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FEASIBILITY OF HIGH-PRESSURE NATURAL CIRCULATION
CATALYTIC RECOMBINERS FOR RADIOLYTIC GASES.

SUMMARY

A study has been made to determine if the driving force due to differences in gas densities at high pressure is sufficient to permit the design of a reasonable natural circulation catalytic recombiner for high power reactors. The only significant pressure drops in the design, considered here, lie in the pellets of the catalyst bed and the wire mesh of the entrainment separator. By designing the catalyst bed with a large flow area, it is possible to reduce the total pressure drop to reasonable values, particularly at the higher pressures (~ 2000 psia).

It is recommended that further development work be done on high pressure catalytic recombination to determine the following design criteria:

1. The explosive limits of $D_2(H_2)$ in the desired diluent at pressures up to 2000 psia and operating temperatures.
2. The operating characteristics of a catalytic type recombiner at pressures up to 2000 psia.
3. The best form of catalyst to use. (Pt coated wire mesh gives low pressure drops, but is not as durable as platinized alumina pellets.)
4. The amount of catalyst needed for quiet, efficient operation under pressure.
5. Safety considerations.

INTRODUCTION

Aqueous homogeneous reactors have a considerable gas disposal problem. External means of recombination must be provided at periods of low temperature operation, such as start-up time. These external recombiners should be designed to handle at least one-third of the maximum total radiolytic gas production expected. Two types of recombiners are:

1. Flame recombiners in which the entering gases are above the explosive limit and burning takes place.
2. Catalytic recombiners in which the gases are diluted below the explosive limit and are recombined in contact with a catalytic surface.

Recombination in the explosive range is hazardous and so the catalytic type recombiner is favored for the larger power reactors. The catalytic recombiners may be divided into the following types:

1. Low-pressure forced circulation recombiner
2. Low-pressure natural circulation recombiner
3. High-pressure forced circulation recombiner
4. High-pressure natural circulation recombiner

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The low-pressure type recombiners are handicapped by excessively large gas volumes. Other disadvantages include the problems involved in reducing the pressure on the gases from operating pressure to recombiner pressure (1 atmos.).

The simplest type of high-pressure recombiner is the natural circulation design. The purpose of this study is to see if such a design is feasible. A complete design is not given, as much development work is needed.

The only diluent considered in this study is D_2O . If helium or oxygen is used to dilute the radiolytic gases entering the catalyst bed, different physical properties, flow rates, etc., will be involved and consequently different chimney heights will be required. However, an approximation of the size of recombiner needed will be shown by this study.

The vessel housing the recombiner should probably serve also as a pressurizer for the reactor.

CALCULATIONS AND RESULTS

A simple schematic design of a high-pressure natural circulation recombiner is presented in Fig. 1. The radiolytic D_2 and O_2 gases enter the recombiner through a cyclone separator to remove some of the entrained fuel solution. The gases are then diluted with D_2O vapor and pass into the natural circulation chimney of the recombiner. Inside the chimney the gases first pass through an entrainment separator made of wire mesh. The gases are then heated to dry out any remaining moisture, and sent through the catalyst bed. Assuming 100% recombination, the gases leaving the bed are essentially pure D_2O . After leaving the chimney at the top the D_2O gas passes through a condenser which removes the heat of recombination and condenses an amount of D_2O equal to that which was formed in the catalyst bed. The remaining D_2O vapor flows down through the annular area outside the chimney and becomes the diluent for more radiolytic gas coming from the gas separator.

The amount of diluent circulated is assumed to be that quantity which will give the following gas composition entering the catalyst bed:

D_2		10 mol %
O_2		5 mol %
D_2O		<u>85</u> mol %
		100 mol %

The heat of formation ($D_2 + 1/2 O_2 \rightarrow D_2O$) is assumed to be [1]

$$\Delta H = 105,000 \text{ Btu/lb mol}$$

Thus, the heat input to the entering gas mixture is 550 Btu/lb. With the aid of the Keenan and Keyes steam tables [2], the temperature of the gas before and after the catalyst bed can be found for several operating pressures. See Table I.

TABLE I
GAS TEMPERATURES BEFORE AND AFTER THE CATALYST BED

p , psia	T_o	ρ	T_f	ΔT , °F	C_p , Btu/lb-°F
500	467	1.078	1434	967	0.571
1000	545	2.244	1424	879	0.629
1500	596	3.617	1397	801	0.690
2000	636	5.325	1356	720	0.768

p = pressure of system,

T_o = temperature of gas mixture entering catalyst bed. (Assumed sat. temp.) °F

ρ = density of D_2O vapor (gas in annulus) at saturation temperature, lb/ft³.

T_f = final temperature of gas leaving catalyst bed, °F.

C_p = mean heat capacity.

The density of the D_2O vapor in the downflow or annular section may be taken as the saturation density given in Table I. The density in the upflow or chimney section above the catalyst bed is estimated from

$$\rho_u = \frac{M}{V_m} = \frac{M}{\left(\frac{X RT}{p}\right)} \quad (1)$$

where ρ_u = gas density in upflow section, lb/ft³.

M = molecular weight of gas = 20 lb/lb-mol for D_2O .

V_m = volume of one molecular weight of gas, ft³/lb mol.

X = compressibility factor of gas (assumed same as H_2O vapor).

R = gas constant = 10.73 lb-ft³/in²-lb mol-°R.

T = temperature, °R.

p = pressure, psia

From equation (1) and the data of Table I, the densities in the chimney section may be determined. They are given in Table II.

TABLE II
GAS DENSITIES IN CHIMNEY OF RECOMBINER

Pressure of system p, psia	Compressibility $\times \sqrt{2}$	Density at final temperature, lb/ft ³
500	0.996	0.494
1000	.987	1.002
1500	.975	1.544
2000	.961	2.136

The driving force for the gas in the recombiner is assumed to be the product of the elevation between the catalyst bed and cooler-condenser and the difference between gas densities in the chimney and annular sections of the recombiner. This is equal to the total pressure drop in the system and is represented by

$$z(\rho_d - \rho_u) = (\Delta p)_t \quad (2)$$

where z = elevation between catalyst bed and condenser, ft.

ρ_d = density in downflow annulus, lb/ft³ (assumed = ρ_{sat}).

ρ_u = density in upflow section, lb/ft³.

$(\Delta p)_t$ = total pressure drop in natural circulation loop of recombiner, lb/ft².

The major pressure drops in the loop are in the entrainment separator and in the catalyst bed. The pressure drop across wire mesh such as is used in the entrainment separator is given by [3].

$$(\Delta p)_w = \frac{f L_w S \rho V_o^2}{2 g_c F^3} \quad (3)$$

$$f = 3.19 (\text{Re})^{-0.266} \quad (4)$$

where $(\Delta p)_w$ = pressure drop across entrainment separator, lb/ft².
 f = friction factor.
 $Re = \frac{G_o}{\mu S}$
 G_o = mass flow rate, lb/hr-ft².
 μ = viscosity, lb/hr-ft.
 S = surface area per unit volume, ft²/ft³.
 V_o = superficial velocity based on empty cross section, ft/sec.
 L_w = thickness of separator bed, ft.
 F = void fraction of wire mesh.
 ρ = density of gas, lb/ft³.

Pressure drop across alumina pellets such as are used in the catalyst bed is given by [4].

$$(\Delta p)_p = \frac{0.691 L_p \mu^{0.15} \rho^{0.85} V_o^{1.85} A_F}{D_p^{1.15}} \quad (5)$$

where $(\Delta p)_p$ = pressure drop across pellets of catalyst bed, lb/ft².
 L_p = thickness of pellet bed, ft.
 A_F = wall effect factor = 1.0 (assumed) for this case.
 D_p = diameter of pellet, ft.

Equation (3) and (5) assume turbulent flow in the recombiner loop. No design will be considered which will give laminar flow in the loop.

Two types of wire mesh will be considered--coarse and fine. They have the following characteristics:

	Coarse	Fine
$S, \text{ft}^2/\text{ft}^3$	110	355
F	0.977	0.968

From these values and equation (4) the ratio of pressure drops for the coarse and fine wire mesh may be found.

$$\frac{(\Delta p)_w \text{ (coarse)}}{(\Delta p)_w \text{ (fine)}} = 0.221 \quad (6)$$

The 1/8" x 1/8" cylindrical pellet will be the only catalyst pellet considered in this study. For turbulent flow through the catalyst bed, the increased volume of the larger sized pellets needed offsets the lower pressure drop and little advantage is realized. Disadvantages, such as the possibility of flash-backs crushing the larger pellets and thereby increasing the pressure drop, also rule out the use of large pellets.

Assuming a G-value of 1.67 molecules D_2 /100 ev, the gas production rate is

$$\omega = 0.0208 P_o \quad (7)$$

where ω = gas production rate in core, lb mols D_2 /min.

P_o = reactor power, Mw.

Using the concentration of D_2 in the gas mixture entering the bed as 10 mol %, the value for the mass flow rate and velocity can be found in terms of power level, density of gas and cross sectional area of bed.

$$G_o = \frac{237 P_o}{A}, \text{ lb/hr-ft}^2 \quad (8)$$

$$V_o = \frac{0.0659 P_o}{\rho A}, \text{ ft/sec} \quad (9)$$

The pressure drop equations may be reduced to expressions containing reactor power level, cross sectional area, and thickness of bed, terms only. For the fine wire mesh

$$(\Delta p)_w \text{ (fine)} = \alpha(p) L_w \left(\frac{P_o}{A}\right)^{1.734} \quad (10)$$

for the 1/8" x 1/8" cylindrical pellets

$$(\Delta p)_p = \alpha(p) L_p \left(\frac{P_o}{A}\right)^{1.85} \quad (11)$$

where $\alpha(p)$ = constant depending on pressure.

In Table III are listed some values of the constant $\alpha(p)$ for the present case.

TABLE III
VALUES OF $\alpha(p)$

Pressure psia	$\alpha(p)$ for fine wire mesh of entrain- ment separator	$\alpha(p)$ for fine wire mesh used as catalyst	$\alpha(p)$ for 1/8"xl/8" cylindrical pellets
500	0.0436	0.0991	1.204
1000	.0224	.0505	.605
1500	.0146	.0330	.394
2000	.0103	.0241	.287

The elevation required to overcome the pressure drops in the entrainment separator and catalyst bed may be found by substituting equations (10) and (11) into equation (2).

Let Z_w = elevation needed to overcome $(\Delta p)_w$, ft.

Z_p = elevation needed to overcome $(\Delta p)_p$, ft.

Then

$$Z_w = \frac{\alpha_w(p) L_w \left(\frac{P_o}{A_w}\right)^{1.734}}{\rho_d - \rho_u} \quad (12)$$

$$Z_p = \frac{\alpha_p(p) L_p \left(\frac{P_o}{A_p}\right)^{1.85}}{\rho_d - \rho_u} \quad (13)$$

Equations (12) and (13) may be plotted with Z vs. P_o for the parameters area, (A) , and pressure, (p) . This has been done for the case under consideration here. Figures (2) through (9) show the results.

The total elevation needed between the catalyst bed and the condenser-cooler is then given by

$$Z = Z_w + Z_p \quad (14)$$

If the catalyst bed is made in the form of a cylindrical annulus as shown in Figure (10), a large increase in flow area can be accomplished without increasing the diameter of the chimney. This type bed may be necessary as the pressure drop across the pellets is much higher than the corresponding drop across wire mesh. Should platinum coated wire mesh prove practicable, this type catalyst bed design will be unnecessary.

As an example, consider a reactor power plant producing power at a level such that radiolytic gas from 150 Mw of reactor heat is being released from solution. At this power level the amount of gas mixture flowing in the chimney is 31.2 lb-mols/min, which is about 600 lbs/min or 280 cu ft/min (at 2000 psia and operating temperatures). To reduce the pressure drop across the catalyst pellets to a reasonable value, a large cross sectional area is needed and so a bed design similar to that shown in Figure (10) is assumed. The annular bed is designed to be two inches thick which should be sufficient to prevent channeling of the gas. The average cross sectional area available to flow of gases through the bed is now a function of the bed length and the average diameter of the bed annulus. For the designs listed in Table IV a catalyst bed area of 10 ft² is used. Since the gases are heated in the bed, the bed diameters (inside and outside) are chosen so that the flow area available to the gases leaving is about 60% greater than the flow area of gases entering the bed.

The height of the chimney should be great enough to house the entrainment separator, the catalyst bed, and to provide sufficient driving force to overcome pressure losses in the separator and bed. The heights sufficient to overcome the pressure drops are read from Figures (8) and (9). Three designs are listed in Table IV.

At lower pressures the specific volume of the gas mixture will be greater and the mass circulation rate will be smaller. Using Figures (2) through (7), the power levels that the chimney designs of Table IV will handle can be determined for pressures of 1500 psia, 1000 psia and 500 psia. Figure (11) shows the power level, P_0 , that the recombiner designs can accommodate versus pressure, holding the inlet mixture at 85 mol % D₂O and 15 mol % combustibles.

It may prove desirable to place the condenser inside the chimney section rather than in the annular space outside the chimney. In this case the entrainment separator and catalyst bed should be placed in the annular section causing the gases to rise in the annulus and fall inside the chimney. Figures (2) through (9) will still represent this case for corresponding cross sectional areas of entrainment separators and catalyst beds.

TABLE IV
NATURAL CIRCULATION CHIMNEY DESIGNS FOR RADIOLYTIC GAS FROM
150 MW POWER AND AT 2000 PSIA

	Design 1	Design 2	Design 3
Chimney size	18" Sch 10 pipe	20" Sch 10 pipe	24" Sch 10 pipe
Height of chimney required to overcome pressure drop in entrainment separator	10 ft	7 ft	3.6 ft
Thickness of entrainment separator	.2 ft	2 ft	2 ft
Area of catalyst bed (cross section)	10 ft ²	10 ft ²	10 ft ²
Thickness of catalyst bed	2 in	2 in	2 in
Length of catalyst bed	4 ft	3.8 ft	3 ft
Height of chimney required to overcome pressure drop in catalyst bed	2.5 ft	2.5 ft	2.5 ft
Total Height of Chimney	18.7 ft	15.3 ft	11.1 ft

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- (2) Keenan, J. H., and Keyes, F. G., "Thermodynamic Properties of Steam," John Wiley Book Co., (1936).
- (3) Wilson, R. H., "Pressure Drop Across Wire Mesh Packing," CF-53-7-198.
- (4) Perry, J. H., Chemical Engineers' Handbook, McGraw-Hill (1950).

Fig. 1. Schematic Design of High Pressure Natural Circulation Recombiner

Fig. 2. Required Height of Chimney for Δp in Entrainment Separator at 500psia

Fig. 3. Required Height of Chimney for ΔP in Catalyst Bed at 500 psia

Fig. 4. Required Height of Chimney for Δp in Entrainment Separator at 1000 psia

Fig. 5. Required Height of Chimney for Δp in Catalyst Recombiner at 1000 psia

Fig. 6. Required Height of Chimney for Δp in Entrainment Separator at 1500 psia

Fig. 7. Required Height of Chimney for Δp in Catalyst Bed at 1500 psia

Fig. 8. Required Height of Chimney for Δp in Entrainment Separator at 2000 psia.

Fig. 9. Required Height of Chimney for Δp in Catalytic Bed at 2000 psia

Fig. 10. Schematic Diagram of Annular Type Catalyst Bed

Fig. 11. Reactor Power Level For Which Recombiner is Designed vs Operating Pressure

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