Fusion Engineering and Design 169 (2021) 112452

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Dynamic simulation tools for isotopic separation system modeling and design

Eduardo Iraola [a,b,*], José M. Nougués [a], Luis Sedano [c], Josep A. Feliu [a], Lluís Batet [b]

[a] Inprocess Technology & Consulting Group, Gran Via de Carles III, 86, 08028 Barcelona, Spain
[b] ETSEIB, Universitat Politècnica de Catalunya, BarcelonaTech (UPC), Department of Physics, Diagonal 647, 08028 Barcelona, Spain
[c] FUS_ALIANZ Science, Engineering & Consulting, C/ Nord 19, Àtic, 43700, El Vendrell, Tarragona, Spain

ARTICLE INFO

Keywords:
Isotope separation system
Tritium
Aspen HYSYS
Dynamic simulation

ABSTRACT

Cryogenic distillation is the best candidate for hydrogen isotopic separation in fusion power plants. The design of the ITER's Isotope Separation System can still undergo major changes due to its close contact with the Water Detritiation System and this will be decisive in the design of future DEMO detritiation facilities. Dynamic simulation tools are key in the analysis and design of tritium processing systems and the use of commercial simulators can be especially valuable to this end and may add capabilities in terms of standardization of industrial-scale fusion plant modeling and conjunction of operator training systems, human-machine interface and process monitoring and control. The present work proposes the use of a commercial dynamic process simulator such as Aspen HYSYS given its abilities to overcome the challenges described. This document presents the implementation of a cryogenic distillation column in Aspen HYSYS by setting the thermodynamic principles upon which the simulation is founded and verifying isotopic separation models with experimental data available in the literature.

1. Introduction

The use of cryogenic distillation technology is based on working at very low temperatures (lower than 150 K) where some gases boil under atmospheric pressure conditions and can thus be separated. For the ITER plant and the forthcoming DEMO, cryodistillation is intended as a continuous multi-stage process with a feed of a mixture of different hydrogen isotopologues and extraction of the most volatile and least volatile components separately. In practice, the process is embodied in boiler/condenser cascades in packed columns. Cryodistillation is the best technology available for the separation of high amounts of hydrogen and it is necessary for a feasible tritium fuel cycle [1].

Dynamic simulation is a widespread tool in the industry for the design and study of processes, and it can be very valuable in the tritium field. This work aims to use experimental data as the first step in modeling tritium processes using a commercial simulator. There are a few experimental documents available in literature related to tritium cryodistillation, such as those of the Tritium Laboratory of Karlsruhe [2] and the National Research and Development Institute for Cryogenic and Isotopic Technologies of Romania [3]. These usually show data regarding overall column geometry and general operating conditions at steady-state operating conditions. However, transient data, necessary to verify dynamic models, is rarely available.

Nonetheless, some literature exists that provides of dynamic operation data. It is the case of the Los Alamos National Laboratory series of publications in collaboration with the Japan Atomic Energy Research Institute [4–6]. Sherman et al. [5] provide two experiments related to the operation of a distillation column with sidestream recycle through equilibrator: the first working with an H-D mixture and the second with an H-D-T mixture, although the latter is incomplete. In this work, we will focus on the first of them and replicate its behavior in simulation.

Aspen HYSYS is the commercial process simulation tool selected to perform the stated work. This software suite has been intensively validated through the years since its creation in the 1990s [7]. This simulator, typically used for refinery and petrochemical related processes, allows use of general unit operations present in many chemical plants such as compressors, vessels, pumps, pipes and distillation columns, all of them present in a tritium plant. Its internal database holds a huge set of thermodynamic data that is necessary for a plant simulator. Its connectivity also involves high potential as a tool for operator training, using it as a simulator of actual operation and for real-time process monitoring in an actual plant as a master monitoring and alarm

* Corresponding author at: Inprocess Technology & Consulting Group, Gran Via de Carles III, 86, 08028 Barcelona, Spain.
E-mail address: eduardo.iraola@inprocessgroup.com (E. Iraola).

https://doi.org/10.1016/j.fusengdes.2021.112452
Received 1 December 2020; Received in revised form 3 March 2021; Accepted 4 March 2021
Available online 21 March 2021
0920-3796/© 2021 Elsevier B.V. All rights reserved.

Table 1
Regressed two-alpha coefficients [10].

Isotopologue	L	M	N
H_2	0.7189	2.5411	10.2000
HD	0.1009	1.0204	1.9102
HT	0.6820	1.2469	0.2000
D_2	1.2584	6.1846	0.0759
DT	0.9783	1.6726	0.2000
T_2	1.0943	1.6009	0.2000

management system. Other products in the Aspentech suite [8] such as Aspen Plus have already started to be selected in previous documents in the literature as the main tool for simulating scenarios in tritium-related environments [9–12]. In contrast with them, Aspen HYSYS shows advantages such as providing tools for continuous analysis from steady-state to dynamic modeling, which allows covering the whole design-cycle of a process.

The present document is organized as follows. Section 2 performs a review of the available data needed to perform a dynamic simulation of tritium cryodistillation. Section 3 shows the modeling process of the experimental distillation column to be benchmarked. Finally, Sections 4 and 5 show and discuss the simulation results obtained in the work.

2. Input data

Data is needed both for the characterization of the chemical species that may appear in the cryogenic columns and for the process itself. This section shortly reviews the necessary inputs for later modeling.

2.1. Physical data

The Peng-Robinson equation of state [13] (Eq. (1)) is an equation of state model with parameters that can be expressed by using the critical properties (T_c, P_c) and the acentric factor ω of each pure component as presented in the set of Eq. (2).

$$P = \frac{RT}{v-b} - \frac{a\alpha}{v^2 + 2bv - b^2} \tag{1}$$

$$a = 0.45723553\frac{R^2 T_c^2}{P_c} \tag{2a}$$

$$b = 0.07779607\frac{RT_c}{P_c} \tag{2b}$$

$$\alpha = \left[1 + m\left(1 - T_r^{0.5}\right)\right]^2 \tag{2c}$$

$$m = 0.37464 + 1.54226\omega - 0.26992\omega^2 \tag{2d}$$

being T_r the reduced temperature:

$$T_r = \frac{T}{T_c} \tag{3}$$

The Peng-Robinson (PR) equation is especially appropriate for hydrocarbons and the coefficients in Eq. (2d) do not apply well to other kind of components. This drawback can be overcome by using the Twu-alpha function [14]:

$$\alpha(T) = T_r^{N/(M-1)}\exp\left[L\left(1 - T_r^{NM}\right)\right] \tag{4}$$

Noh et al. [10] found that this model fairly predicted the actual vapor pressures of hydrogen isotopologues under cryogenic conditions. The cited work fitted the three parameters *L*, *M* and *N*, unique for each pure component, to the properties of the six diatomic isotopologues for the use of the PR-Twu equation of state. Their results are shown in Table 1.

For complete use of the Peng-Robinson Twu-Alpha equation of state, the critical point of the pure substances and other properties are also needed. A review of the available literature leads to the data compilation shown in Table 2. To elaborate this selection of properties, it has been preferred to point at each individual reference rather than general compilations, whenever possible.

Table 2
Physical properties of the six hydrogen isotopologues. If not specified, the source is Souers [15]. Homonuclear properties are given for normal ortho-para rotational state forms.

	H_2	HD	HT	D_2	DT	T_2
MW (g/mol) [16]	2.016	3.022	4.024	4.028	5.030	6.032
T_b (K) [17]	20.397	22.14	22.906	23.665	24.372	25.04
T_c (K)	33.19	35.91 [18]	37.13	38.35	39.42	40.22
P_c (kPa)	1313	1483.9 [18]	1570	1650	1770	1850
ρ_c (mol/m^3)	15200	15923.6 [18]	16393	16700	16949	17700

Table 3
Design data of the first column in TSTA (I) [4].

	Feed	Top product	Bottoms product
mole frac.			
H_2	0.00014	0.00053	0
HD	0.01000	0.04100	$4.70{\cdot}10^{-9}$
HT	0.00930	0.03600	$1.70{\cdot}10^{-6}$
D_2	0.24800	0.91800	0.01700
DT	0.48300	0.00470	0.64900
T_2	0.24900	$6.10{\cdot}10^{-8}$	0.33500
Flowrate (mol/h)	15.120	3.960	11.160

Table 4
Design data of the first column in TSTA (II) [4].

Number of stages	84	
Design reflux ratio	20.96	
HETP	5	cm
Packed Length	4.11	m
Inside Diameter	2.84	cm
Operating Pressure	860	Torr
Condenser Temperature	23.97	K
Reboiler Temperature	24.97	K
Condenser Duty	27.26	W
Reboiler Duty	28.08	W

In cryogenic isotope separation systems, it is generally useful to remove the intermediate species HT and DT. Withdrawing a sidestream from intermediate stages of the column and passing this stream through equilibrators at room temperature before recycling it into the column is beneficial to this objective. The sidestream recycle through equilibrator shifts the mixture composition thanks to the chemical equilibrium attained and reduces the amount of the undesired species, allowing also to reduce the number of columns necessary to obtain a certain product purity [5]. To represent these intermediate operations, we use the equilibrium constant data set provided by Jones [19]. For the specific case of the $H_2 + D_2 \leftrightarrow 2HD$ reaction, the preferred equilibrium constant value is the one at 25 °C [20].

2.2. Process data

This work is dedicated to the dynamic simulation of cryogenic distillation systems. Therefore, it was found necessary to rely on experimental dynamic data for testing. The chosen scenario was developed by Sherman et al. [5], where the first column of the Tritium Systems Test Asembly (TSTA) of the Los Alamos National Laboratory was used to demonstrate the effectiveness of the sidestream recycle equilibration. The design of the same distillation column was previously

Table 5
Characterization data for the first experiment of Sherman et al. [5].

HD Feed composition	2:1
Reflux ratio	8
Column pressure	808 Torr
Feed/Recycle Stage	29
Withdrawal stage	9
Flowrates (sccm)	
Feed	3037
Top	2971
Bottoms	66
Recycle	1008

presented the same author [4] and its main details are shown in Table 3 and Table 4. We refer to the number of theoretical plates, feed and product flow rates, feed and product compositions, feed stage, pressure and reflux ratios when talking about design data. These data will be used for the modeling of the distillation tower.

Regarding the transient operation of Sherman et al. [5], we will focus on the first experiment of this work, in which the distillation column is filled with an H-D mixture with 2:1 proportion. The column is operated at single-column mode (no sidestream equilibration) with a reflux ratio of 8 until it reaches steady-state conditions. During this period the top and bottom products are withdrawn and recombined to be reused as feed for the same column. Then, the operation of the sidestream recycle is started and run for several hours, and the evolution of the composition in the reboiler is given. The main data available for the experiment is shown in Table 5.

After observing the results of Figure 4 in the paper of Sherman et al. we determined that the feed composition of 2:1 H:D could not be given in equilibrium (the exact composition is not shown), and we estimated its actual composition by trial and error. The results are mole fractions of approximately 0.3343, 0.6647 and 0.0010 for H_2, HD, and D_2 respectively.

3. Methodology

This section will show how Aspen HYSYS can be used to model the cryogenic distillation column given the available data, from the thermodynamic data to the flowsheet modeling.

Regarding the dynamic simulation mode, Aspen HYSYS uses the implicit fixed-step size Euler algorithm as the integration algorithm. The program allows the user to set the desired time step and volume, energy and composition balances are solved at different frequencies. Specifically, volume differential equations are solved every time step, while composition and energy are solved every second and tenth time step by default. This provides a trade-off between accuracy and speed [21]. In the present work, the default values are used and a time step of 0.2 s has been shown sufficient to provide a stable simulation.

3.1. Property package design

The six isotopologues H_2, HD, HT, D_2, DT and T_2 are defined as *hypothetical components*, following the terminology used by Aspen HYSYS. This has been done by specifying the physical properties used by the PR-Twu equation of state, mainly the critical point previously shown in Table 2 and the specific PR-Twu parameters from Noh et al. [10] shown in Table 1. Furthermore, before any simulation, it is necessary to specify the thermodynamic model used by the simulation. As previously stated, the PR-Twu equation of state is chosen.

Note that, even though the hydrogen and deuterium components already exist in the HYSYS database, they are defined separately in this work for consistency between the six components that need setup.

Regarding the isotopic exchange reactions taking place in equilibrators, the individual reactions must be added to the property package to be available for use in the model. In the work developed in this document, no reactor unit operation will be used since this task will take place via spreadsheet programming, but the reaction set will be defined for later assessments where several exchange reactions take place at the same time and its manual automation is non-trivial. In this case, tabular data such as that shown in Table 6 can be input to configure equilibrium reaction calculations in the property package.

Table 6
Jones's tabular data for the definition of the equilibrium isotopic exchange reaction $H_2 + D_2 \leftrightarrow 2HD$ [19].

T (K)	K_{eq}	T (K)	K_{eq}
50	1.33	700	3.78
100	2.26	800	3.83
200	2.90	900	3.87
298	3.26	1000	3.90
300	3.26	1250	3.94
400	3.48	1500	3.96
500	3.62	2000	3.97
600	3.72	2500	3.97

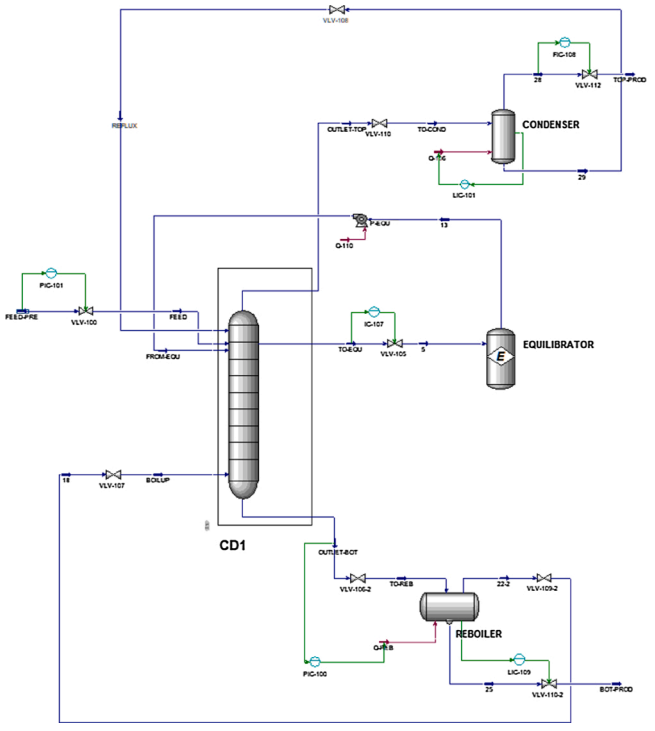

Fig. 1. Aspen HYSYS flowsheet of the dynamic cryogenic distillation column.

Table 7
Mole fraction comparison between design and steady-state model in Aspen HYSYS.

	Feed		Top Product		Bottoms Product	
	Design	HYSYS	Design	HYSYS	Design	HYSYS
H_2	0.00014	0.00014	0.00053	0.00053	0	0
HD	0.01000	0.01001	0.04100	0.03819	$4.70{\cdot}10^{-9}$	$3.95{\cdot}10^{-9}$
HT	0.00930	0.00931	0.03600	0.03552	$1.70{\cdot}10^{-6}$	$9.42{\cdot}10^{-7}$
D_2	0.24800	0.24814	0.91800	0.90018	0.01700	0.01674
DT	0.48300	0.48327	0.00470	0.02558	0.64900	0.64571
T_2	0.24900	0.24914	$6.1 \cdot 10^{-8}$	$5 \cdot 10^{-7}$	0.33500	0.33754

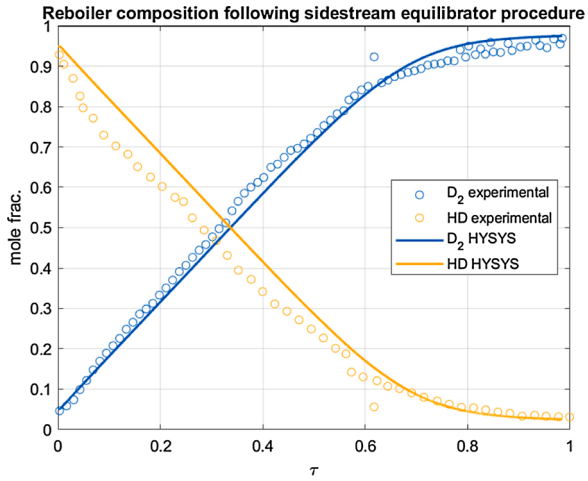

Fig. 2. Comparison of transient results between experimental and simulation data. The horizontal axis shows the dimensionless time parameter and the vertical axis represents the mole fraction of the mixture in the reboiler.

3.2. Cryodistillation modeling

This section will show the development of a scaled model of the column described in Section 2.2. The model will be sized so that it processes ten times the design feed shown by Sherman et al. This scaling has two purposes: to provide better convergence for the Aspen HYSYS numerical solver, and to let us build a verified model of a cryodistillation column working in similar conditions as those envisaged in ITER—the ITER Isotope Separation System is foreseen to process a maximum plant throughput between 150 Pa m^3/s and 200 Pa m^3/s [22,1], approximately ten times the TSTA design feed.

First, a shortcut distillation object fed with the design feed composition is used to check if the property package is well-defined. The obtained product flows, purity, number of theoretical stages and condenser and reboiler flow rate shall agree with the input design.

The next step involves defining a new steady-state distillation column flowsheet object specified by the number of stages, pressure profile and reflux and product rate specification. The results shall match those of the shortcut design (duties and output flow rates) and also let the user have a first glance of the internal profiles estimated for the column. Of special interest are the reboiler and condenser operating conditions.

After the steady-state analysis, the model is moved to the dynamic simulation environment. In this step, it is chosen to separate the column environment from the condenser and reboiler objects as shown in Fig. 1 for further flexibility of the model.

Here, PI controllers are set up to govern the feed flow, the top pressure of the column, product flows and the liquid level of both reboiler and condenser unit operations. General recommendations made by Yamanishi et al. [23] for configuring the control architecture of the system are followed. Geometric considerations and static head now contribute to the pressure-flow solver and the packing internals of the column are then adapted to fit a pressure drop of approximately 5 Torr as stated by Sherman [24] along the column. The column height is set to the original 4.11 m packed length and the diameter is sized to fit the scaled throughput. The transient simulation is run until steady-state and product purity of Table 7 is achieved.

The previous steps led to obtaining a functional model of a column operating at the design H-D-T feed conditions. In order to transition to the H-D conditions of the experiment to be reproduced, the column is injected with the new feed conditions until all tritium is withdrawn from the system. Controllers are adjusted to shift to the reflux and product flows previously shown in Table 5.

4. Results

The model developed in Section 3 is used in this section to reproduce the experimental transient results.

As described in the original procedure, the equilibrator starts working after a period of steady-state operation of the column. The sidestream equilibration is kept for several hours. In the simulation, the sidestream is withdrawn at the 9th stage and returned through the feed stage. The isotopic exchange equilibrium for the $H_2 + D_2 \leftrightarrow 2HD$ reaction at 25 °C of room temperature is calculated with the data available in Section 2. The comparison between the original data and the actual transient simulation is shown in Fig. 2.

5. Conclusions

In this work, the Twu-Alpha parameters from Noh et al. [10] were used and verified in the Aspen HYSYS simulator to replicate a transient operation experiment of a cryodistillation isotope separation column changing from single-column operation to sidestream recycle operation.

Non-idealities not represented in the PR-Twu equation of state can lead to small discrepancies with real experiments. However, the use of the equation of state for hydrogen isotopologues allows fast and efficient simulations of cryogenic systems with fair accuracy and the main transient behavior is replicated in the simulations. Further work involves testing scenarios in which small quantities of HT play an important role and, therefore, assessing the accuracy of the model. Next steps will also include adjusting the binary interaction parameters, not considered in this document, to improve the vapor-liquid equilibrium performed by the Peng-Robinson equation of state.

There have been few attempts of modeling tritium processing systems using commercial simulators such as Aspen HYSYS. Aspen HYSYS has the advantage of yielding accurate and fast calculation performance, which will be critical when coupling several cryodistillation columns to simulate a complete Isotopic Separation System and when creating an interface to work as an Operator Training System. The results shown in this document open a path to stakeholders in the industry to become new agents in future fusion projects, taking advantage of their expertise in complex chemical processes.

Declaration of Competing Interest

The authors report no declarations of interest.

Acknowledgements

This work has been possible thanks to co-funding of the Centro para el Desarrollo Tecnológico Industrial of the Spanish Ministry of Science and Innovation (IDI-20200750) and to the Industrial Doctorates Plan of the Government of Catalonia (2018 DI 0048).

References

[1] M. Glugla, A. Antipenkov, S. Beloglazov, C. Caldwell-Nichols, I.R. Cristescu, I. Cristescu, C. Day, L. Doerr, J.-P. Girard, E. Tada, The ITER tritium systems, Fusion Eng. Des. 82 (2007) 472–487, https://doi.org/10.1016/j.fusengdes.2007.02.025.

[2] I. Cristescu, A. Bükki-Deme, R. Carr, N. Gramlich, R. Groessle, C. Melzer, P. Schaefer, S. Welte, Review of the TLK activities related to water detritiation, isotope separation based on cryogenic distillation and development of barriers against permeation, Fusion Sci. Technol. 71 (2017) 225–230, https://doi.org/10.1080/15361055.2017.1288057.

[3] A. Bornea, M. Zamfirache, L. Stefan, I. Stefanescu, A. Preda, Experimental investigation on hydrogen cryogenic distillation equipped with package made by ICIT, Fusion Sci. Technol. 67 (2015) 266–269, https://doi.org/10.13182/FST14-T7. URL: https://www.tandfonline.com/doi/full/10.13182/FST14-T7.

[4] R.H. Sherman, Cryogenic hydrogen isotope distillation for the fusion fuel cycle, Fusion Technol. 8 (1985) 2175–2183, https://doi.org/10.13182/FST85-A24605.

[5] R.H. Sherman, D.J. Taylor, J.W. Barnes, T. Yamanishi, M. Enoeda, S. Konishi, K. Okuno, Application of sidestream recycle to the separation of hydrogen isotopes

by cryogenic distillation, Proc. Symp. Fusion Eng. 1 (1993) 77–79, https://doi.org/10.1109/fusion.1993.518287.
[6] R.H. Sherman, D.J. Taylor, T. Yamanishi, M. Enoeda, S. Konishi, K. Okuno, The role of sidestream recycle in hydrogen isotope separation and column cascade design, Fusion Eng. Des. 28 (1995) 392–396, https://doi.org/10.1016/0920-3796(95)90066-7.
[7] G. Sobočan, P. Glavič, Optimization of ethylene process design, in: R. Gani, S. B. Jørgensen (Eds.), European Symposium on Computer Aided Process Engineering – 11, vol. 9, Elsevier, 2001, pp. 529–534, https://doi.org/10.1016/S1570-7946(01)80083-3.
[8] Aspentech, Aspen HYSYS, 2019. https://www.aspentech.com/en/products/engineering/aspen-hysys.
[9] M. Aldehani, F. Alzahrani, M.N.A. TSaoir, D.L.A. Fernandes, S. Assabumrungrat, F. Aiouache, Kinetics and reactive stripping modelling of hydrogen isotopic exchange of deuterated waters, Chem. Eng. Process. Process Intensif. 108 (2016) 58–73, https://doi.org/10.1016/j.cep.2016.07.008.
[10] J. Noh, A.M. Fulgueras, L.J. Sebastian, H.G. Lee, D.S. Kim, J. Cho, Estimation of thermodynamic properties of hydrogen isotopes and modeling of hydrogen isotope systems using Aspen Plus simulator, J. Ind. Eng. Chem. 46 (2017) 1–8, https://doi.org/10.1016/j.jiec.2016.07.053.
[11] S. Reyes, T. Anklam, W. Meier, P. Campbell, D. Babineau, J. Becnel, C. Taylor, J. Coons, Recent developments in IFE safety and tritium research and considerations for future nuclear fusion facilities, Fusion Eng. Des. 109-111 (2016) 175–181, https://doi.org/10.1016/j.fusengdes.2016.03.034.
[12] J. Wilson, J. Becnel, D. Demange, B. Rogers, The ITER Tokamak exhaust processing system design and substantiation, Fusion Sci. Technol. (2019) 1–8, https://doi.org/10.1080/15361055.2019.1642089, 00.
[13] D.-Y. Peng, D.B. Robinson, A new two-constant equation of state, Ind. Eng. Chem. Fundam. 15 (1976) 59–64, https://doi.org/10.1021/i160057a011.
[14] C.H. Twu, J.E. Coon, J.R. Cunningham, A new cubic equation of state, Fluid Phase Equilib. 75 (1992) 65–79, https://doi.org/10.1016/0378-3812(92)87007-A.
[15] P.C. Souers, Hydrogen Properties for Fusion Energy, University of California Press, 1986.
[16] D.R. Burgess Jr., Thermochemical Data, in: P. Linstrom, W. Mallard (Eds.), NIST Chemistry WebBook, NIST Standard Reference Database Number 69, National Institute of Standards and Technology, Gaithersburg MD, 2020, p. 20899, https://doi.org/10.18434/T4D303. https://webbook.nist.gov/chemistry/.
[17] P.C. Souers, Cryogenic hydrogen data pertinent to magnetic fusion energy, Technical Report, California University, Livermore, United States, 1979. http://inis.iaea.org/search/search.aspx?orig_q=RN:10490410.
[18] H. Hoge, J. Lassiter, Critical temperatures, pressures, and volumes of hydrogen, deuterium, and hydrogen deuteride, J. Res. Natl. Bureau Stand. 47 (1951) 75, https://doi.org/10.6028/jres.047.010.
[19] W.M. Jones, Thermodynamic functions for tritium deuteride. The dissociation of tritium deuteride. equilibria among the isotopic hydrogen molecules, J. Chem. Phys. 17 (1949) 1062–1064, https://doi.org/10.1063/1.1747113.
[20] M. Kinoshita, Y. Naruse, Parameter setting method for control system of cryogenic distillation column, J. Nucl. Sci. Technol. 18 (1981) 595–607, https://doi.org/10.1080/18811248.1981.9733295.
[21] Aspen Technology, Aspen HYSYS Dynamic Modeling Guide. Technical Report, Burlington, MA, USA, 2011.
[22] M. Glugla, D. Murdoch, A. Antipenkov, S. Beloglazov, I. Cristescu, I.-R. Cristescu, C. Day, R. Laesser, A. Mack, ITER fuel cycle R&D: consequences for the design, Fusion Eng. Des. 81 (2006) 733–744, https://doi.org/10.1016/j.fusengdes.2005.07.038.
[23] T. Yamanishi, M. Enoeda, K. Okuno, R.H. Sherman, A control method for the cryogenic distillation column with a feedback stream, Fusion Technol. 29 (1996) 232–243, https://doi.org/10.13182/FST96-A30710.
[24] R.H. Sherman, J.R. Bartlit, D.K. Veirs, Experimental results from hydrogen/deuterium distillations at the tritium systems test assembly, Fusion Technol. 6 (1984) 625–628, https://doi.org/10.13182/FST84-A23144.