETC personnel. Other potential sources of discrepancy between the two methods are not as easily identified.

Ammonia conversion was calculated from the changes in ammonia concentration in the gas stream entering and leaving the reactor. Ammonia conversion and reactor operating conditions are presented in Table 10. A visual picture of ammonia conversion as a function of operating time is shown in Figure 12. Average ammonia decomposition for HSTR-1 based on impinger sampling was 91.7 and 87.4 percent based on condensate sampling. During the 60 h of operation for which reliable data were available, no deactivation of the catalyst was observed.

exhausted or would be exhausted during the next sampling period, the silica gel was replaced.

In addition to impinger samples, samples of the condensate were collected twice a day for ammonia analysis. A condensate sample was also collected for the raw coal gas by FETC personnel on a daily basis. Condensate sampling began after ~40 h of operation to check and confirm ammonia concentrations being obtained by impinger sampling.

Ammonia analysis was performed by FETC personnel using a Dionex Series 4000 ion chromatograph with an IonPac[®] CS14 column. The eluent was 10 millimolar methane sulfonic acid at a flow rate of 2 mL/min. Detection was by conductivity.

4.4.4 Results

The average composition of the coal gas during this 100-h test is provided in Table 8.

Table 9. Ammonia Concentration in Coal Gas as Determined by Impinger and Condensate Sampling

Cumulative time of operation (h)	Ammonia concentration (ppmv)			
	Impinger sampling		Condensate sampling	
	Before reactor	After reactor	Before reactor	After reactor
40.83	2,128	315		
42.30	3,311	167		
46.58			3,009	442
50.83	3,641	131		
52.25			3,009	238
53.67	2,712	170		
62.00			3,009	487
63.73	3,564	199		
71.70	3,391	173		
74.00			3,273	310
80.22	4,016	478		
86.00			3,273	549
88.50	3,627	629		
94.77	3,425	149		
98.00			2,251	239

Table 10. Ammonia Conversion and Reactor Operating Conditions

Cumulative time of operation (h)	Ammonia conversion (%)		Reactor temperature (°C)	Reactor pressure (psig)	Gas hourly space velocity (h ⁻¹)
	Impinger	Condensate			
40.83	85.18		794	143	4,975
42.30	94.94		777	146	4,975
46.58		85.31	784	148	4,975
50.83	96.40		795	134	4,975
52.25		92.09	784	138	4,975
53.67	93.70		769	153	4,975
62.00		83.81	777	150	4,975
63.73	94.42		777	149	4,975
71.70	94.90		790	149	4,975
74.00		90.53	786	148	4,975
80.22	88.11		785	146	4,975
86.00		83.25	771	148	4,975
88.50	82.65		778	146	4,975
94.77	95.66		795	145	4,975
98.00		89.36	787	147	4,975

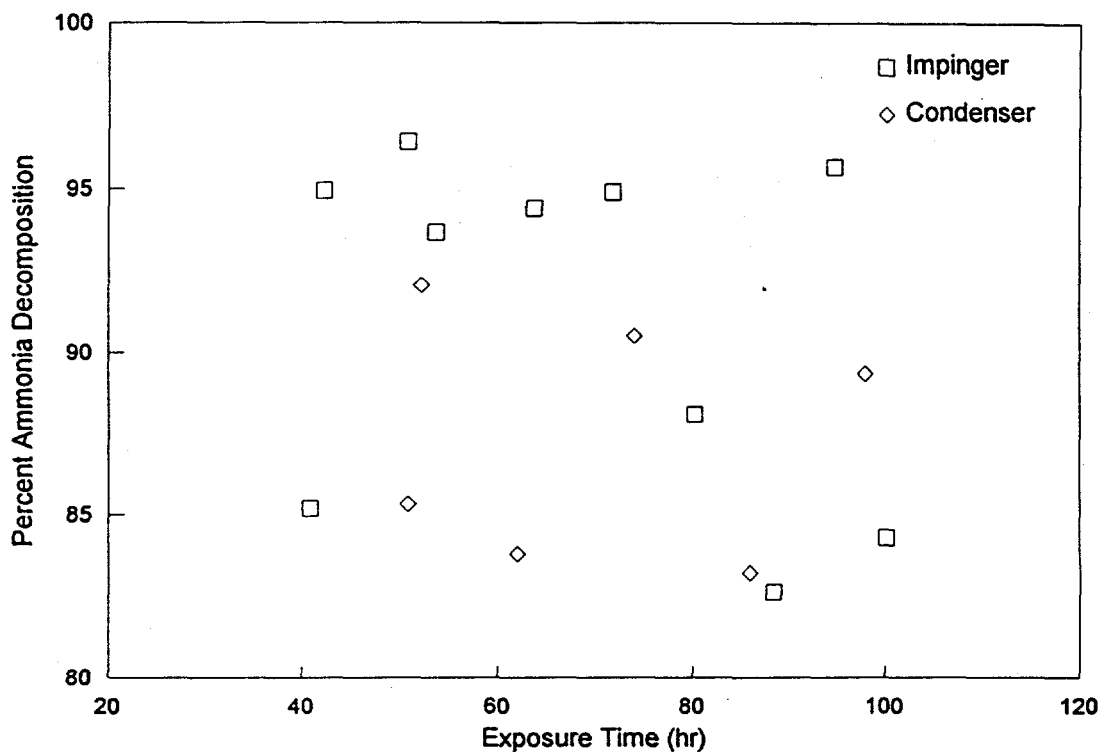


Figure 12. Performance of ammonia decomposition catalyst as function of exposure time.

4.4.5 Conclusions

Ammonia decomposition catalyst, HSTR-1, was exposed to real coal gas for 100 h at an average temperature of 780 °C, pressure and 146 psig, and gas hourly space velocity of 4,975 h⁻¹. During this test period, ammonia decomposition averaged 91.7 percent based on impinger sampling and 87.4 percent based on condensate sampling. Over the last 60 h for which reliable data were available, no catalyst deactivation could be detected.

5.0 CONCLUSIONS AND RECOMMENDATIONS

The efforts to develop a successful combination of an NH_3 decomposition catalyst with an H_2S sorbent were not successful. Numerous combinations containing catalysts based on Ni, Co, Mo, and W with zinc titanate exhibited low-to-moderate activity for NH_3 decomposition.

For the high-temperature catalytic decomposition approach, the HTSR-1 catalyst, a nickel-based catalyst on a refractory support, gave excellent results. The two 100-h tests with HTSR-1 demonstrated that it is capable of decomposing up to 90 percent of the NH_3 present in typical coal gasifier gases.

Further work is needed to reduce the cost of the catalyst and develop it in a form, such as monolith, that could operate in the presence of particles. Also continued research is needed for development of catalysts that would work at relatively lower temperatures down to 500 °C. Fundamental studies of what catalyst forms are active for NH_3 decomposition, what causes catalyst poisoning, and how it can be minimized could lead to the development of a successful combined sorbent-catalyst.

6.0 REFERENCES

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