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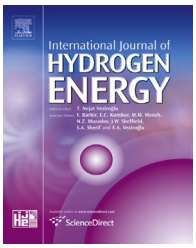

Simulation studies of the characteristics of a cryogenic distillation column for hydrogen isotope separation

Rupsha Bhattacharyya[*], *Kalyan Bhanja*, *Sadhana Mohan*

Heavy Water Division, Bhabha Atomic Research Centre, Trombay, Mumbai 400 085, Maharashtra, India

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ABSTRACT

Cryogenic distillation is a very widely applied technique for separating the components of a mixture of hydrogen isotopes, which may contain up to six isotopic species. This work presents a simple dynamic model for evaluating the major performance characteristics of a cryogenic distillation column for hydrogen isotope separation. Liquid hold up, pressure drop and flooding point have been predicted for the tower using known packing characteristics and fluid properties. The column has been modelled as an equivalent tray tower using a known value of the height equivalent to theoretical plate (HETP) and its dynamic behaviour and separation performance under various conditions have been predicted. The effect of parameters like temperature, reflux ratio, feed point location and feed composition on the final isotopic contents of top and bottom products at steady state has also been studied. Consequences of neglecting the decay heat of tritium in distillation calculations have also been evaluated. This model is intended for preliminary column design work using data from literature, without the need for prior experimental activity.

Introduction

Separation of the isotopes of hydrogen has always been an important unit operation in the field of nuclear energy production. For use as the coolant and moderator in pressurized heavy water based reactors (PHWRs) employing natural uranium oxide as the fissile material, the separation and concentration of heavy water from natural sources of light water is a well known and mature process technology, using unit operations like vacuum distillation and catalytic exchange [1]. For the fusion energy field, gaseous hydrogen isotopes like deuterium and tritium are the postulated fuels. The pure form of each isotope can be separated and recovered in a cryogenic distillation column cascade [2]. For fusion applications, the deuterium-tritium streams are the desired products. Hydrogen isotopes have normal boiling points ranging from 20 to 25 K [3], so a cryogenic refrigeration system based on helium as the working fluid is an auxiliary requirement. Thus cryogenic distillation of hydrogen isotopes is a very energy intensive process.

There is a significant body of literature pertaining to the cryogenic distillation of hydrogen isotopes, with a number of papers being available on both theoretical and experimental studies. Among the theoretical investigations, mention can be made of the work of Busigin et al. [4] in which simulation

* *Corresponding author*. Tel.: +91 022 2559 2962.
E-mail address: rupshabhattacharyya1986@gmail.com (R. Bhattacharyya).
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studies on the detailed design of an isotope separation system for ITER application, for two different kinds of tritium breeding blankets have been presented. Special emphasis has been placed on the cryogenic distillation system (columns and equilibriators promoting isotope exchange reaction), which is at the very heart of hydrogen isotope separation. Optimization of the distillation column cascade to minimize the total tritium inventory handled in it has also been carried out in this work. A simplified semi-rigorous model for predicting transient and steady state behaviour of cryogenic distillation columns for hydrogen isotope separation that allows reduction of computational time has also been developed [5]. This improvement has been brought about by using the concept of a supertray, which is actually a block of trays of a column over which the concentration profiles are very flat. Hence this leads to a reduction in the number of equations to be solved for the column. The model has been well validated by experimental studies. A first principle, theoretical study of the transport phenomena involved in cryogenic distillation of hydrogen isotopes has been carried out by some researchers to illustrate the fact that not all the components of the mixture simultaneously reach equilibrium in the vapour and liquid streams leaving a theoretical stage [6]. Specific simulation packages applicable to hydrogen isotope distillation have also been developed [7] where the non-ideality of the hydrogen isotope solutions has been taken into account and the modified algorithm also ensures faster convergence to the solution. Distillation columns with side streams originating in an equilibriator have also been simulated [8] and location of the side stream was seen to strongly influence the column performance. Decay heat of tritium and difference in the molal latent heats of vaporization of the various species were also taken into account in the model used in this study. To account for the radioactivity of tritiated species, a reactive distillation column model was formulated by a different group of researchers [9] and the code was integrated with a commercial process simulator's distillation module to study column performance. Simulations were validated with experiments performed at Laue-Langevin Institute's tritium extraction plant.

Several experimental studies on hydrogen isotope distillation at both laboratory scale and plant scale facilities have been carried out and very important design and operating data have been extracted from them. For example, in a work by Enoeda et al. [10], pressure drop behaviour of the cryogenic distillation column and values of HETP (about 4–6 cm under the conditions used in the study) have been determined experimentally. In another study, a two column arrangement for cryogenic hydrogen isotope distillation has been used to obtain design and operating data for ITER design (e.g. HETP values). A computer code for simulating column performance has been developed. Laser Raman Spectroscopy has been used for determination of the isotopic compositions of the process streams [11]. Test data pertaining to the development and use of two new stainless steel packings for cryogenic hydrogen distillation have been presented recently by Bornea et al. [12]. Experiments on a laboratory scale distillation cryogenic column cascade have been described, parametric studies have been carried out and data for simulation of large scale systems suitable for ITER applications have been collected elsewhere [13]. In another work, full scale columns placed in vacuum jackets where built and their hydraulic behaviour and separation performance with various packings were experimentally studied. Practical problems during column operation were identified [14]. Practical considerations in the design of cryogenic columns for hydrogen isotope separation have been addressed in yet another work [15]. An experimental facility named TRENTA which is a prototype of the ITER WDS and ISS protium separation column, has been operated at TLK and this work presents design and operating data and information about integration of these two processes so that tritium leakage into the environment can be reduced [16]. Since the hydrogen isotope feed stream is derived from a purge gas stream of helium after passing it through a palladium based membrane permeator, the effect of helium on the column's separation performance, control and condensor performance has been studied experimentally. Removal of helium before feeding the hydrogen isotopes to the column has been found to be necessary if the feed has more than 1% helium [17]. A two column cascade has been proposed as an alternative to the TSTA four column cascade for hydrogen isotope separation as would be required for fusion plants like ITER, with reduction in the number of instruments though there is an increase in the tritium inventory being handled here [18].

In this work, an equilibrium-based dynamic model for simulating the behaviour of a packed cryogenic distillation column for hydrogen isotope separation has been presented. The primary aim of the work is to bring together all the necessary data, correlations, and model equations for the distillation system in one place and to simulate the general behaviour of the distillation column, carry out parametric studies and explore possible design alternatives through simulations, without the need for new experimental work. Important hydrodynamic parameters of column behaviour have also been evaluated using physical property data of the isotopic system and packing characteristics of the column, for which equations from literature have been used. The column configuration has been kept conventional and emphasis is placed on the use of the model for prediction of dynamic composition profiles i.e. the separation behaviour in the column operated under various conditions. The model consists of a set of coupled ordinary differential equations representing material balances along with the vapour liquid equilibrium relationships for the hydrogen isotope system, which have been integrated over time using standard techniques. Generally a cascade of three to four columns is usually employed to recover deuterium and tritium in nearly pure form while removing protium [3]. In this work the focus is on the development of a simulation program, thus results for the first column have been presented in maximum detail and the entire cascade configuration has not been analysed here. The model represents the packed column as a tray tower by using known values of the HETP for the particular type of packing used and gas and liquid flow rates prevailing in it. Model equations such as the ones presented here can be integrated using codes written by the user without depending on expensive commercially available process simulation packages, which very often do not have complete pure component and phase equilibrium data for isotopic systems such as the one considered in this study. Moreover the solution of sets of simultaneous algebraic equations needed in steady state