LEGIBILITY NOTICE

A major purpose of the Technical Information Center is to provide the broadest dissemination possible of information contained in DOE's Research and Development Reports to business, industry, the academic community, and federal, state and local governments.

Although a small portion of this report is not reproducible, it is being made available to expedite the availability of information on the research discussed herein.

LA-UR -88-1230

[illegible]

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

LA-UR--88-1230

DE88 009134

TITLE: OPERATION OF THE TSTA ISOTOPE SEPARATION SYSTEM WITH 100 GRAM TRITIUM

AUTHOR(S): Robert H. Sherman and John R. Bartlit
Materials Science and Technology Division
Tritium Science and Technology Group
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

Hiroshi Yoshida, Toshiniko Yamanishi, Taisei Naito, Shingo Hirata and
Yuji Naruse
Department of Thermonuclear Fusion Research
Japan Atomic Energy Research Institute
Tokai Research Establishment, 319-22
Tokai-mura, Naka-gus, Ibaraki-ken, JAPAN

SUBMITTED TO: Third Topical Meeting on Tritium Technology in Fussion, Fusion, and Isotopic Applications - Toronto, CANADA - May 1-6, 1988

By acceptance of this article, the publisher recognizes that the U.S. Government retains a nonexclusive, royalty-free license to publish or reproduce the published form of this contribution, or to allow others to do so, for U.S. Government purposes.

The Los Alamos National Laboratory requests that the publisher identify this article as work performed under the auspices of the U.S. Department of Energy.

MASTER

Los Alamos National Laboratory
Los Alamos, New Mexico 87545

FORM NO 836 R4

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

*OPERATION OF THE TSTA ISOTOPE SEPARATION SYSTEM WITH 100 GRAM TRITIUM

Robert H. Sherman and John R. Bartlit
Materials Science and Technology Division
Tritium Science and Technology Group
Los Alamos National Laboratory, MS C348
Los Alamos, New Mexico, 87545 USA
505-667-1410

Hiroshi Yoshida, Toshihiko Yamanishi, Taisei Naito, Shingo Hirata, and Yuji Naruse
Department of Thermonuclear Fusion Research
Japan Atomic Energy Research Institute
Tokai Research Establishment, 319-22
Tokai-mura, Naka-gun, Ibaraki-ken, JAPAN
0292-82-5577

ABSTRACT

In March of 1988 full operation of the 4-column isotope separation system (ISS) was realized in runs that approximated the design load of tritium. Previous operations had been fraught with operating difficulties principally due to external systems. This report will examine the recent highly successful 6-day period of operation. During this time the system was cooled from room temperature, loaded with hydrogen isotopes including 109 grams of tritium, integrated with the transfer pumping, impurity injection, and impurity removal systems, as well as the remote computer control system. At the end of the operation 12 grams of tritium having a measured purity of 99.987 % (remainder deuterium) were offloaded from the system. Observed profiles in the columns in general agree with computer models. A Height Equivalent to a Theoretical Plate (HETP) of 5.0 cm is confirmed.

INTRODUCTION

The Isotope Separation System (ISS) at the Tritium Systems Test Assembly (TSTA) was designed in 1977, constructed by Arthur D. Little, Inc. (Cambridge, MA), and placed initially in service during 1981[1]. A schematic diagram is shown in Figure 1. In the intervening years many experiments have been performed on individual columns, pairs of columns, and even the entire 4-column system. The tritium inventory has been gradually raised until in June-July of 1987 a 100-gram level was reached. Previous 4-column experiments have suffered from a series of difficulties, not entirely due to the ISS.

---

*This work is supported by the US Department of Energy, Office of Fusion Energy, and the Japan Atomic Energy Research Institute.

Recent measurements on one and two column cascades of the ISS are reported elsewhere at this conference.[2]

START UP SCENARIO

The column system was cooled from room temperature by simultaneously starting to fill liquid N2 into the radiation shield and the helium refrigerator. In about 4 hours columns I and T were cooled to the point that hydrogen isotopes, loaded into the condensers from uranium storage beds, would condense. These two columns cool more rapidly because of the extra coolant lines along the column length. Within 8 1/2 hours liquid was obtained in the reboilers of these two columns. After an additional 3 1/2 hours, the column D was cold and liquid filled into the reboiler. Column H required a total of 17 1/2 hours to fill liquid into the reboiler because it was not fed from a uranium bed through the condenser, but received its charge by flow through feed and product lines from the other columns. For reasons of safety, the uranium bed supplying the charge for column D was not heated until the uranium beds supplying columns I and T were substantially empty. The schedule for cooldown could be reduced by a factor of about 2 if all of the beds could be heated simultaneously.

OPERATING SCENARIO

The overall composition of the isotopic mixture introduced into the system was: hydrogen 11.1 grams, deuterium 218.0 grams, and tritium 109.2 grams or mole fractions of 0.071, 0.696, and 0.233 respectively. This is not the nominal mixture for which the columns were designed. Through the transfer pumping system full flows were established which were close to the design values. After initial cooldown, the coolant flows available to remove tritium decay heat from columns I and T were shut off completely. Therefore the packed sections of the columns were operated adiabatically during the run. Over a period of 4 days, the columns were operated in a mostly stable mode with little operator intervention required. During this period, the conditions listed in Table I were set, initially manually at the local control panel, and later through instructions to the TSTA computer system (MDAC) after transferring to the remote control mode.

The system was designed to vent ^3He, H_2 and HD from the top of column I and this task was performed periodically at flow rates varying between 50 and 200 cm^3/min. During the course of the run 0.16 moles of 3He were removed together with 1.78 moles of protium and 0.51 moles of deuterium. Thus by the end of the run the net composition of isotopes in the overall TSTA system were H:D:T = 0.050 : 0.710 : 0.240. Nominally 1% H_2 added to the feed stream continuously would have maintained constant composition, but this was not done during this run.

During the course of the run, gas samples were periodically withdrawn for analysis by gas chromatography. The chromatographs employ 5A molecular sieve columns coated with Fe_2O_3

and cooled with liquid N_2 and using neon as a carrier gas; both thermal conductivity and ion chamber detectors were used. A typical analysis required 37 minutes. The relative locations of the sample taps and feed points are shown in Figure 1. At the same time, all column variables were archived by the data acquisition system (MDAC) every minute. From these archives it is possible to reconstruct the operational history of the system and calculate many valuable parameters relative to the system performance.

Near the end of the run, 12 grams of tritium were withdrawn from the liquid pool in the reboiler of column T both to prove that capability, and to have a source of pure T_2 available for experiments. At the time of withdrawal, gas chromatographic analysis indicated a purity of at least 99.995%. By the time the material had been pumped into a 50-liter tank, there was some degradation, and the final analysis was 99.987% T_2 with the balance being D_2 (as DT).

Table I
Nominal Conditions for Column Operation

	Column I	Column H	Column T	Column D
Feed Flows, cm^3/min:				
From T(top)	4300			
From T(Bot)	200			
From D(Top)	1100	3900		
From D(Bot)	400			
From I(Top)		1500		
From I(Bot)			4500	
From H(Bot)				5400
Totals:	6000	5400	4500	5400
Top Fraction	0.25	0.0	0.956	0.926
Product Flows, cm^3/min:				
Top	1500	0*	4300	5000
Bottom	4500	5400	200	400
Totals:	6000	5400	4500	5400
Pressure, torr	860	800	700	700
Reboiler Power, W	37	17	25	20

---

* 50 - 200 cm^3/min were removed intermittently

RESULTS

A. Separation Characteristics

Steady state simulations of column characteristics were made using our computer modeling codes and measured feed stream composition, product flow rates, and previously measured HETP. Figure 2, 3, and 4 show the expected composition profile for columns I, T, and D together with the observed analytical data. It can be seen that the agreement is relatively good. At present we cannot explain why some sample data, especially within the lower section of the columns, do not conform well to the predictions whereas the reboiler analyses, in general, do conform. Similar comparisons for column H showed a much poorer agreement which probably can be attributed to dynamic behavior of the system and the resultant problems with measuring composition after there has been a system upset. Thus, in general, the predictions of the modeling codes are substantiated. As a result, we can verify the column design value of 5.0 cm for HETP.

B. Vapor Velocity and Pressure Drop

From the measured values of composition, reboiler power, and liquid level, it is possible to derive the vapor velocity of the material entering the column from the reboiler. A plot of the column pressure drop as a function of vapor velocity is shown in Figure 5. Similar measurements have been made in previous years and these data are also shown on the graph for comparison.[3]

The earlier measurements were made under conditions of total reflux, whereas the present measurements are under system recycle operation (all product streams combined and fed to the inlet). As a consequence, pressure drops would be expected to be larger due to the disturbance of the feed stream. The comparison is quite good and would seem to indicate that deuterium-tritium mixtures behave similarly to pure D_2.

PROBLEM AREAS

One area of previous difficulty lay in the interface with the external transfer pumping system which circulates the recycle D2 stream into the feed of column H (see figure 1.) Pressures developed by the recycle flow increase the pressure in the flow lines connecting the condenser of column I with to the feed of column H to the point that it was not possible to obtain flow from the condenser. It was necessary to install a small pump in the condenser product line to assure positive flow out of column I.

A second area of previous difficulty lay in the interface with the impurity injection and removal systems. Condensable impurity concentrations of more than a few ppm will result in rapid plugging of feed lines. Many runs were unsatisfactory because plugging of some line occurred.

On a lesser scale, it has not always been possible to maintain desired flow from the reboiler of column I to the feed of column T. The product lines are obviously somewhat undersized. This should be correctable by the creation of a larger pressure drop between the two columns. While this might be accomplished by changing the operating pressure levels of the two columns, this has proven difficult to control. Therefore it will be necessary to install a pump in this line to create the pressure difference.

Flow meters constitute another problem area. Currently installed flowmeters have unstable zero readings and are of such a design that zero and gain adjustments must be made within the glovebox. Replacements are on hand and awaiting installation.

The helium refrigerator needs better stabilization. Small variations in expansion engine speed can result in changes in cooling power supplied to the columns which exceed the dynamic range of the temperature control heaters.

While the gas chromatograph analytical system is satisfactory for the present experiments, the 30-40 minute delay to receive results is not satisfactory to effect really stable control. Furthermore, peak elution times are dependent on composition which complicates automation of the process. A Raman spectroscopic analytical system is under development which will permit analyses in less than one minute. In addition, such an instrument is absolute, not requiring calibration with standard mixtures.

CONCLUSIONS

A very successful period of full operation of the TSTA ISS has been completed. The results compare well with theoretical modeling results. The previously determined value of 5.0 cm for HETP is substantiated. Pressure drops measured for pure D_2 may be used to predict operation with D-T mixtures.

REFERENCES

[1] J.R. Bartlit, W.H. Denton, and R.H. Sherman "Hydrogen Isotope Distillation System for the Tritium Systems Test Assembly", Proc. 3rd Topl. Mtg. Technology of Controlled Nuclear Fusion, Santa Fe, NM, May 9-11, 1978, CONF-780508, Vol.2, pp 778-783.

[2] T. Yamanishi, H. Yoshida, T. Naito, S. Hirata, Y. Naruse, R.H. Sherman, J.L. Anderson, and J.R. Bartlit, "Single Column and Two-Column H-D-T Distillation Experiments at TSTA", Fusion Technology, in press.

[3] R.H. Sherman, J.R. Bartlit. and D.K. Veirs, "Experimental Results from H/D Distillations at the Tritium Systems Test Assembly", Fusion Technology 6(625)1984.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

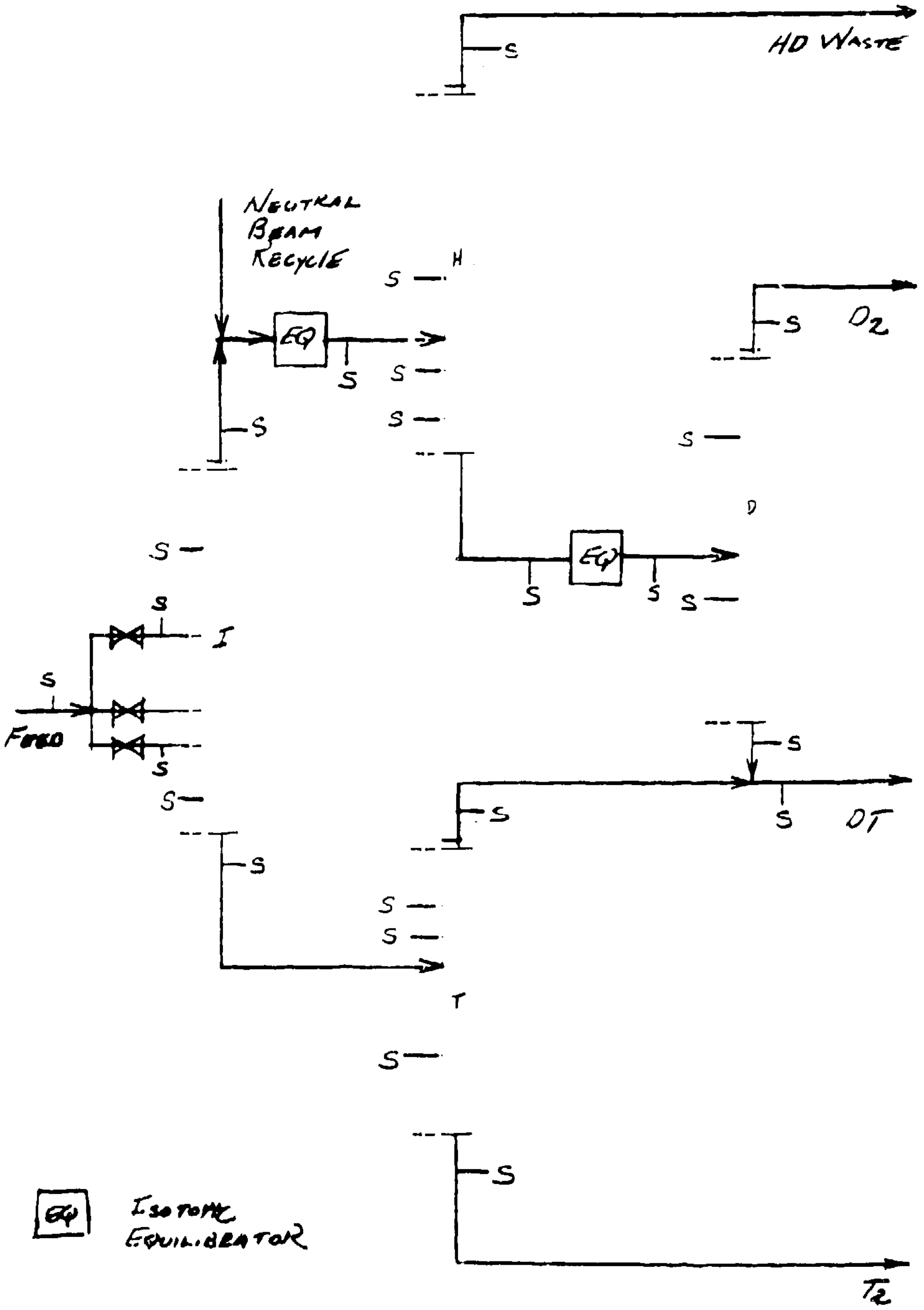
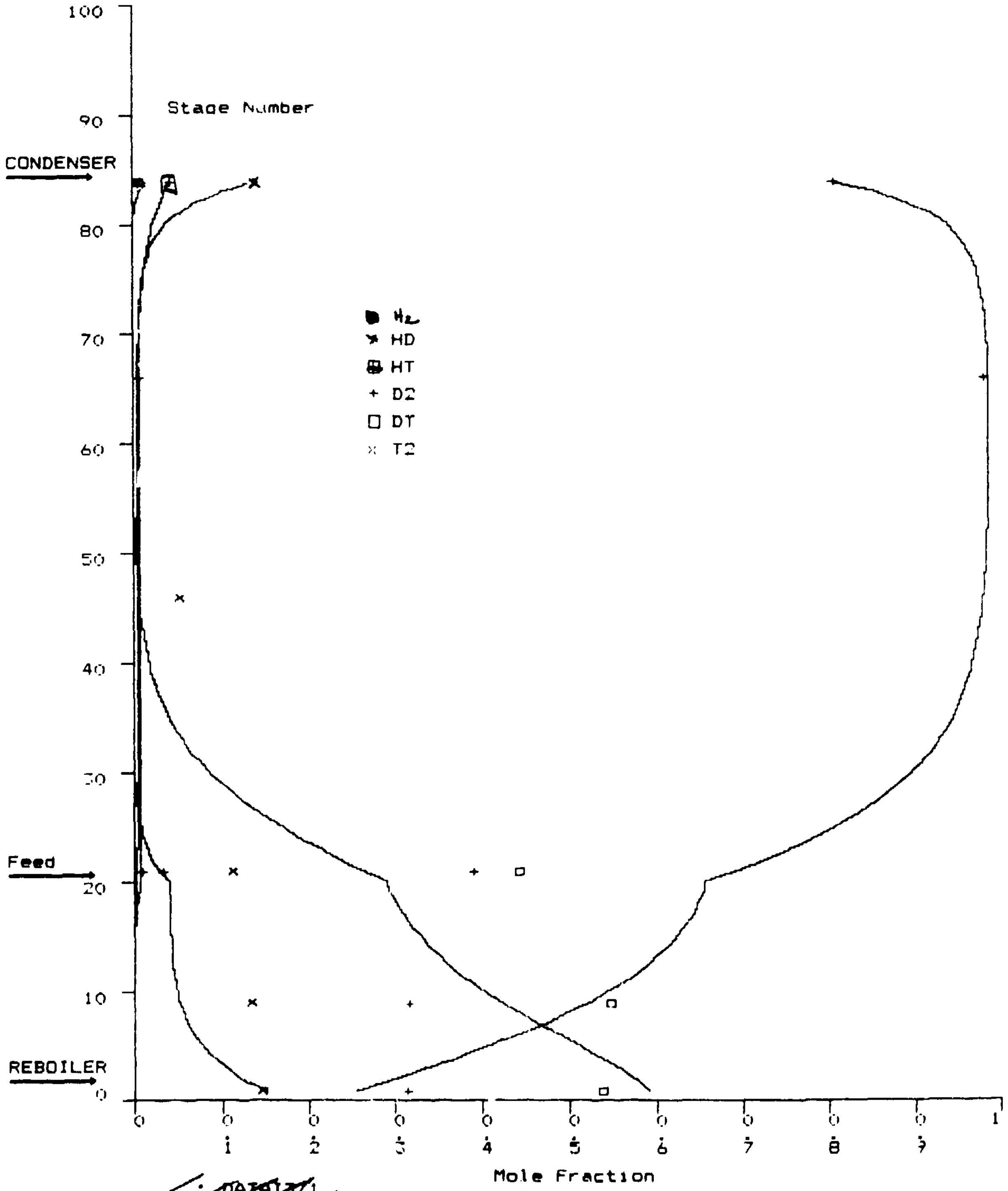


FIGURE 1.

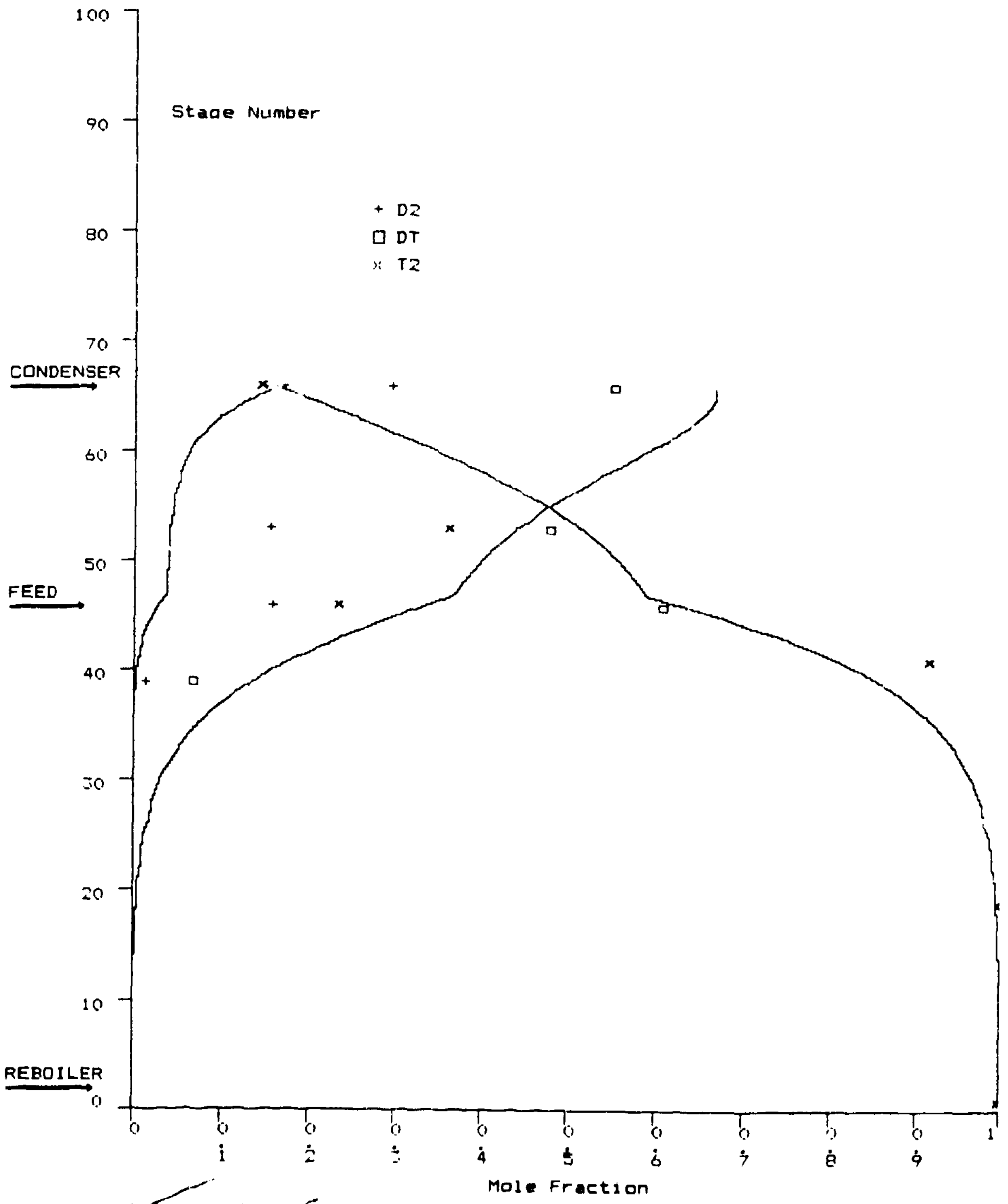
Schematic Diagram of the Isotope Separation System showing Feed and Sample Taps. Relative heights are to scale.



~~File : DATA13/1~~
~~: III.PLT~~
~~: 4/ 7/1988~~

Figure 2.

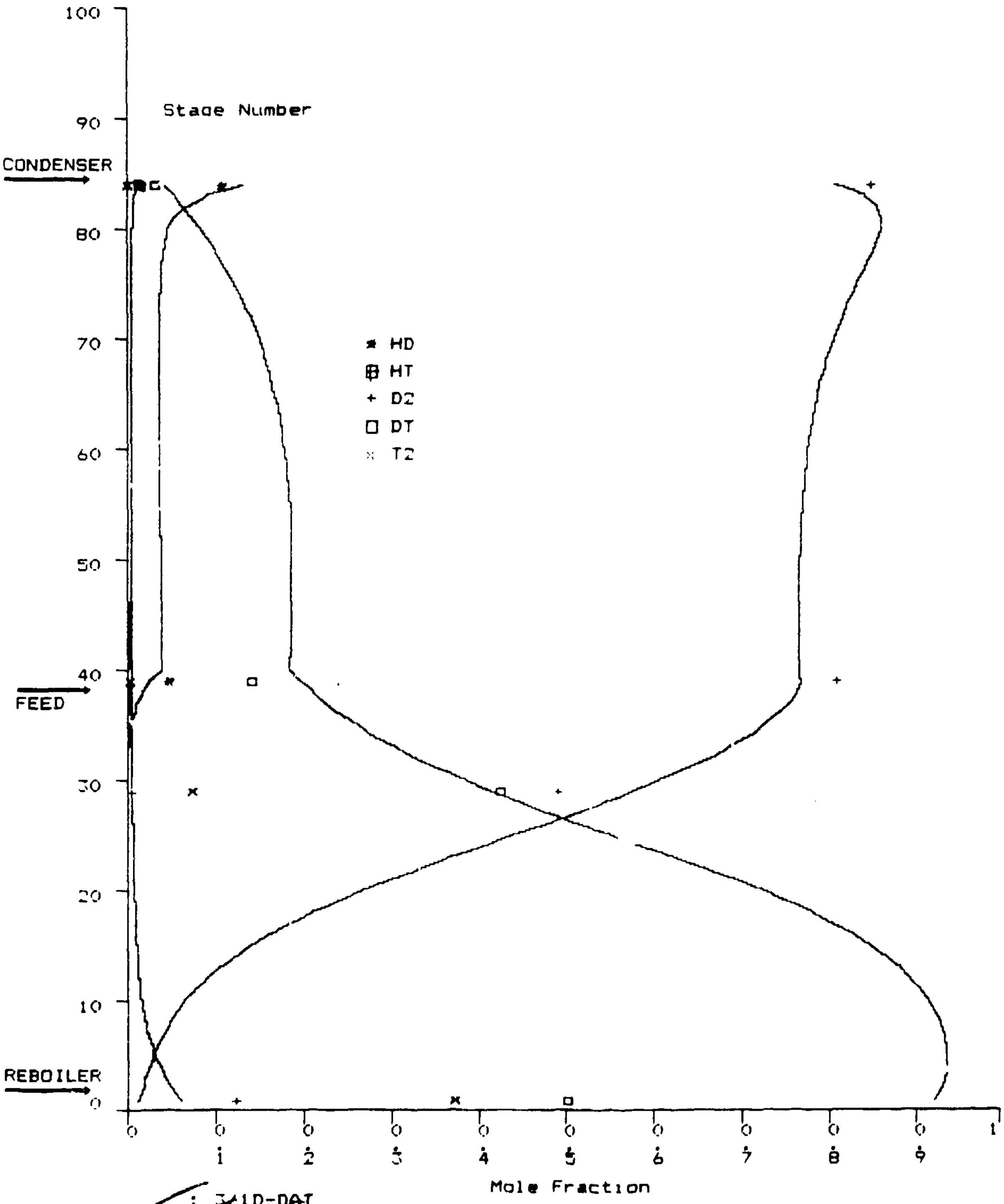
Column I. Comparison of analyses with modeled composition profile



~~File : T2P.PLT~~ ~~11/3/1988~~

Figure 3.

Column T. Comparison of analyses with modeled composition profile



~~File : 3/1D-DAT~~
~~D2D.PLT~~
~~[illegible] 7/1988~~

Figure 4.

Column D. Comparison of analyses
with modeled composition profile

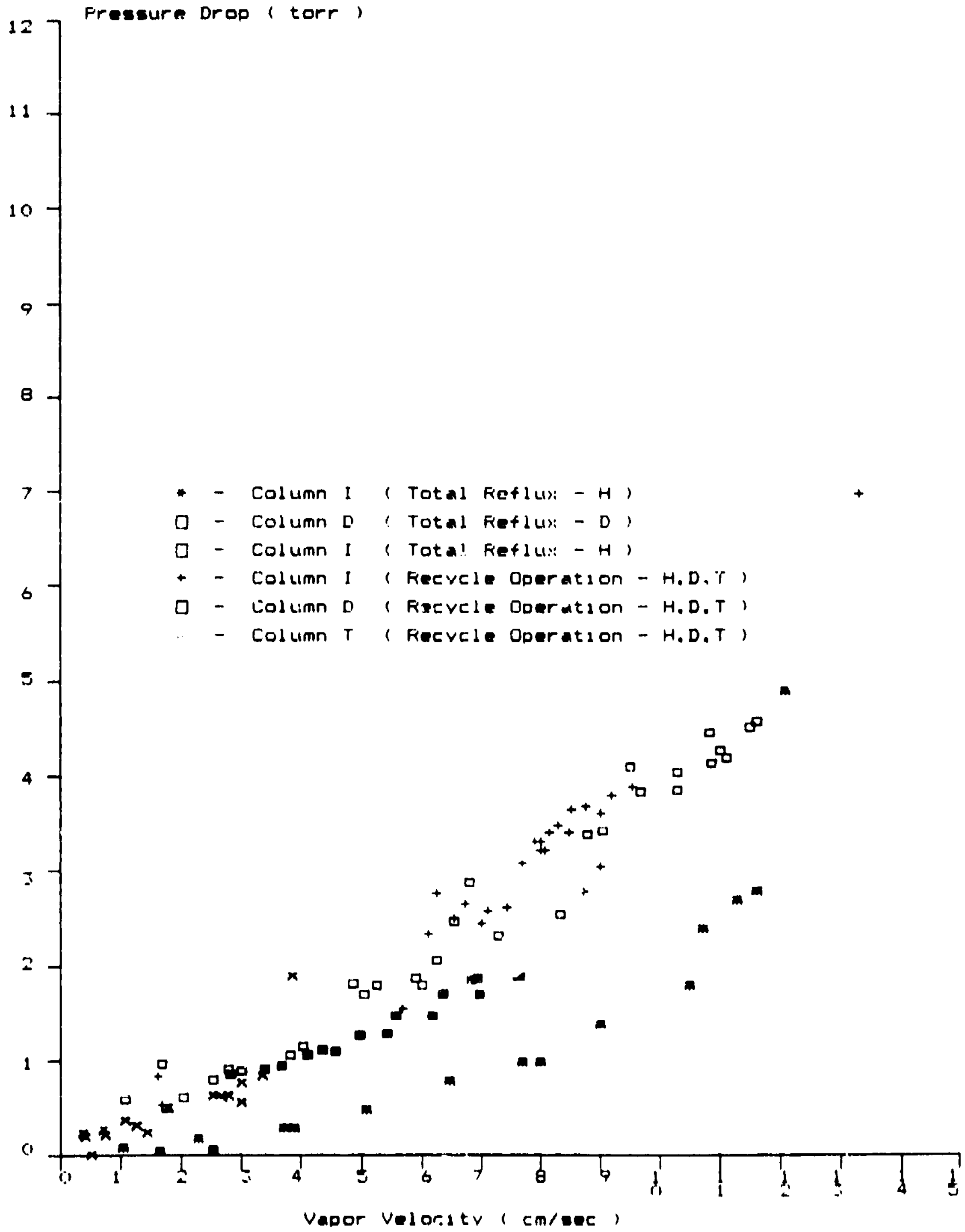


Figure 5.

Observed Vapor Velocity for H,D,T operation, compared with earlier single Substance/Column Data