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FLOW ANALYSIS OF DIFFUSER-GETTER-DIFFUSER SYSTEMS

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Tritium clean-up systems typically deploy gas processing technologies between stages of palladium-silver (Pd/Ag) diffusers/permeators. The number of diffusers positioned before and after a gas clean-up process to obtain optimal system performance will vary with feed gas inert composition. A simple method to analyze optimal diffuser configuration is presented. The method assumes equilibrium across the Pd/Ag tubes and system flows are limited by diffuser vacuum pump speeds preceding or following the clean-up process. A plot of system feed as a function of inert feed gas composition for various diffuser configuration allows selection of a diffuser configuration for maximum throughput based on feed gas composition.

I. INTRODUCTION

Like the design of many tritium clean-up systems¹, the Savannah River Site (SRS) Tritium Facilities uses an impurity removal process between stages of palladium-silver (Pd/Ag) alloy diffusers/permeators. The diffuser performs initial tritium removal before impurity processing (using a SAES[®] St 909 at SRS), followed by further diffuser tritium removal. The design of an SRS diffuser-getter-diffuser system was to include a third diffuser stage. The location of this third diffuser to maximize system performance was to be determined.

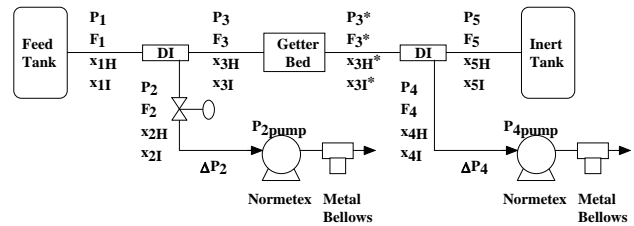
II. BACKGROUND

Hydrogen isotopes (Q₂) can be stored as metal hydrides, but other process gases are stored in tanks before tritium recovery and release of waste gases from the facility. These gases are typically fed to a diffuser to recover Q₂ and concentrate impurities. The diffuser effluent then enters an impurity processor to remove impurities and “crack” tritiated species followed by another diffuser for further Q₂ recovery.

The number of diffusers needed preceding and following a getter bed and the choice of series or parallel configuration needs to be determined. The purpose of this paper is to outline a simple methodology for analyzing diffuser-getter-diffuser configurations and present the results obtained from spreadsheet calculations

III. NOTATION AND METHODOLOGY

The Figure 1 schematic shows the gas flows and notation used: F is flow in sccm, P is pressure in kPa, and x is mole fraction. Subscript numbers identify the stream: odd-numbers for flows in or out of diffuser tubes, even numbers for permeate flows. The subscript H refers to Q₂ properties and I to inerts (i.e. non-hydrogen isotopes) properties. The “*” symbol denotes changed stream properties by a getter bed, but these changes will be neglected for this analysis.



x_{3H} controlled to fixed value to prevent methane decomposition in a diffuser (DI)

Fig. 1. Diffuser-Getter-Diffuser Notation

As previously described², it is desired to maintain the 1st stage diffuser effluent Q₂ partial pressure at an elevated value to reduce diffuser coking from methane. Thus a value for x_{3H} will be specified for these analyses. The tube-side Q₂ partial pressures at the exit of the diffusers are assumed to be in equilibrium with the shell pressures: P₂ = x_{3H}·P₃ and P₄ = x_{5H}·P₅ with x_{2H} = x_{4H} = 1.

A protium pump curve was measured for a 15 m³/hr Normetex pump backed by MB-601 Metal Bellows pump with the two heads connected in series. The pump curve correlates pump inlet pressure with gas flow and is

$$\log_{10}(P_{\text{pump}}, \text{kPa} \cdot 7.5) = [0.7826 + 2.129 \cdot (G - 0.5) - 0.2916 \cdot (6 \cdot G^2 - 6 \cdot G + 1) + 0.03103 \cdot (20 \cdot G^3 - 30 \cdot G^2 + 12 \cdot G - 1)], \quad (1)$$

where G = pump inlet flow (sccm) / 755

Mass balances correlate flows, F₁ = F₂ + F₃ and F₃ = F₄ + F₅. A Q₂ mass balance around each diffuser adds

additional equations relating flows and compositions: $F_3 = F_2(x_{2H}-x_{1H})/(x_{1H}-x_{3H})$ and $x_{5H} = (F_3x_{3H}-F_4x_{4H})/F_5$.

Pressure drops through the diffusers and getter bed will be neglected for these analyses ($P_1 = P_3 = P_5$) and a system (tube-side) operating pressure selected. The pressure drop between the diffuser shell and a vacuum pump (a function of flow and pipe size) will also be neglected for simplicity of presentation ($\Delta P_2 = \Delta P_4 = 0$).

With these stated assumptions, the analysis was reduced to the specification of one more variable to completely define the system. The system feed gas inert gas composition, x_{1H} , was chosen to start the calculations and the results are presented as a function of x_{1H} .

The calculation sequence for fixed values of x_{1H} , x_{3H} , and system pressure for the 1st diffuser was: 1) Select an initial estimate of F_2 , 2) Calculate F_3 using Q_2 mass balance equation $F_3 = F_2(x_{2H}-x_{1H})/(x_{1H}-x_{3H})$, 3) Calculate F_1 using mass balance equation $F_1 = F_2 + F_3$, 4) Calculate ΔP_2 (taken as zero for this analysis) using F_2 , 5) Calculate pump inlet pressure (P_{2pump}) using $P_2(= x_{3H} \cdot P_3) - \Delta P_2$, 6) Calculate P_{2pump} using F_2 in Equation 1, 7) Repeat calculation Steps 1 through 6 until P_{2pump} from Step 5 and Step 6 were equal. This process was repeated for a range of feed compositions.

The calculation sequence for the 2nd diffuser was similar to that used for the 1st diffuser, but differed in that its inlet flow (F_3) and composition (x_{3H}) were known, but its outlet conditions (F_5 and x_{5H}) were unknown and were to be calculated.

The calculation methods described above were forward-calculation sequences: x_{3H} is specified and x_{5H} is calculated. A backward-calculating sequence used selected x_{5H} to then calculated upstream parameters. One artifact of the backward-calculation method is that F_2 can be calculated using the Q_2 mass balance equation $F_2 = F_3(x_{1H}-x_{3H})/(x_{2H}-x_{1H})$ which exceeds the pump speed calculated using Equation 1. The implication of this inconsistency will be discussed later.

Diffuser configurations analyzed will be denoted as XY. X and Y indicate the number of diffuser in a particular position with X plus Y totaling up to number of diffusers available –three for this paper. For example, a 21 configuration has two diffusers operating in parallel before a St 909 bed followed by a single diffuser.

IV. RESULTS

Figure 2 shows the results for a system tube-side pressure of 101 kPa and x_{3H} equals 20%. Fixing the

system pressure and x_{3H} , specified P_2 and implicitly determined F_2 from the pump curve equation - independent of system feed composition. Figure 2 shows that as x_{1H} increased, the system throughput increased, but the system Q_2 outlet concentration also increased. When a system maximum Q_2 outlet concentration is desired, backward calculations can be performed. The results for x_{5H} equal to 0.001 (0.10%) are shown in Figure 3.

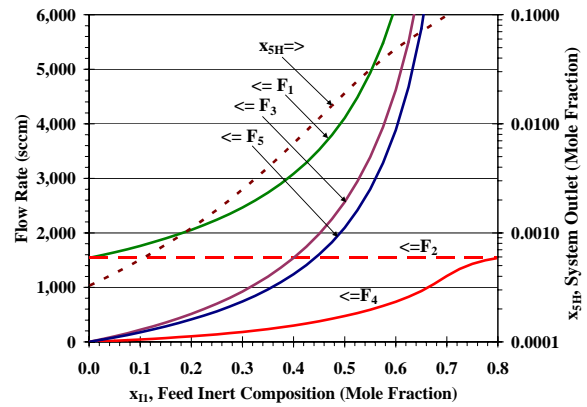


Fig. 2. 11 Configuration for 101 kPa and $x_{3H} = 20\%$: Forward Calculation

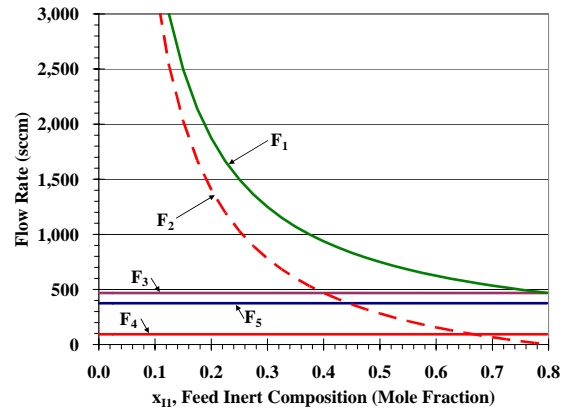


Fig. 3. 11 Configuration for 101 kPa, $x_{3H} = 20\%$, with x_{5H} of 0.10%: Backward Calculation

Figure 3 shows as the feed became leaner in inerts (richer in Q_2), F_2 calculated from the Q_2 mass balance equation increased. Figure 3 does not show is that at some feed compositions, F_2 calculated from the mass balance exceeds F_2 calculated using Equation 1. To better understand this apparent conflict, the results for Figure 2 and Figure 3 are combined and presented in Figure 4.

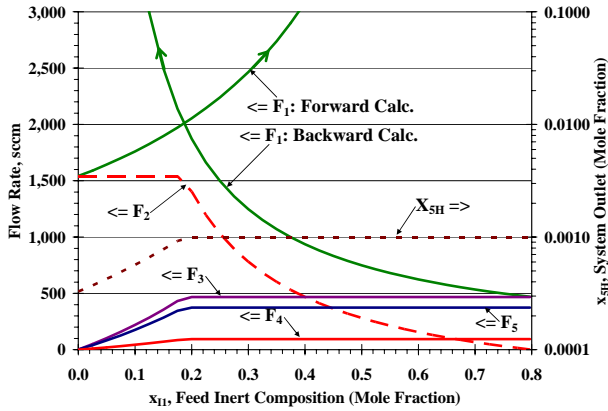


Fig. 4. 11 Configuration Results at 101 kPa, $x_{3H} = 20\%$ and $x_{5H} = 0.10\%$ Using Forward and Backward Calculations

Figure 4 shows the 1st stage diffuser pump is running at a fixed rate for low values of x_{11} , and as x_{11} increases, F_1 increases, using the forward calculations, with a resulting increase in outlet Q_2 concentration. To maintain x_{5H} below some desired value as x_{11} increases, the system feed rate must be decreased per the backward calculations. The point where the feed flow (F_1) plots from the forward and backward calculations intersect will be referred to as the feed transition point (FTP). Before the FTP, the 1st diffuser vacuum pump limits the system feed rate while after the FTP, the 2nd diffuser vacuum pump limits the system flow rate.

The FTP at x_{3H} equals 20% and x_{5H} equals 0.10% is shown for pressures of 51 kPa (380 torr), 101 kPa (760 torr), and 203 kPa (1520 torr) are shown in Figure 5. Figure 6 shows the FTP for 101 kPa, x_{5H} equals 0.10% for values of x_{3H} of 15%, 20%, and 25%.

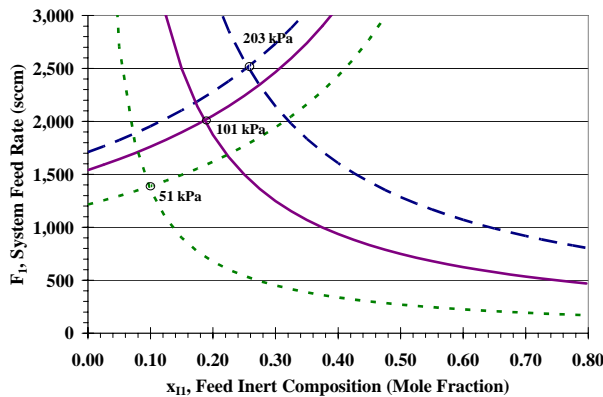


Fig. 5. 11 Configuration Results for $x_{3H} = 20\%$ and $x_{5H} = 0.10\%$ for Various Pressures

The Figure 4 results have been extended for different diffuser configurations. Figure 7 shows the F_1 forward calculation results for one and two diffusers in front of the getter bed (1Y and 2Y configurations) along with the F_1 backward calculation results for one and two diffusers following the getter bed (X1 and X2 configurations) for a pressure of 101 kPa, x_{3H} of 20%, and x_{5H} of 0.10%. Also shown are the results for a 03 configuration.

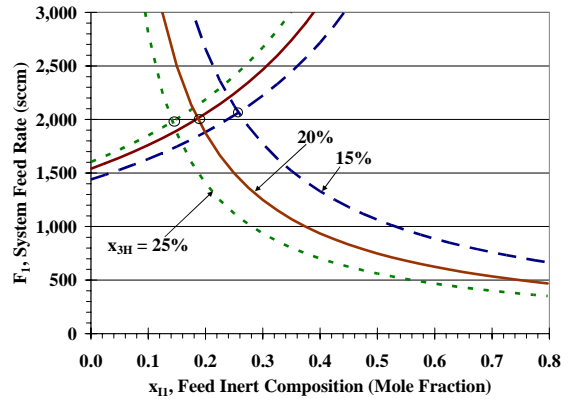


Fig. 6. 11 Configuration Results at 101 kPa, $x_{5H} = 0.10\%$ and $x_{3H} = 15\%, 20\%, \text{ and } 25\%$

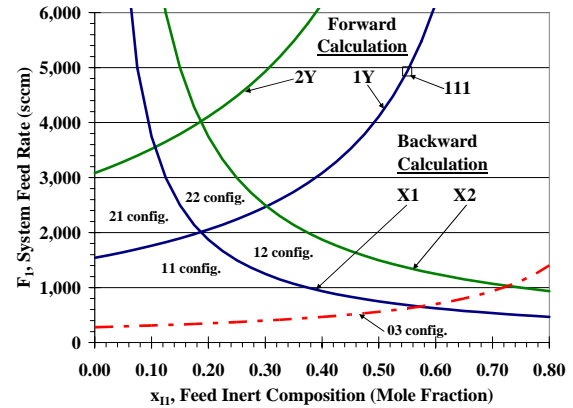


Fig. 7. Various Diffuser Configuration for 101 kPa, $x_{3H} = 20\%$, and $x_{5H} = 0.10\%$

V. DISCUSSIONS

Figure 2 shows the performance of a diffuser-getter-diffuser can be estimated as a function of system feed tank composition. The large, asymptotic nature of the system flows as x_{11} increases would eventually negate the equilibrium assumption of Q_2 partial pressures across the diffuser membrane. The addition of another diffuser

stage to a system location is equivalent to adding an additional vacuum pump when using the equilibrium assumption.

Figure 4 shows that when adding the criteria of a maximum x_{5H} value, the system equations are over-defined. The reduction in F_1 and F_2 flow rates after the FTP indicates the 1st diffuser vacuum pump is operating at less than its capacity.

Figure 5 shows doubling the system pressure does not double the system feed flow for all cases, but the impact of increased pressure on system feed flow rate is greater after the FTP. For example, at an inert feed composition of 30%, doubling the pressure from 51 kPa to 101 kPa increased system feed rate from 451 sccm to 1248 sccm: doubling the pressure again to 203 kPa only increased the flow to 2142 sccm. The changes in system flows with changes in system pressure are due to the shape of the vacuum pump flow-pressure relationship.

The impact of running the diffuser system at different intermediate Q_2 partial pressures (x_{3H}) can be seen in Figure 6. Before the FTP, there is a less pronounced change in maximum system flow. After the FTP, the maximum system flow can be reduced for increased x_{3H} values. For example, when x_{1I} equals 30%, the maximum system feed is 1772 sccm when x_{3H} equals 15%, but is reduced to 935 sccm when x_{3H} equals 25%. The lower flow after the FTP at higher x_{3H} values is attributed to the larger volume of Q_2 that are to be removed by the 2nd diffuser pump.

Figure 7 shows that for a known feed tank inert composition, the optimum diffuser configuration can be selected. For example, for 10% inert feed, the 21 configuration will give a larger system throughput than a 12 configuration. Conversely, for 40% inert feed, the 12 configuration allows larger system throughputs than the 21 configuration. Figure 7 also shows for inert feeds greater than 72.5%, the 03 configuration allows the fastest system throughput.

By extension of the calculation methods presented, analysis of a 111 diffuser configuration could be done: one diffuser before the St 909 bed followed by two diffusers in series. These results were not presented due to the uncertainty of the pump curve correlation at low pressures. Figure 2 would show the outlet Q_2 concentration from the 3rd diffuser stage roughly two orders of magnitude lower than that from 2nd diffuser stage for the same feed rate.

When the outlet from the 3rd diffuser stage was limited to 0.10% Q_2 as used in the previous examples, the forward calculation method gave a maximum feed rate of

4931 sccm for 55% inerts in the feed – this datum is indicated in Figure 7. Further study is needed to fully verify and compare the 111 configuration to other diffuser-getter-diffuser configurations.

VI. CONCLUSIONS

The simple analysis method presented gave great insight into optimal diffuser configuration when additional diffusers are added to the design and operation of a system with various composition feed gas. True system performance requires estimating diffuser-to-pump pressure drops and validation of the equilibrium assumption across the diffuser tube. A wider range of vacuum pump curves would allow a more in-depth analysis of different system configurations.

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