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Estimation of thermodynamic properties of hydrogen isotopes and modeling of hydrogen isotope systems using Aspen Plus simulator

Jaehyun Noh[a], Alyssa Marie Fulgueras[a], Leah Jessica Sebastian[a], Hyeon Gon Lee[b], Dong Sun Kim[a], Jungho Cho[a,*]

[a] Department of Chemical Engineering, Kongju National University, 275 Budae-dong, Seobuk-gu, Cheonan-si, Chungcheongnam-do 331-717, South Korea
[b] National Fusion Research Institute (NFRI), 169-148 Gwahak-ro, Yuseong-gu, Daejeon-si 305-806, South Korea

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ABSTRACT

Physical properties of hydrogen isotopes, hydrogen (H_2), hydrogen-deuterium (HD), hydrogen-tritium (HT), deuterium (D_2), deuterium-tritium (DT), and tritium (T_2) were estimated through vapor pressure prediction, and validated by steady-state simulation of ITER isotope separation system (ISS). Peng–Robinson (PR) equation of state with Twu alpha function was selected for modelling which showed favorable prediction from the experimental vapor pressures of each hydrogen isotopes. The steady-state simulation of ITER ISS using Aspen Plus consists of four distillation columns and seven equilibrium reactors with four purified products: D_2, T_2, HD, and DT. Converged solution from simulation produced potential scenario for actual ITER ISS process.

Introduction

For the practical production of fusion energy, seven entities, South Korea, the United States, European Union, China, Russia, Japan, and India, have gathered for the international nuclear fusion experiment as a joint development project in progress. The International Thermonuclear Experimental Reactor (ITER) project implies about achieving the road from plasma physics to production of nuclear fusion power, in which the word “iter” also means “way” in Latin. Fig. 1 is a conceptual diagram showing the ITER tritium fuel cycle. The gas fuel with several other components including tritium is fed to a tokamak’s torus chamber which consists of separation/purification/recovery system of components, and then mixed again with the emitted gas. Tokamak uses strong magnetic field to confine plasma into a stable flow of plasma current in a donut-shaped vacuum chamber. The tokamak torus chamber is surrounded by magnetic field coils and transformers. Nuclear fusion is generated by injecting gas inside the vacuumed torus, heating by magnetic and electric field to form high-temperature plasma, and adding microwaves to further increase the temperature and squeeze the plasma. Tritium, which is introduced into a torus, is released combined with other gases containing hydrogen isotopes from nuclear fusion reaction and proliferation process. The gas mixture discharged from the torus proceeds to the tokamak exhaust processing (TEP) to separate the hydrogen isotopes with other impurities, while the deuterium and tritium are separated by cryogenic distillation in the isotope separation system (ISS). Then, the separated hydrogen isotopes through ISS are supplied for storage and delivery system (SDS). In the water detritiation system (WDS), the remaining amount of deuterium and tritium are recovered, and qualitative–quantitative analysis is performed in the analysis system (ANS). In the fueling system (FS) and neutral beam injector (NBI), the functions supplying the torus should be depended on the application of the gas supplied from the SDS. The gases to be fed into the nuclear fusion reaction as fuel are T_2, D_2 and DT, and the gases to be injected in order to stop the nuclear fusion reaction are Ne, Ar, He, O_2 and N_2, and the like [1–5]. This research proceeds with the study of the ISS process.

ITER ISS is the system for purifying the desired component and composition using cryogenic distillation and the equilibrium reaction in which gas mixture of hydrogen isotopes from TEP and WDS are fed [6,7]. On the other hand, since cryogenic distillation is used in the ISS, there is a significant amount of hydrogen isotopes being liquefied, and longer retention time of tritium holdup during operation is the best process. Helium refrigerator is used to operate at low temperature of about 15–20 K. From TEP and WDS to ISS, the main compositions to be introduced

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[*] Corresponding author.
E-mail addresses: jhcho@kongju.ac.kr, pronjh1217@naver.com (J. Cho).

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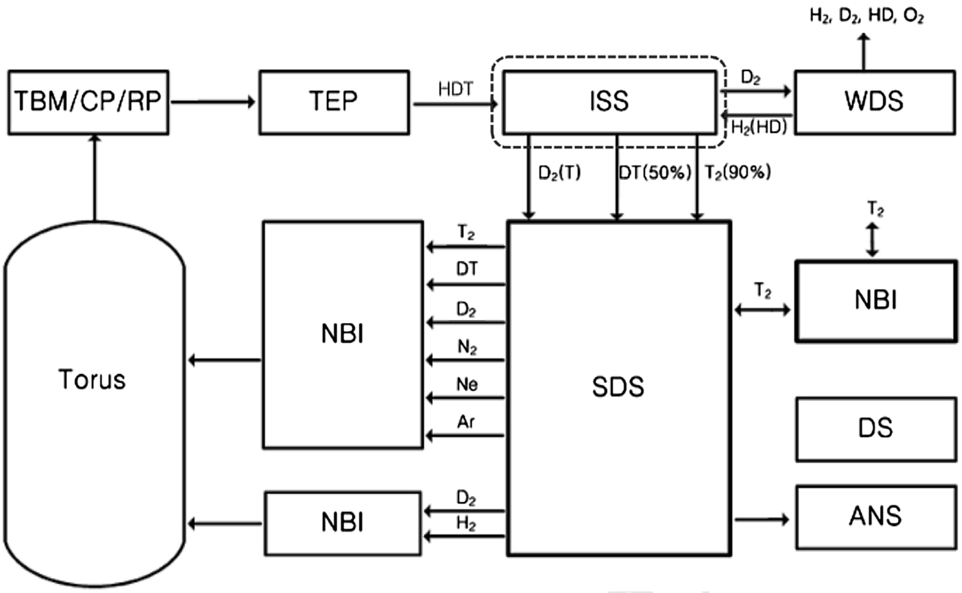

Fig. 1. Block diagram of ITER fuel cycle.

are the 6 hydrogen isotopes: H_2, HD, HT, D_2, DT, and T_2. In the ISS process, out of the six mixed gas, D_2, DT, and T_2, which are used as a raw material for ITER, should be purified with the desired compositions. To produce the desired composition of D_2, DT, and T_2 from ISS, a total of four distillation columns and several equilibrium reactors (Equilibrator) should be installed. In the equilibrium reactor, the following reversible reactions take place: $2HD \leftrightarrow H_2 + D_2$, $2HT \leftrightarrow H_2 + T_2$, and $2DT \leftrightarrow D_2 + T_2$.

Table 1, adapted from Song et al. [8], shows the equilibrium constant of isotopes based on temperature for each equilibrium reaction. Generally, these equilibrium reactions have slow reaction rate at very low temperature and fast reaction rate at room temperature in terms of D_2 and T_2 generation. The desired molecular bond form can lead to a reaction when the supplied composition was set in the equilibrium reactor by optimizing the position of the equilibrium reactor [9].

ITER ISS process uses a cryogenic distillation and catalytic reaction from temperatures 14 to 30 K. Catalytic reaction takes place in equilibrium in which the reactor should be installed in an optimum position depending on the concentration profile of the column. In addition, with WDS linked to the ISS process, a highly complex optimization technique will be required. Therefore, in order to design and optimize this process, process simulation should be performed. However, in Korea, there are few to none organizations that have performed process simulation of the cryogenic distillation process for the ITER ISS. One of the reasons is because domestic technology for the ISS process is still in the concept establishing phase and international technology also failed to facilitate the tracking of related data which is critically low. Another reason is the lack of technology and the physical property data to perform the cryogenic distillation process simulation. In this study, the necessary physical properties for H_2, HD, HT, D_2, DT, and T_2 components were obtained using the equation of state which allows the simulation of the ISS process.

Estimation of thermodynamic properties

Estimation of fixed properties of pure components

As shown in Table 2, the physical properties of the pure component for hydrogen isotopes, such as H_2, HD, D_2, HT, DT and T_2, must be obtained in order to estimate the phase equilibrium properties using the equation of state such as Peng–Robinson [10] and Soave–Redlich–Kwong [11]. However, from the compatibility of chemical process simulator such as Aspen Tech Corporation's Aspen Plus, Invensys' PRO/II with PROVISION, etc., only few properties are built-in for H_2, HD and D_2, and no properties are available for HT, DT, and T_2. Thus, in this study, to estimate the thermodynamic properties of H_2, HD, D_2, HT, DT, and T_2 required for the separation process, related experimental data were collected which were used for regression analysis to obtain model equation parameters, and, finally, to estimate thermodynamic properties (Table 3).

The pure component properties for the six hydrogen isotopes based on comprehensive study of experimental data available in literature [12–14] are shown in Tables 3 and 4. The Gibbs free energy for each of the component is presented in which the value for pure elements, H_2, D_2 and T_2, are defined as zero (0), while the values for all the other compounds are defined by a specific value. The value of Gibbs free energy of formation, $\Delta G^{\circ}_{f,i}$, for the compounds are estimated from the standard Gibbs free energy change for a given chemical reaction. With these physical property values, the calculation for the equilibrium reactor is carried out (Table 4).

Table 1
Equilibrium constant for isotopes.

Reaction formula	Equilibrium constant				
	273 K	298 K	400 K	500 K	600 K
$[H_2][D_2]/[HD]^2$	3.18	3.25	3.48	3.62	3.72
$[D_2][T_2]/[DT]^2$	3.79	3.82	3.88	3.92	3.94
$[H_2][T_2]/[HT]^2$	2.42	2.56	2.97	3.24	3.44

Table 2
Fixed properties for using equation of state model.

Parameter	Description
T_c	Critical temperature
P_c	Critical pressure
ω	Acentric factor

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