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THE SEPARATION OF HYDROGEN ISOTOPES
BY CRYOGENIC DISTILLATION

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Application Of Sidestream Recycle To The Separation Of Hydrogen Isotopes By Cryogenic Distillation

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ABSTRACT

Withdrawal of a sidestream from an isotopic-separation distillation column followed by equilibration of species and reinjection is a technique for reducing the required number of columns and hence reducing tritium inventories in fusion-energy fuel processing systems. Measurements have been made with a single column and the three-component H_2 -HD- D_2 system withdrawing streams of essentially pure H_2 and D_2 . This was followed by single column measurements with the full HDT system and a column inventory of ca. 16.8 moles ($H/D/T = 0.043/0.528/0.429$). Rather pure T_2 (ca. 98.7 mol%) was produced as one of the products. This would previously have required two separate distillation columns with an inventory of ca. 16 moles of T_2 as compared with 7.2 moles of T_2 in these experiments. The dynamic response of the H-D system was followed using *in-situ* laser Raman spectroscopy to obtain product compositions and column composition profiles.

INTRODUCTION

Normally one cryogenic distillation column is required for each specified component to be extracted as a product from a distillation column system. Withdrawal of a sidestream, followed by isotopic equilibration at room temperature, and reinjection into the distillation column is a powerful technique for reducing the required number of columns and hence reducing tritium inventories with significant application to the processing of fusion fuel streams.[1]

Thus in a H-T system, the species equilibrate to H_2 -HT- T_2 . In a normal column the distillate stream could be chosen to be either H_2 or H_2 +HT. Correspondingly the reboiler product would be constrained to be HT+ T_2 or T_2 . In either case a significant quantity of tritium in the form of HT must appear as a product. HT is, in general, a rather undesirable molecule as fusion fuel process systems try to eliminate H-bearing species and drastically reduce the amount of T-bearing species which must be treated as waste.

In normal operation the three species vary in concentration from the bottom to the top of the column, with the T_2 concentrating at the bottom, the H_2 concentrating at the top, and the HT peaking somewhere in the center of the column. If a sidestream is withdrawn from the distillation column at a point where the HT species is near a maximum and passed over a catalyst bed, the isotopic equilibrium,

is effected and the HT molecule is reduced, increasing the H_2 and T_2 species which, after reintroduction into the column, will appear at the top and bottom, respectively. Composition profiles are shown in Figs. 1 and 2 for the H_2 -HD- D_2 system, without and with sidestream recycle, as predicted by dynamic modeling.

Fig. 1 Equilibrium Composition Profile BEFORE sidestream recycle. Feed is at stage 29. Dashed line - H_2 , dotted line - HD, Solid line - D_2 (lower-left corner of figure).

Utilizing this approach it is possible to reduce the number of columns required by a factor of as much as two. This will permit a significant reduction in the inventory of tritium held up in the distillation system as well as the complexity and controllability of the overall system, especially in a full HDT isotope separation system.

Fig. 2 Equilibrium Composition Profile WITH sidestream recycle. Feed is at stage 29, sidestream recycle is from stage 9 to stage 29. Dashed line - H_2 , dotted line - HD, Solid line - D_2 (lower-left corner of figure).

EXPERIMENTAL

A. Experiments with H_2 and D_2

For this research, the H_2 -HD- D_2 system was chosen for initial study. The first column of the TSTA (Tritium Systems Test Assembly) 4-column cascade was chosen for the tests because it has 84 theoretical stages, 3 alternate feed-injection points, and seven sample withdrawal locations.[2] The column process piping was modified to permit a stream to be withdrawn from any of the sample taps through a flow control valve, equilibrated over a precious metal catalyst bed at ambient temperature, and pumped back into any other sample tap. Fig. 3 shows the column layout schematically.

The column was cooled to *ca.* 20K and filled with 14.2 moles of a hydrogen isotopic mixture with the composition H:D=2:1. Approximately 0.002% of T_2 was eluted from the walls of the system during the course of the measurements, but this had no significant effect on the performance of the system. Tritium appeared as HT or DT in the top and bottom products.

After a period at total reflux, the top and bottom streams were withdrawn at rates of *ca.* 2971 and 66 sccm ($\sim 2\%$) respectively, recombined and re-injected into the main feed at a rate of *ca.* 3037 sccm. The reflux ratio was 8. During this time the composition of the various streams were determined by *in-situ* Raman spectrometry. Raman analyses were taken every two minutes, each observation being the average of 100 1-second scans. After a period of several hours, when the composition had reached a steady state, sidestream recycle was initiated at the rate of 1008 sccm using the withdrawal and reinjection stages illustrated in Fig. 2.. Fig. 4 shows the dynamics observed during a several hour period of Raman measurements of the reboiler (bottom) stream composition with data points taken every minute. The flows chosen are similar to those which might be expected in a system designed to recover T_2 from an H-T breeding-blanket system.

Fig. 3 Schematic diagram of TSTA Column I as modified for sidestream recycle.

After the data in Fig. 4 was obtained, the sidestream recycle flow was stopped and the system allowed to return to normal 1-column operation. A portion of the data taken every 2 minutes over an 8-hour period are shown in Fig. 5.

Following the return to steady state, sidestream recycle was again started. This time the reboiler flow was 184 sccm ($\sim 6\%$) and the sidestream recycle flow was 1999 sccm. The dynamics were followed for a period of several hours taking Raman spectra of the reboiler stream every 2

Fig. 4 Reboiler composition following initiation of sidestream recycle (1008 sccm). Solid line - D_2 , dotted line HD, dashed line - H_2

minutes. The results are shown in Fig. 6. It was unfortunately necessary to terminate the experiment after only about 5 hours. If this experiment had been allowed to proceed for about 20 hours the reboiler composition must have returned to containing some HD as dictated by mass balance considerations and predicted by static and dynamic modeling.

Fig. 5 Reboiler composition without sidestream recycle - return to normal single-column operation. Solid line - D_2 , dotted line HD, dashed line - H_2

B. Experiments with H, D, and T.

A second set of experiments was performed having charged the columns with a 16.8 mole mixture having the composition $H:D:T=0.043:0.528:0.429$; a reflux ratio of 8 was used. Because of the requirements to perform other experiments with the TSTA process loop it was not possible to obtain significant dynamic data for the distillation column

Fig. 6 Reboiler composition following initiation of sidestream recycle (1999 sccm). Solid line - D_2 , dotted line HD, dashed line - H_2

response. However operating the system in a similar manner to the H-D experiments above it was possible to extract a reboiler stream with a composition of up to 2.6% DT, 97.4% T_2 , and no significant quantity of D_2 . At the same time the top stream was mostly HD and D_2 with a little DT, having an HDT ratio of $H:D:T=0.21:0.79:0.10$. Without sidestream recycle the reboiler composition was $D_2:DT:T_2=0.078:0.516:0.406$.

DISCUSSION

Without the use of sidestream recycle, it has traditionally been necessary to make use of a 2-column cascade to produce streams of pure H_2 and T_2 from an H_2 -HT- T_2 mixture. Each of the two columns would have a significant inventory of tritium. The use of the sidestream recycle configuration permits the elimination of one distillation column. The column flow rates must be adjusted and the column profile is modified. The resultant tritium inventory is reduced by a factor of almost two.

ACKNOWLEDGMENT

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