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Advanced Isotope Separation Technology for Fusion Fuel

Xin Xiao*, Henry T. Session, Robert Rabun

Savannah River National Laboratory, Aiken, SC

*E-mail: steve.xiao@srnl.doe.gov

803-646-8360

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D-T fusion is the easiest nuclear fusion reaction among known fusion reactions. Since tritium is extremely rare, it is artificially produced by irradiating lithium metal. The separation, isolation, and storage of the tritium isotope has been a major focus of the Savannah River Site for many decades. Thermal Diffusion, Fractional Absorption, and Cryogenic Distillation have all been used in the past and each has significant operational and safety challenges. A process known as the Thermal Cycling Absorption Process (TCAP) was invented at SRS, and due to its overwhelming advantages in safety, efficiency, size, and reduced tritium inventory, it has replaced all other hydrogen isotope separation processes at SRS. The working principles and current development of hydrogen isotope separation using TCAP at SRS are explained as a potential advanced isotope separation process for the fusion fuel cycle.

Keywords: TCAP; tritium; fusion, isotope separation

I. INTRODUCTION

Nuclear fusion provides enormous energy and could potentially be an invaluable power producing source when it becomes controllable. The fusing of light atomic nuclei – nuclear fusion – is the same reaction that has been powering the Sun and stars since their formation. The concentration of D₂O in water is 155 ppm. Each liter of seawater could produce the energy equivalent of 300 liters of gasoline from D-D fusion. The easiest nuclear fusion reaction is D-T fusion, however, naturally occurring tritium is extremely rare on Earth. For use in sustained controlled nuclear fusion, tritium needs to be produced by breeding and purification processes. In the present design of nuclear fusion reactors tritium has a low burn up rate and therefore a large portion of the D-T fuel will be recycled. One step in the recycling process will be hydrogen isotope separation.

Since hydrogen isotopes have almost identical physical and chemical properties, their separation process is unique and difficult compared to removal of other impurities

such as inert gasses. Thermal diffusion, fractional absorption, and cryogenic distillation were methods used at the Savannah River Site (SRS) for tritium purification [1]. These were all replaced by the thermal cycling absorption process (TCAP) due to its demonstrated unequivocal advantages. TCAP is currently the sole process for purifying tritium at SRS.

II. THERMAL DIFFUSION (TD)

The thermal diffusion process was operated from 1955 to 1986 at SRS. Thermal diffusion consists of a vertical column with a hot wire in the center and an outer cooling jacket. Typically, gas mixtures of 45 to 85% tritium, very small amount of deuterium and with remainder protium, were fed into the middle of the column. The lighter hydrogen isotopes migrated to the hot center and the heavier isotopes moved to the cold outer wall. By gravity, the lighter isotopes at the center moved upward and heavier isotopes near the outer wall moved downward. In steady state, the lighter isotopes are concentrated at the top of the column and heavier isotopes at the bottom. The product, tritium, is withdrawn at the bottom, and raffinate at the top. The thermal diffusion process enriches tritium to better than 95%. Typically, the raffinate will contain approximately 8% tritium. This is sent to a cryogenic still for further tritium recovery. The hot wire at the center of the Thermal Diffusion column catalyzes the hydrogen isotopic exchange. The mixed molecules HD, HT, and DT reach equilibrium with H₂, D₂, or T₂. A Thermal Diffusion column operates at low density and capacity.

II. FRACTIONAL ABSORPTION (FA)

A fractional absorption process was operated at SRS from 1964 to 1968. It was replaced by the cryogenic still. Fractional absorption is a batch chromatography process using palladium powder. To improve the gas flow rate, asbestos was mixed with Pd

powder and this mixture was used to pack a column. Palladium preferentially absorbs lighter isotopes and leaves heavier isotopes in the gas phase. As the gas mixture passed through the column the heavier isotopes eluded first followed by the lighter isotopes. Just prior to the elution of the lighter isotopes the effluent gas is stopped. The heavier isotope, tritium, was collected as it eluded first, and the remaining lighter isotopes in the palladium column were recovered as the column was heated and regenerated. The typical feed was 20% tritium to produce 40% product and 8% raffinate which was subsequently fed to cryogenic still.

III. CRYOGENIC STILL (CS)

The cryogenic distillation process that had a large capacity and was operated at SRS from 1967 to 2004 in two columns. One of the columns consisted of a 20 ft column that was maintained close to liquid hydrogen temperature and a pressure of 900 torr (Figure 1). A reboiler is at the bottom of the column providing hydrogen vapor. A condenser at the top of the column formed liquid hydrogen. Isotopic molecular concentration equalized between counter current phases: down-flow of liquid phase and up-flow of vapor phase. The normal boiling points of the hydrogen isotope species are listed below [2]. The six molecular species are separated by the slight differences in their boiling points and vapor pressures. No atomic isotope exchange occurs at this low temperature. Therefore, gas is removed from the middle of the column, warmed to room temperature, re-equilibrated over a catalyst bed, and returned for separation. In practice, there are more than 6 separatable species. For example, H₂ has two spin isomers (*para*-H₂ and *ortho*-H₂). Equilibrium compositions at ambient temperature are typically about $\frac{3}{4}$ *ortho*-H₂ and $\frac{1}{4}$ *para*-H₂.

Species	H ₂	HD	HT	D ₂	DT	T ₂
Boiling Point, Kelvin	20.4	22.1	23.6	23.7	24.3	25.0

The SRS cryogenic distillation system served two main functions. The first was to separate protium from H-D-T gas mixtures to produce a protium raffinate low enough in tritium that it could be disposed of as waste to the stack. With the protium removed, the second main function was to separate the deuterium-tritium fraction into various concentrations of enriched tritium product depending on the facility's needs at the time. While the actual product concentrations depended on requirements that varied from week to week, the cryogenic still was capable of separating the feed into nearly pure fractions of deuterium and tritium.

The cryogenic distillation system was not automated so it required operator attention 24/7 when the still was at operating temperature and pressure. Power interruptions could cause compressor/refrigerator shutdown and require immediate de-inventory of the still to a relief tank. Because the cryogenic still condenser was oversized, up to 40% of the throughput for a given operating cycle had to be recycled in later operations.

IV. THERMAL CYCLING ABSORPTION PROCESS (TCAP)

TCAP has been in 24/7 production operation at SRS since 1994. TCAP is a continuous gas chromatographic separation process. It consists of a column packed with palladium on kieselguhr (Pd/k), a plug flow reverser (PFR) packed with plain kieselguhr, and a single valve connecting the two columns together, as illustrated in Figure 2. Since the 1930s, palladium was known to have the highest hydrogen isotopic effect [3,4], but a continuous process utilizing this property for hydrogen isotope separation did not appear for almost 50 years until the invention of the TCAP at SRS [5]. Palladium columns have been used by others in fractional absorption processes [6]. However, it was the system design incorporating the PFR that enabled radiological

confinement and a continuous operation, in which the carrier gas, hydrogen, is isotopically separated. The PFR provides a temporary gas storage buffer while maintaining the concentration profile generated by the Pd/k column. The aspects of hydride technology, which provide interim hydrogen storage, generate pressure, and create vacuum have been innovatively applied to create the TCAP process. During operation, the Pd/k column is heated to desorb hydrogen gas and generate pressure, (which is the driving force within a TCAP system), to transfer gas to the PFR, and cooled to absorb hydrogen gas and create a vacuum which pulls gas back into the Pd/k column. By moving gas back and forth in the Pd/k column, hydrogen isotopes are separated at each of the column's separation stages. The Pd/k column's effective number of theoretical stages is enhanced by the fact that the gas can be repeatedly reprocessed in the same column, thus achieving efficient isotope separation. During each cycle, a gas mixture is fed into the Pd/k column and pure product and raffinate are withdrawn from the two ends of the column. The TCAP invention is an engineering achievement with simple design and advanced control logic. A simple on/off valve is the only mechanical moving part. It is a robust design with inherent safety features. Should power or cooling be lost, the tritium gas is safely contained within the TCAP columns until the process resumes. Since the only mechanically moving part in the system is a valve, system maintenance is minimal. It can be scaled from very small to large with plenty of versatility depending on throughput.

Since the 1st generation TCAP systems are approaching design life, considerable effort has been spent at SRS developing, testing, and evaluating new innovative concepts to advance the technology, reduce the footprint to conserve glovebox space, and increase efficiency [7,8]. As a result of the successful

demonstration of these innovations, external customers have approached SRNL to acquire TCAP units for tritium purification [9].

V. TCAP APPLICATION FOR FUSION FUEL CYCLE

The new TCAP development further improved the heat transfer design, increased throughput, and reduced footprint (Figure 3). The process was intensified with direct electric heaters at full duty cycle for column heating, and liquid nitrogen (LN_2) as a quench type of column cooling. PFR was replaced by an inverse column that prefers heavier hydrogen isotope, so that hydrogen isotopes are separated twice during each cycle. As a result, the TCAP throughput doubled or the inventory reduced by half for the same throughput [10]. Testing has demonstrated that TCAP can easily separate the naturally occurring 150ppm D_2 , thus producing pure protium. A compact design column further reduced the footprint and increased the heat transfer efficiency [11]. With the overall improvement, the current TCAP design can reduce tritium inventory by almost two orders of magnitude over the cryogenic still process. Because of the small TCAP footprint it can be placed inside a glovebox. Operational data showed a reduction of tritium emission by an order of magnitude after the cryogenic still was replaced by the 1st generation TCAP. With the current TCAP development, tritium emission is expected to reduce further by 1 - 2 orders of magnitude. During system upsets or power outage, the hydrogen isotopes remain absorbed on the palladium column and in a safe state. When operation is resumed the TCAP recovers the separation profile within one cycle.

Shine Medical Technologies licensed TCAP technology in support of linear accelerator D-T fusion for Mo-99 production. The D-T fusion is used to generate neutrons. Mo-99 and other short half-life medical isotopes are produced in the uranium-bearing targets by irradiating them with thermal neutrons. After delivery of the 1st H-D

demonstration TCAP, Shine has selected and ordered additional TCAPs for D-T separation to support medical isotope production.

The Laboratory for Laser Energetics (LLE) at the University of Rochester acquired a TCAP from the Savannah River National Laboratory (SRNL) to support their fusion energy research. LLE researchers study inertial confinement fusion (IFC) of D-T fuel using powerful lasers. The SRNL TCAP system removed all protium from the LLE tritium supply. Test shots with the purified tritium increased neutron yield significantly.

The current TCAP prototype is designed for weapons tritium purification. It would require major scale up efforts for fusion energy application such as ITER. Concepts have been developed focusing on intensified heat transfer and mass transfer, multiple columns, and advanced control algorithm to adapt to a throughput for fusion energy up to 100 std liters/min. The new design TCAP with inverse column can reduce the raffinate tritium concentration by a factor of 100. Since TCAP is small enough to be placed inside a glovebox (as oppose to cryogenic distillation), tritium emission is expected to reduce by another factor of 10 based on SRS operation experience. TCAP is capable of providing much higher ratio of throughput to hydrogen isotope inventory than that of the cryogenic distillation. Besides overall smaller size of TCAP, the new TCAP design doesn't require refrigeration system which is part of the footprint in the cryogenic distillation system. If used in fusion energy, TCAP technology could reduce tritium inventory by a factor of about 90, reduce tritium emission by a factor of 1000, reduce footprint to 1/15th, and provide an inherent safe robust process compared to cryogenic distillation.

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Figure 1 Cryogenic distillation process operated at SRS 1967-2004 (photo taken for upstairs portion and downstairs portion of the same column).

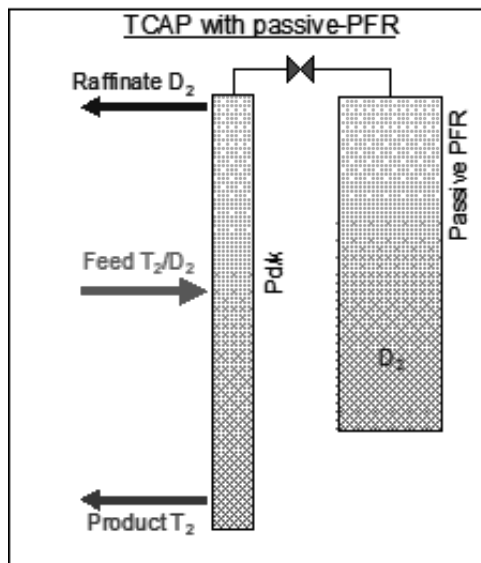


Figure 2 Illustration of TCAP Working Principle with a Pd/k Column and Plug Flow Reverser (PFR).

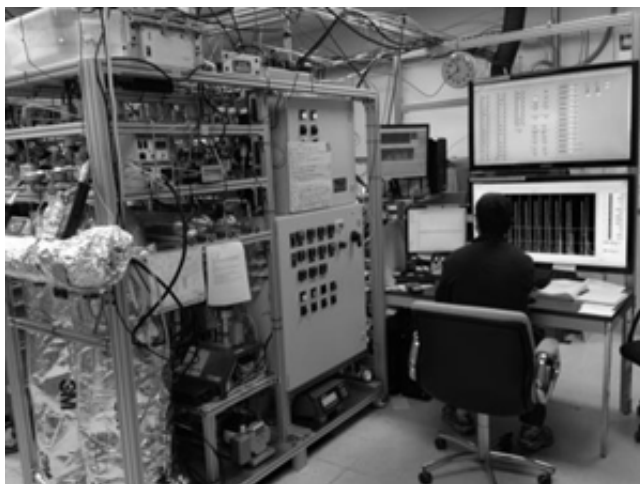


Figure 3 State of the art TCAP prototype