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INTRODUCTION

A means of recombining the radiolytic off gas is necessary in the operation of homogeneous reactors. The natural circulation recombiner was built to check the feasibility of a gas recombiner operating without a blower or external evaporator to supply diluent. The highly explosive nature of hydrogen and oxygen mixtures (assuming the explosive limit to be 15% stoichiometric H_2 and O_2) can be kept safe by using a suitable diluent.

SUMMARY

The recombiner consists of a cylindrical shell which houses a flue or chimney containing a catalyst bed, cooling coil and heaters. A metered quantity of hydrogen and oxygen is introduced into the annular space and recombination takes place as the gas mixture is swept through the catalyst by convection. The unit operated smoothly with a hydrogen flow of 1.06 scfm, using steam diluent at 100 psia. Using air diluent at 15 psia, the unit operated satisfactorily with hydrogen flow up to 0.20 scfm.

DESCRIPTION OF APPARATUS AND OPERATION

The low pressure natural circulation recombiner test unit (Fig. 1) consisted of a 12 inch schedule 80 pipe, 8 feet long, housing a 4 inch stainless steel chimney 6 feet in length. A 6 inch roll of platinized Yorkmesh catalyst was placed at the lower end of the chimney. Three 450 watt jacketed heaters were installed under the catalyst to maintain a dry catalyst prior to startup and initiate thermal convection. An air cooled condenser, containing approximately 28 feet of 1/2" copper water tubing, was coiled in concentric fashion about the annular space at the top of the chimney. A liquid level probe was provided to maintain a predetermined water level by opening a solenoid drain valve as required. Thermocouples were spotted at various points on the unit for control purposes and temperature recording. The test unit was insulated with a 3 inch thickness of magnesia to reduce heat losses.

For startup operation, the recombiner unit was pressured to 85 psig with a building steam source. The heaters were turned on and approximately an hour was required to reach equilibrium and to obtain satisfactory convective circulation in the system. To simulate off gas introduction, metered quantities of hydrogen and oxygen were fed into the system at approximately the midsection of the unit. The gases were introduced at this point to give the maximum mixing with the diluent before being swept through the catalyst. The recombination of the gases ($2 \text{ H}_2 + \text{O}_2$) was indicated by immediate temperature rise of the thermocouple located adjacent to the catalyst. The heaters and steam source were cut off. The operating pressure of 100 psia was maintained by controlling the air coolant flow. The condensate formed was kept at constant volume by the liquid level probe.

RESULTS

The natural circulation recombiner operated smoothly, using steam diluent, with hydrogen flow up to 1.06 scfm and operating pressure of 100 psia. For the initial run, hydrogen and oxygen were added in small quantities (0.265 scfm and 0.133 scfm, respectively) and the flow rates were gradually increased to the maximum capacity of the flow meter (1.06 scfm of H_2). On subsequent runs, the maximum flow rate was introduced at once without causing flashbacks. The maximum flow rate corresponds to a hydrogen concentration of 3.81 mol per cent. The maximum temperature attained in the chimney was 792°F .

Air was substituted for steam as diluent on later runs. The unit operated satisfactorily at 15 psia with hydrogen flow up to 0.20 scfm. Further increases in hydrogen flow resulted in flashbacks. The hydrogen concentration corresponds to 3.5 mol per cent and the maximum chimney temperature was 597°F .

Plot of steam diluent flow as a function of hydrogen flow (Fig. 2) indicated the recombiner steam diluent flow to be approximately 20 per cent greater than the design condition. The steam flow was calculated from experimental data as shown in the appendix. Likewise, the plot of concentration (mol %

stoichiometric gas) as a function of hydrogen flow (Fig. 3) indicated the concentration of the gas mixture during the test operation to be approximately 20 per cent less than the design case. The two plots are indicative of the recombiner having capacity greater than design condition, which can be attributed to the conservative pressure drop correlation used.

Tabulation of hydrogen, oxygen, steam diluent flows and mol per cents of hydrogen and ($2 \text{ H}_2 + \text{O}_2$) are given in Table 1.

<u>TABLE 1</u>							
H_2		O_2		H_2O		Mole Per Cent	
<u>scfm</u>	<u>mol/min</u>	<u>scfm</u>	<u>mol/min</u>	<u>lb/min</u>	<u>mol/min</u>	<u>H_2</u>	<u>$(2 \text{ H}_2 + \text{O}_2)$</u>
0.265	.000741	.133	.000375	.863	.0477	1.52	2.29
0.532	.00148	.266	.00075	1.12	.0622	2.30	3.46
0.798	.00222	.399	.00112	1.25	.0694	3.05	4.58
1.06	.00296	.532	.0015	1.32	.0732	3.81	5.24

CONCLUSION

Satisfactory operation of the natural circulation test recombiner merits consideration in future design of gas recombiners.

Recommendations for future gas recombiner design are:

- 1) To provide a shield about the chimney to minimize heat losses.
- 2) To install the heater inside the chimney for maximum heating efficiency.

APPENDIX

Design Calculation

The design basis is the following:

- 1) Concentration of ($2 \text{ H}_2 + \text{O}_2$) at catalyst; 5 mole per cent
- 2) Diluent; steam
- 3) Desired capacity; 1 scfm
- 4) Operating pressure; 100 psia

With design flow of 1.0 scfm ($2 \text{ H}_2 + \text{O}_2$), the inlet and outlet conditions will be:

Inlet Conditions

	<u>Mol Wt.</u>	<u>Flow</u>	<u>Flow</u>	
H ₂	2	0.67 scfm	0.00185	lb mol/min
O ₂	32	0.33 scfm	0.00092	"
H ₂ O	18	19.00 scfm	0.05290	"

Exit Conditions

	<u>Mol Wt.</u>	<u>Flow</u>	<u>Flow</u>	
H ₂ O Formed	18	0.67 scfm	0.00186	lb mol/min
H ₂ O Diluent	18	<u>19.00</u> scfm	<u>0.05290</u>	"
		19.67 scfm	0.05476	

The heat of recombination is 104,000 Btu/lb mol at 18°C and 1 atm. ⁽¹⁾

Then, the heat input from recombination is

$$(104,000 \frac{\text{Btu}}{\text{lb mol}}) (0.00186 \frac{\text{lb mol}}{\text{min}}) = 193.5 \text{ Btu/min}$$

Since,

$$0.05476 \frac{\text{lb mol}}{\text{min}} \times \frac{18 \text{ lb}}{\text{lb mol}} = 0.986 \text{ lb/min}$$

Then, ΔH due to recombination is

$$193.5 \frac{\text{Btu}}{\text{min}} \div 0.986 \frac{\text{lb}}{\text{min}} = 196.3 \text{ Btu/lb of H}_2\text{O formed}$$

The temperature rise across the catalyst was obtained by consideration of the enthalpy of the product steam:

$$H \text{ at } 100 \text{ psia, } 328^\circ\text{F} = 1187.2 \text{ Btu/lb}$$

$$\Delta H \text{ Heat input} = 196.3 \text{ Btu/lb}$$

$$H \text{ products} = 1383.5 \text{ Btu/lb}$$

From the steam tables, the final temperature = 710°F

$$\Delta T \text{ rise} = 382^\circ$$

The natural convection loop functions with the difference in vapor densities as the driving force. To optimize, the pressure drops in the convective circuit must be kept low.

In sizing the chimney, the major resistance in the convective loop was assumed to be through the catalyst.

The pressure drop through the wire mesh catalyst is

$$\Delta P = \frac{f L S \rho V_o^2}{2 g F^3} \quad (2)$$

where, f = Friction factor, function of Re

L = Bed depth, ft.

S = Area per unit volume of bed, ft^2/ft^3

ρ = Fluid density, lb/ft^3

V_o = Velocity based on empty tower cross section, ft/sec

g = Conversion factor, ft/sec^2

F = Void fraction of bed, dimensionless

Volumetric flow ratio:

Upflow volume at 100 psia, $710^\circ F$

$$V = 0.05476 \left(\frac{14.7}{100} \right) \left(\frac{1170}{492} \right) \times 359 = 6.87 \text{ ft}^3/\text{min}$$

Downflow volume at 100 psia, $328^\circ F$

$$V = 0.05476 \left(\frac{14.7}{100} \right) \left(\frac{788}{492} \right) \times 359 = 4.65 \text{ ft}^3/\text{min}$$

The densities of the hot and cold legs of the vapors are:

$$\rho_{\text{hot}} = \frac{0.05476}{6.86} \times 18 = 0.1436 \text{ lb/ft}^3$$

$$\rho_{\text{cold}} = \frac{0.05476}{4.65} \times 18 = 0.211 \text{ lb/ft}^3$$

$$\Delta \rho = 0.0674 \text{ lb/ft}^3$$

Pressure drops through various diameters of catalysts were calculated.

It was assumed the catalyst resistance was 80% of the total convective loop resistance.

$$\text{The chimney height} = \frac{\Delta P_{\text{Total}}}{\Delta \rho}$$

Table 2 gives the relationship of catalyst resistance, overall convective resistance and chimney height as a function of chimney diameter.

Heater Design Basis

1. Design flow rate = 1 scfm ($2 \text{ H}_2 + \text{O}_2$)
2. Explosive limit concentration = 15 mol per cent ($2 \text{ H}_2 + \text{O}_2$)
3. Diluent concentration = 85 mole per cent H_2O
4. Saturation condition at 100 psia; 328°F

Calculated minimum steam flow rate within explosion limit = 5.67 scfm

Calculated pressure drop of convective loop = 0.035 lb/ft^2

Enthalpy at saturated condition = 1187 Btu/lb

Density (ρ_A) of vapor at saturated condition = 0.2258 lb/ft^3

Calculated density (ρ_B) to provide the

$$\text{Driving force} = 0.2202 \text{ lb/ft}^3$$

$$\text{Calculated } \Delta H = 9 \text{ But/lb}$$

$$\text{Calculated heater requirement} = 0.05 \text{ kw}$$

For additional capacity to overcome heat losses to the annular space and convenience in fabrication in a 1 inch shaft, a 450 watt cartridge type heater was installed. During the initial test runs, difficulty was encountered in attaining convective circulation in a reasonable amount of time prior to gas mixture ($2 \text{ H}_2 + \text{O}_2$) introduction. Flashbacks occurred resulting in damage to the unit.

For the final heater design, three 450 watt cartridge type heaters were installed.

Calculation for Steam Diluent Flow (lb/min)

The heat of formation ($\text{H}_2 + 1/2 \text{ O}_2 \rightarrow \text{H}_2\text{O}$) is assumed to be $104,000 \text{ Btu/lb mol}$

The heat output due to recombination at a typical H_2 input is

$$q = \frac{0.265 \text{ ft}^3}{\text{min}} \times \frac{60 \text{ min}}{\text{hr}} \times \frac{1 \text{ lb mol}}{359 \text{ ft}^3} \times 104,000 \frac{\text{Btu}}{\text{lb mol}} = 4680 \text{ Btu/hr}$$

Assuming 100% recombination and no heat loss to annular space, the steam flow up the chimney is

$$q = (h_7 - h_5) W = 4680 \text{ Btu/hr.}$$

TABLE 2

<u>Chimney Dia. (Ins.)</u>	<u>ΔP Catalyst lb/ft²</u>	<u>ΔP Total lb/ft²</u>	<u>Chimney Height Ft.</u>
3	0.851	1.064	15.8
4	0.295	0.369	5.5
5	0.134	0.167	2.5
6	0.067	0.084	1.25

Chimney diameter was sized at 4 inches on the basis of reasonable height to space other internal components such as catalyst, heaters and coolant coils. The shell diameter was arbitrarily sized at 12 inches. This provided an annular volume sufficiently large to accommodate surges of hydrogen and oxygen and still be within the non-explosive range.

Condenser Design Basis

1. Heat to be removed = 17,065 Btu/hr
2. Air in, assumed = 70°F
3. Air out, assumed = 220°F
4. Air supply at 80 psig
5. Coolant tubing = 1/2" cu. water tubing, type K

Calculated mass flow of air = 474 lb/hr

Calculated air velocity = 102.5 ft/sec

For two concentric cooling coil design, the coolant air flow through each equals 237 lb/hr.

Calculated heat transfer coefficient, $h = 102 \frac{\text{Btu}}{\text{hr ft}^2 \text{ } ^\circ\text{F}}$

Calculated surface area required = 0.97 ft²

Total length of 1/2" copper water tubing = 7.03 ft

Calculated pressure drop through tubing = 0.906 psi/ft

Total overall length of 1/2" copper tubing (including additional capacity) = 28 ft.

Where h_7 = Enthalpy of fluid above the Catalyst bed = 1274 Btu/lb

h_5 = Enthalpy of fluid entering the chimney = 1184 Btu/lb

$$W = \frac{4680}{1274-1184} = 51.8 \text{ lb/hr} = 0.863 \text{ lb/min steam diluent flow.}$$

REFERENCES

- (1) Hougen, O. A. and Watson, K. M., "Chemical Process Principles," John Wiley and Sons, Inc.
- (2) Wilson, R. H., "Pressure Drop Across Wire Mesh Packing," CF-53-7-198.

I. K. Namba

IKN:job

Attachments

FIGURE 1

EUGENE DIETZGEN CO.
MADE IN U. S. A.NO. 340R-10 DIETZGEN GRAPH PAPER
10 X 10 PER INCHSTEAM DILUENT FLOW
#/MIN.

FIGURE 2

EUGENE DIETZGEN CO.
MADE IN U. S. A.

NO. 340R-10 DIETZGEN GRAPH PAPER
10 X 10 PER INCH

CONCENTRATION AS A FUNCTION OF H_2 FLOW
MOL PERCENT ($2H_2 + O_2$) VS SCFM HYDROGEN

FIGURE 3