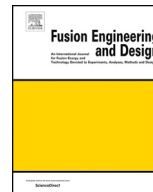




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A distillation column for hydrogen isotope removal from liquid lithium

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ABSTRACT

Recovery of tritium from plasma-facing components in fusion devices will be vital to future full-scale operation. Liquid, low-Z materials have demonstrated many inherent advantages over solid first wall materials. To this end, a thermal treatment method in the form of a distillation column for extraction of hydrogen isotopes from liquid lithium has been designed, developed, and constructed at the Center for Plasma-Material Interactions at the University of Illinois at Urbana-Champaign. Use of induction heating and lithium condensation stages are the two qualities that set this design apart from other thermal treatment systems. Induction heating capabilities were modeled using the COMSOL Multiphysics software, which were validated when commissioning the physical heater module. Proof-of-concept tests were performed in the prototype column, which were undertaken as batch processes to investigate the efficacy with which the column could remove hydrogen gas from lithium-rich and lithium hydride-rich samples. All of the tests reported used lithium hydride as a surrogate for lithium deuteride and lithium tritide. The design process and results from the initial tests will be discussed, along with the envisioned placement of this treatment scheme in a fully-functional lithium loop.

1. Introduction

As lithium has gained popularity in the fusion community as an alternative wall material due to its ability to enhance confinement [1] and consume impurity and cold fuel particles [2], it has become clear that the biggest roadblock to the universal application of lithium (Li) as a first wall material is its ability to retain tritium (T). These concerns exist because on-site tritium inventory is limited due to availability and radiation safety [3].

Due to these concerns, various methods [4–7] have been proposed with the sole purpose of separating tritium from lithium and lithium alloys. Catalytic separation using yttrium has been studied in the context of the Internal Fusion Materials Irradiation Facility (IFMIF) lithium loop [4], but appears to suffer from low tritium yields. The combination of molten salt extraction and electrolysis [6] has demonstrated the ability to evolve tritium at rates that approach reactor-relevant operation; however, yields from this process are still too low, and the technology itself suffers from the presence of impurities and the production of unwanted by-products. The most promising techniques for recovering tritium from lithium are those that take advantage of the thermophysical properties of the lithium-lithium hydride (lithium deuteride, lithium tritide) phases, such as those proposed in Ref. [7]. What is noticeably absent from these systems is a way to actually evolve tritium and deuterium gas from lithium solutions. This absence was the

primary motivator for the work accomplished at the University of Illinois.

To fill in this missing step, specifically with regards to the loop system proposed in Ono's work [7], a distillation column for hydrogen isotope evolution from lithium was designed, developed, constructed, and tested at the Center for Plasma-Material Interactions [8]. Two components of this design set it apart from other conventional distillation columns. First, induction heating was used as the primary driver for evolution, since induction drive heats metals more rapidly. The induction heating capabilities were modeled in the COMSOL Multiphysics simulation software [9]. The accuracy of the simulation was verified with a commissioning test, where the working coil was used to heat the actual column structure. Second, condensation stages were built into the column to capture lithium vapor. These stages, and an evaluation of how much lithium was able to travel up and through the column, were based on the work done by Goldston [10] on the Lithium Vapor Box Divertor.

Proof-of-concept tests were then performed in the fully-constructed, prototype distillation column. These tests were conducted with samples having various hydrogen molar ratios in mixtures that contained both lithium (Li) and lithium hydride (LiH). The effectiveness of the column at removing hydrogen was then gauged by monitoring the partial pressures as the “bucket” of the column was heated using the induction heater. The results from these tests will be described later; however, one

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of the more important conclusions that was drawn was that the hydrogen concentration in the lithium sample was pivotal to the resultant evolution rate.

This paper presents the design, development, and operation of the prototype distillation column constructed at the Center for Plasma-Material Interactions (CPMI) to recover hydrogen gas from hydrogenated lithium samples. The thermodynamics and kinetics which govern hydrogen release in this system have also been thoroughly explored at CPMI at smaller scales. Section 2 describes the theory and design criteria used for the column development. This includes the simulation of the induction heating capabilities of the actual source. Section 3 presents the results from the commissioning and Proof-of-concept tests, wherein a lithium-rich and a lithium hydride-rich sample were tested. Section 4 discusses the impact of the results from this prototype technology, and what these results mean within the context of larger liquid lithium loop systems. Section 5 summarizes the results and describes future experimental work.

2. Theory and design

The prototype distillation column was designed to extract hydrogen from batch lithium melts and collect the evaporated lithium. Two novelties of design seek to accomplish these goals: inclined condensation stages and induction heating. Induction heating is the driving force behind hydrogen desorption and was modeled using the COMSOL® Multiphysics software. Clean lithium collection and potential diversion occur at the condensation stages. Lithium flow is analyzed through a mass and energy balance. Design considerations allow for safe operation, efficiency, and future extensions.

2.1. Column

Induction heating drives thermal desorption of hydrogen in the lithium melt. As temperatures increase, hydrogen and lithium vapor flow upward and encounter the first condensation stage (Fig. 1a), which is kept at 315 °C to allow for lithium condensation but limit hydrogen co-deposition [11]. Hydrogen is then directed toward a small outlet into the second condensation section, creating nozzled flow. The second condensation stage works to condense more of the lithium vapor and funnel the hydrogen towards the top of the chamber. The hydrogen reaches a small aperture (1.14 mm) that restricts the flow of hydrogen into the region of the chamber containing the residual gas analyzer (RGA). As the hydrogen exits the top of the column, it could be directed towards gas separation technologies and eventually back into the reactor.

The condensation stages are the key feature for lithium collection. The upward angle of 45° and temperatures above lithium's melting temperature allow for flow of lithium back down toward the bucket. In the proposed loop system, the flow of lithium off of the stages could be used to recycle clean lithium back to the in-vessel, plasma-material interface. Each stage is connected to the rest of the column by two set screws, making the stages completely modular. Stages could be removed or added to meet the specifications of the system. Stages can also be manufactured readily, such that an array of distillation columns would be easily attainable. Buildup of hydrogen inside the chamber could be a concern. However, the gas relief lip (Fig. 1b) allows for flow of gas to the portion of the chamber outside the column walls, where it can be pumped out, preventing this buildup. The bucket (Fig. 1b) is grooved to prevent warping during the rapid rise in temperature during operation. The bucket was welded into an 203 mm ConFlat flange to allow easy attachment to the vacuum chamber.

2.2. COMSOL heating

The presence of solid hydrides in lithium melts requires temperatures near 690 °C, the melting temperature of lithium hydride, to

promote thermal desorption of hydrogen. Near these temperatures, the solid hydride precipitate, known as the β phase, dissolves into solution into what is known as the α phase. This type of chemistry has been observed in lithium-lithium hydride systems [8,12,13], and is the fundamental principal which governs hydrogen isotope recovery from thermal treatment systems. This is of particular importance when no hydrogen originally exists in the mixture in the α phase. As such, treatment temperatures near the melting point of lithium hydride are necessary for appreciable recovery.

Induction heating's efficiency at heating metals gives it a distinct advantage over the use of resistive heating. COMSOL Multiphysics® was used to model the induction heating of the column's lower bucket [9]. The induction heating module is based on the coupling of heat transfer in solids, Lenz's Law, and Ampere's Law. Induction heating uses a coil carrying high frequency current to create alternating magnetic fields in the workpiece. These alternating fields create eddy currents, which heat the workpiece through Ohmic dissipation. Changes in resistivity throughout heating require the physics to be coupled, such that these changes can inform the rest of the physics.

The physics is based on three fundamental equations, the thermal energy equation for heat transfer in solids and a combination of Lenz's Law and Ampere's Law [9]:

$$\rho C_p \frac{\partial T}{\partial t} + \rho C_p \mathbf{u} \cdot \nabla T = \nabla \cdot (k \nabla T) + Q, \quad (1)$$

$$(i\omega\sigma - \omega^2\epsilon_0\epsilon_r)\mathbf{A} + \nabla \times \frac{\mathbf{B}}{\mu_0\mu_r} - \sigma\mathbf{v} \times \mathbf{B} = \mathbf{J}_e, \quad (2)$$

$$\mathbf{B} = \nabla \times \mathbf{A}, \quad (3)$$

where ρ is the material density, C_p is the constant-pressure heat capacity of the material, T is the temperature of the material in K , \mathbf{u} is the advection term for the thermal energy equation, k is the heat conductivity of the material, Q is the heat source, i is the imaginary root, ω is the angular frequency at which the induction coil is driven, $\epsilon_0\epsilon_r$ is the product of the permittivity of free space and that of the specific material, \mathbf{A} is the magnetic vector potential defined by Eq. (3), \mathbf{B} is the magnetic field, $\mu_0\mu_r$ is the product of the permeability in vacuum and in the specific material, σ is the electrical conductivity of the material, \mathbf{v} is the velocity vector, and \mathbf{J}_e is the current driven in the material. The coupling of the physics is in the heat source term [9]:

$$Q = \frac{1}{2} \text{Re}(\mathbf{J} \cdot \mathbf{E}^*) + \frac{1}{2} \text{Re}(i\omega\mathbf{B} \cdot \mathbf{H}^*), \quad (4)$$

where \mathbf{J} is the total current, \mathbf{E}^* is the complex conjugate of the electric field, \mathbf{B} is the magnetic field, and \mathbf{H}^* is the complex conjugate of the auxiliary field.

The COMSOL model was constructed as a 2-dimensional axisymmetric system, where the physical coil was mimicked by five coils with a pitch of 1.5 cm, an inner diameter of 6 mm, and an outer diameter 8 mm. However, this coil model does not exactly match the physical continuous coil with the aforementioned pitch and diameters. The simulation was run with currents and frequencies that are feasible for the power supply to generate. Fig. 2 shows the temperatures achieved by the COMSOL model after one hour of heating. Based on the model, temperatures needed to promote dissolution of the hydrogen into the α phase and, therefore, hydrogen extraction are attainable.

2.3. Stages and mass and energy balances

As was mentioned in the Introduction, the way in which the stages are meant to condense and capture lithium is based on the work done by Goldston [10,14] on the Lithium Vapor Divertor Box. The stages are held in excess of 320 °C in order to ensure that the lithium that is captured remains liquid. Eventually, outlets at these stages will be integrated into the design to divert clean lithium flow into a separate reservoir, which will be used as a holding tank for lithium that will be

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