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(54) **SYSTEM FOR EXTRACTION OF TRITIUM FROM LIQUID METAL COOLANTS**

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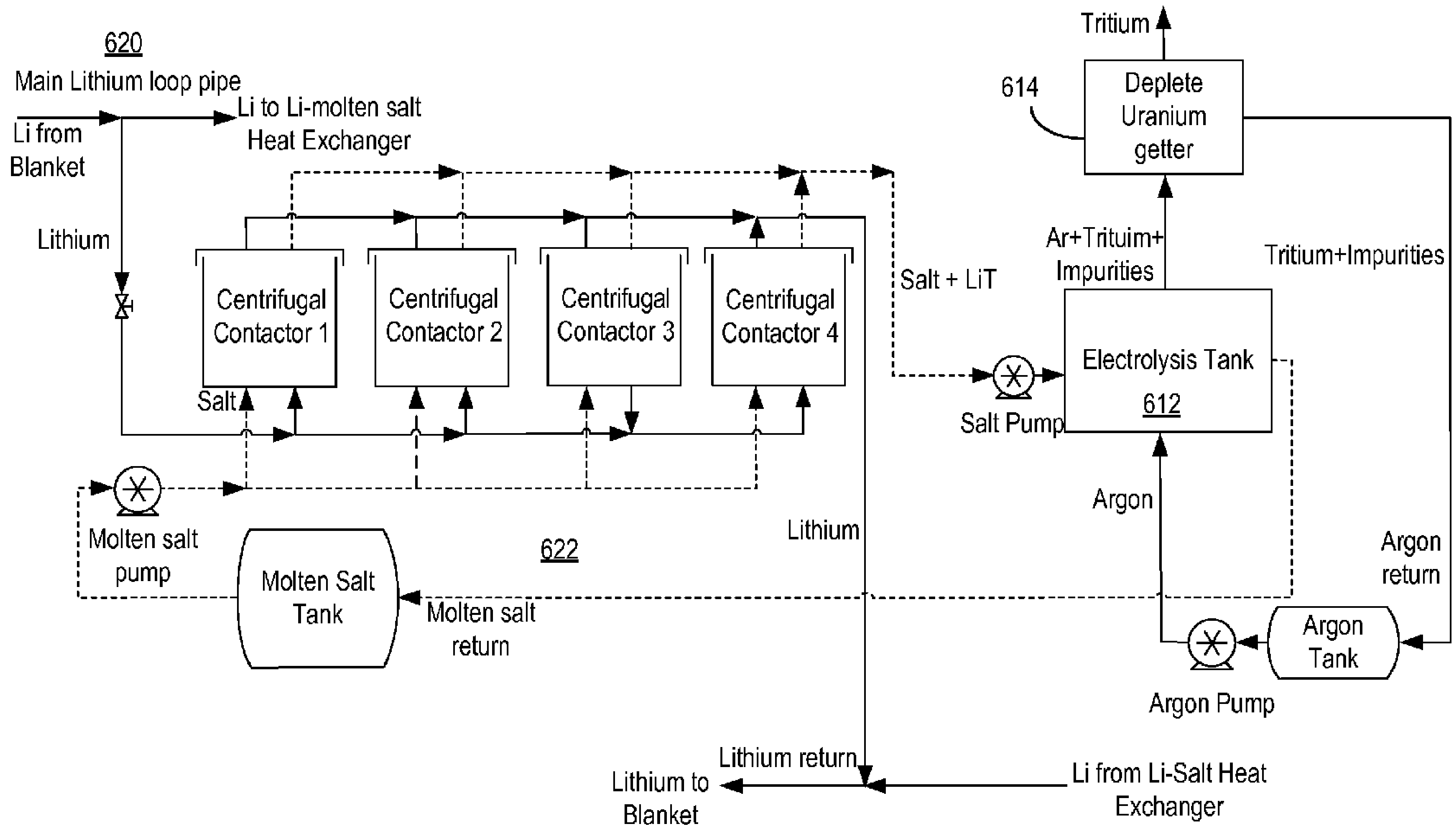
CPC *C25C 3/34* (2013.01)
USPC **205/705**; 204/243.1

(57) **ABSTRACT**

A method for removing tritium from liquid lithium includes mixing the liquid lithium containing trace amounts of tritium with a molten salt and forming a salt of lithium and tritium. The method also includes separating the liquid lithium from the salt of lithium and tritium and circulating the molten salt in an electrolyzer to form molecular tritium. The method further includes bubbling an inert gas through the electrolyzer to remove the molecular tritium and circulating the argon and removed molecular tritium in a titanium getter to recover the tritium.



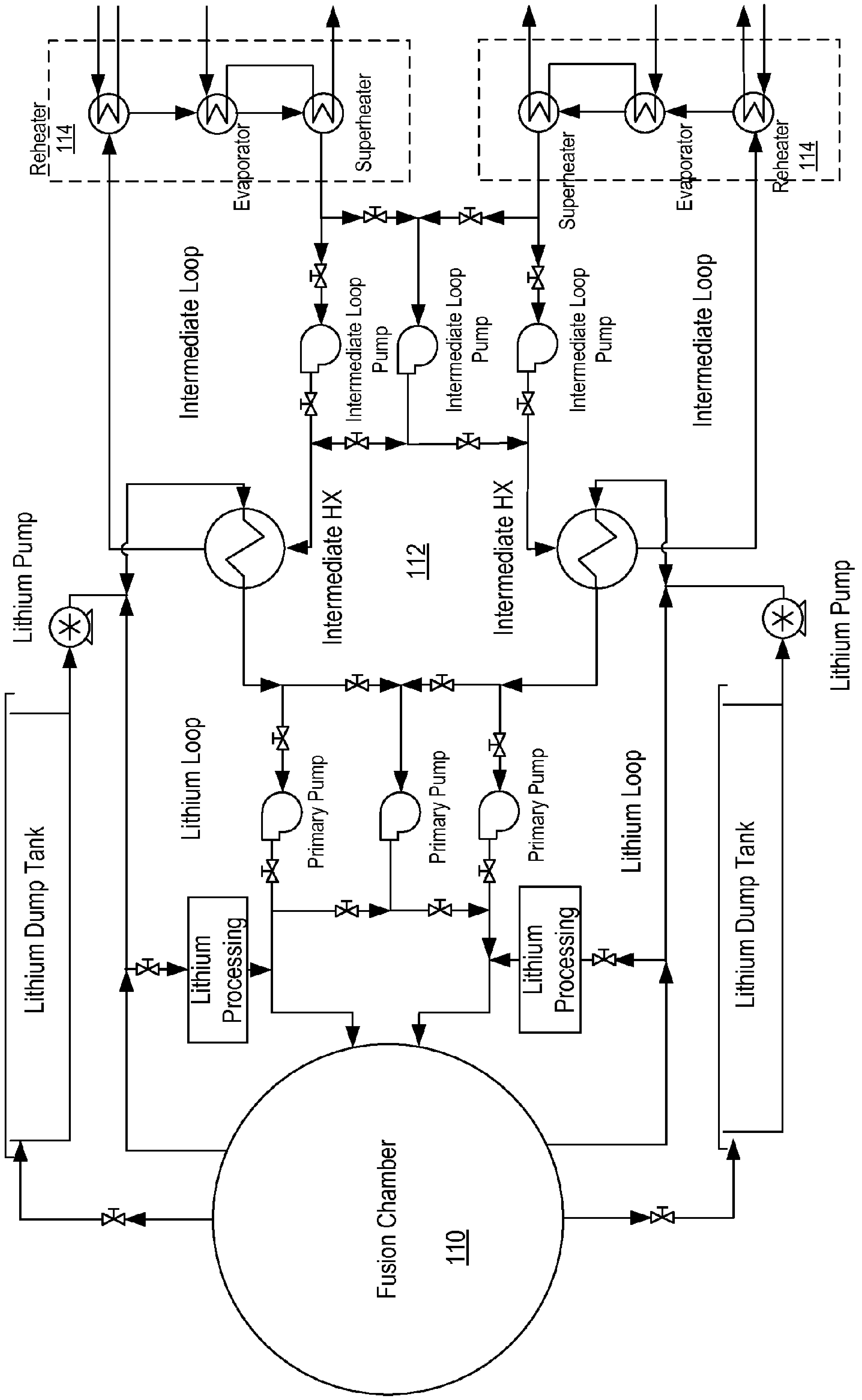

Fusion Chamber
110
Lithium Dump Tank
Lithium Dump Tank
Lithium Pump
Lithium Pump
Lithium Processing
Lithium Processing
Lithium Loop
Lithium Loop
Primary Pump
Primary Pump
Primary Pump
Intermediate HX
Intermediate HX
112
Intermediate Loop Pump
Intermediate Loop Pump
Intermediate Loop Pump
Intermediate Loop
Intermediate Loop
Reheater
114
Evaporator
Superheater
Superheater
Evaporator
Reheater
114

FIG. 1

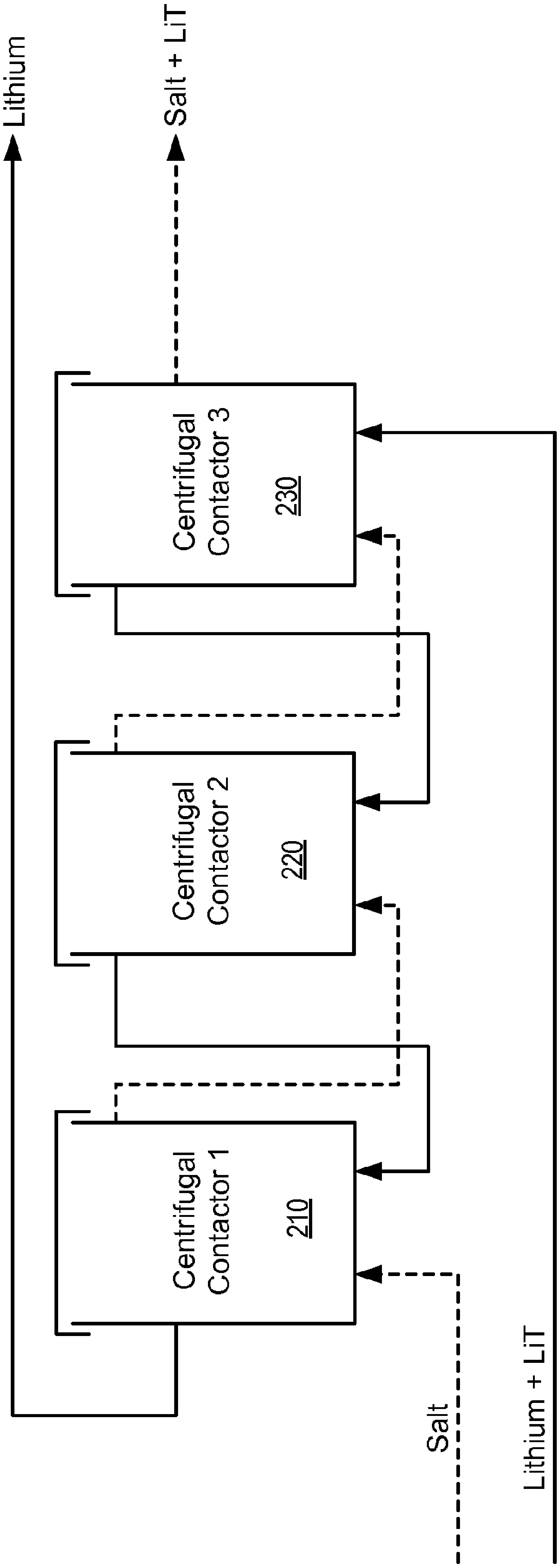

Lithium
Salt + LiT
Centrifugal Contactor 3
230
Centrifugal Contactor 2
220
Centrifugal Contactor 1
210
Salt
Lithium + LiT

FIG. 2

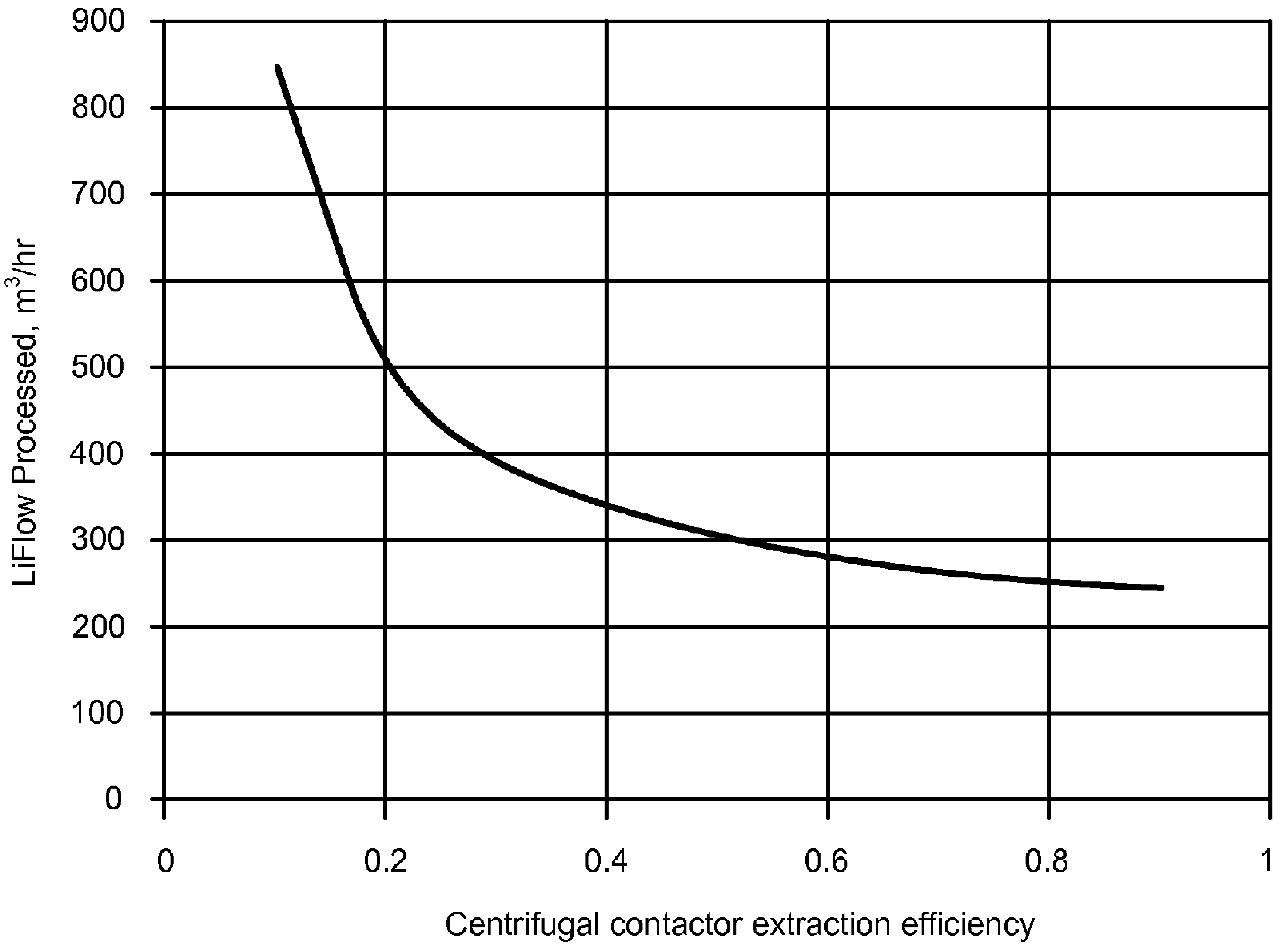

900
800
700
600
500
400
300
200
100
0
LiFlow Processed, m³/hr
0
0.2
0.4
0.6
0.8
1
Centrifugal contactor extraction efficiency

FIG. 3

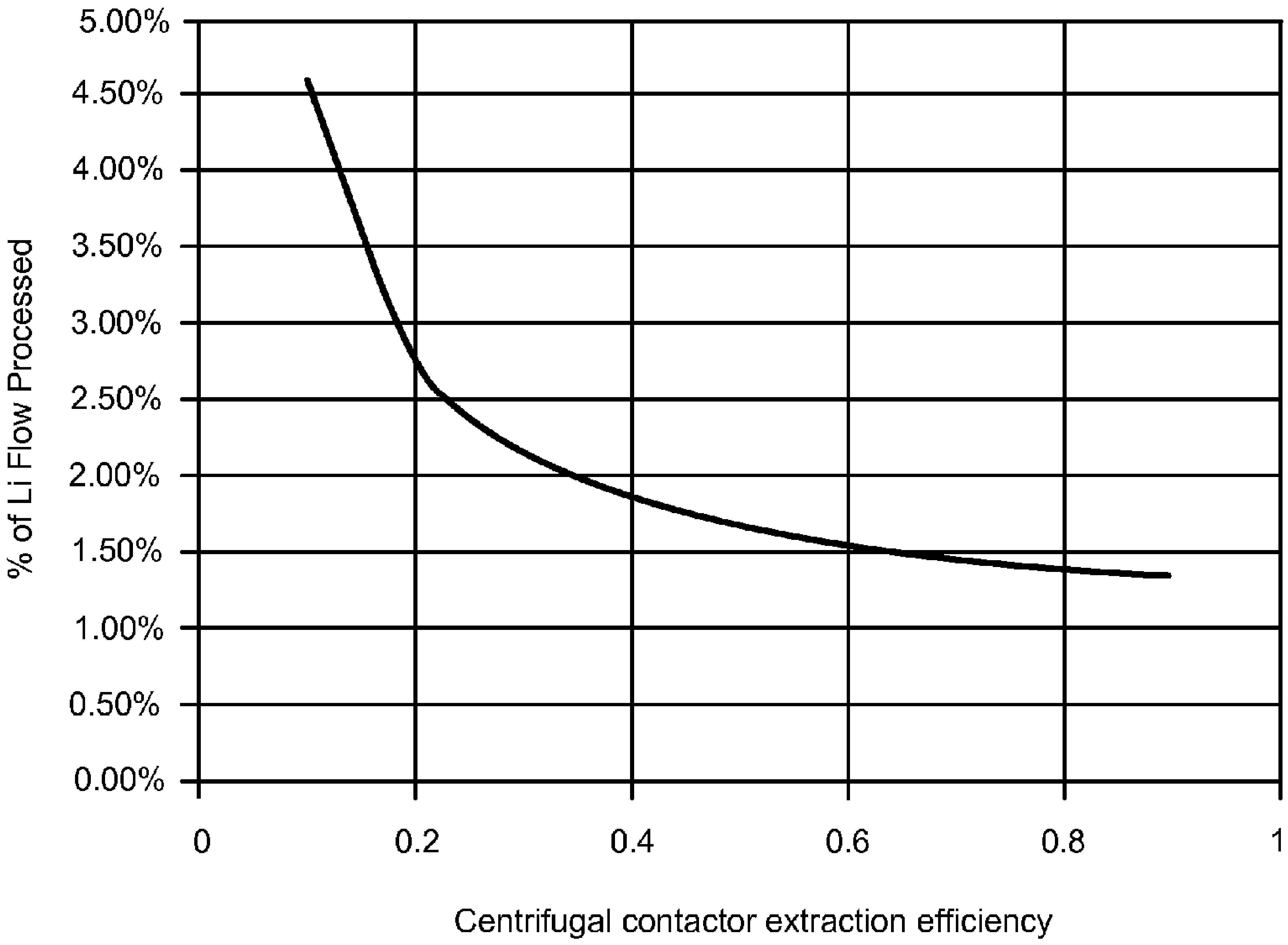

5.00%
4.50%
4.00%
3.50%
3.00%
2.50%
2.00%
1.50%
1.00%
0.50%
0.00%
% of Li Flow Processed
0
0.2
0.4
0.6
0.8
1
Centrifugal contactor extraction efficiency

FIG. 4

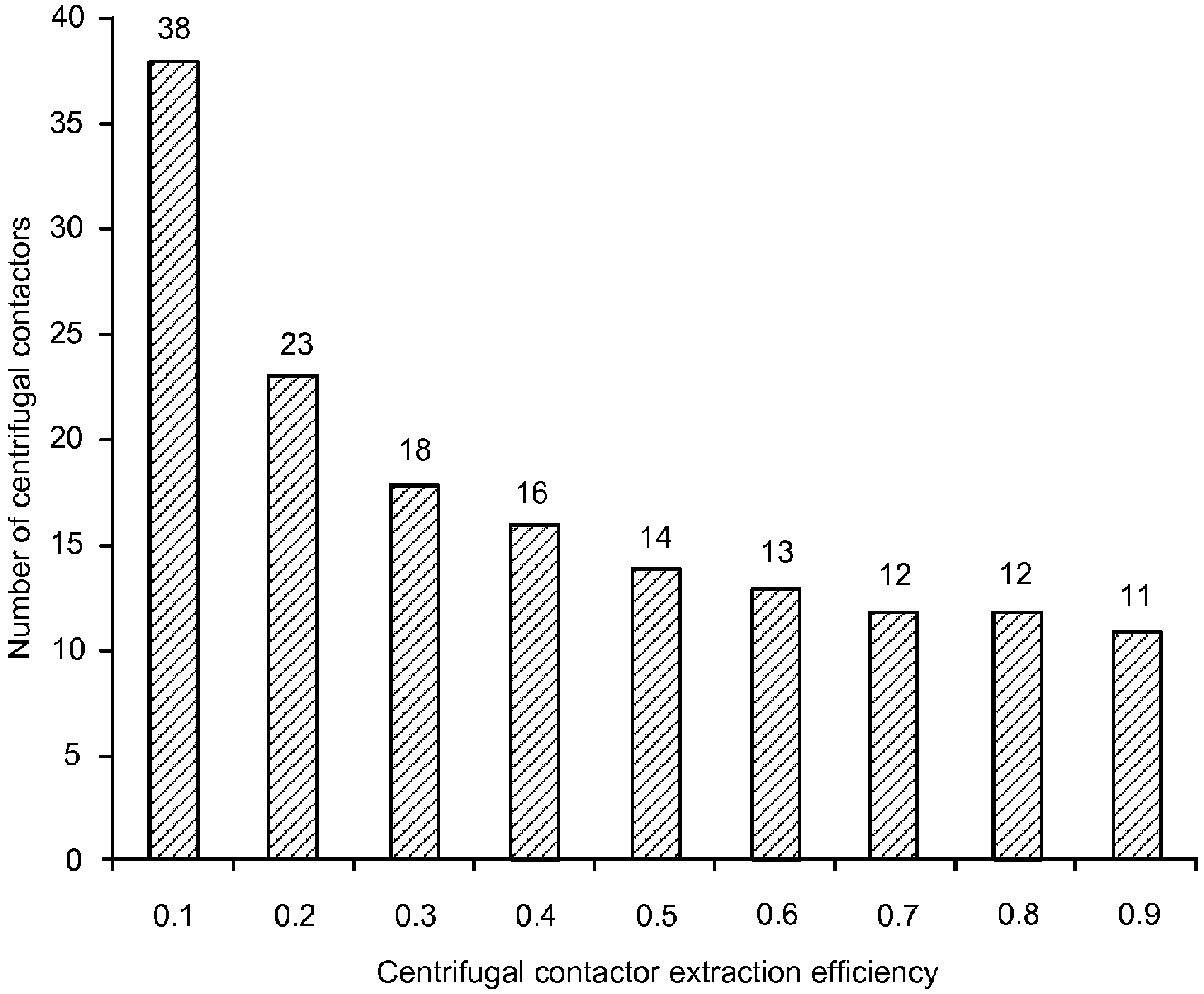

Number of centrifugal contactors
40
35
30
25
20
15
10
5
0
38
23
18
16
14
13
12
12
11
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
Centrifugal contactor extraction efficiency

FIG. 5

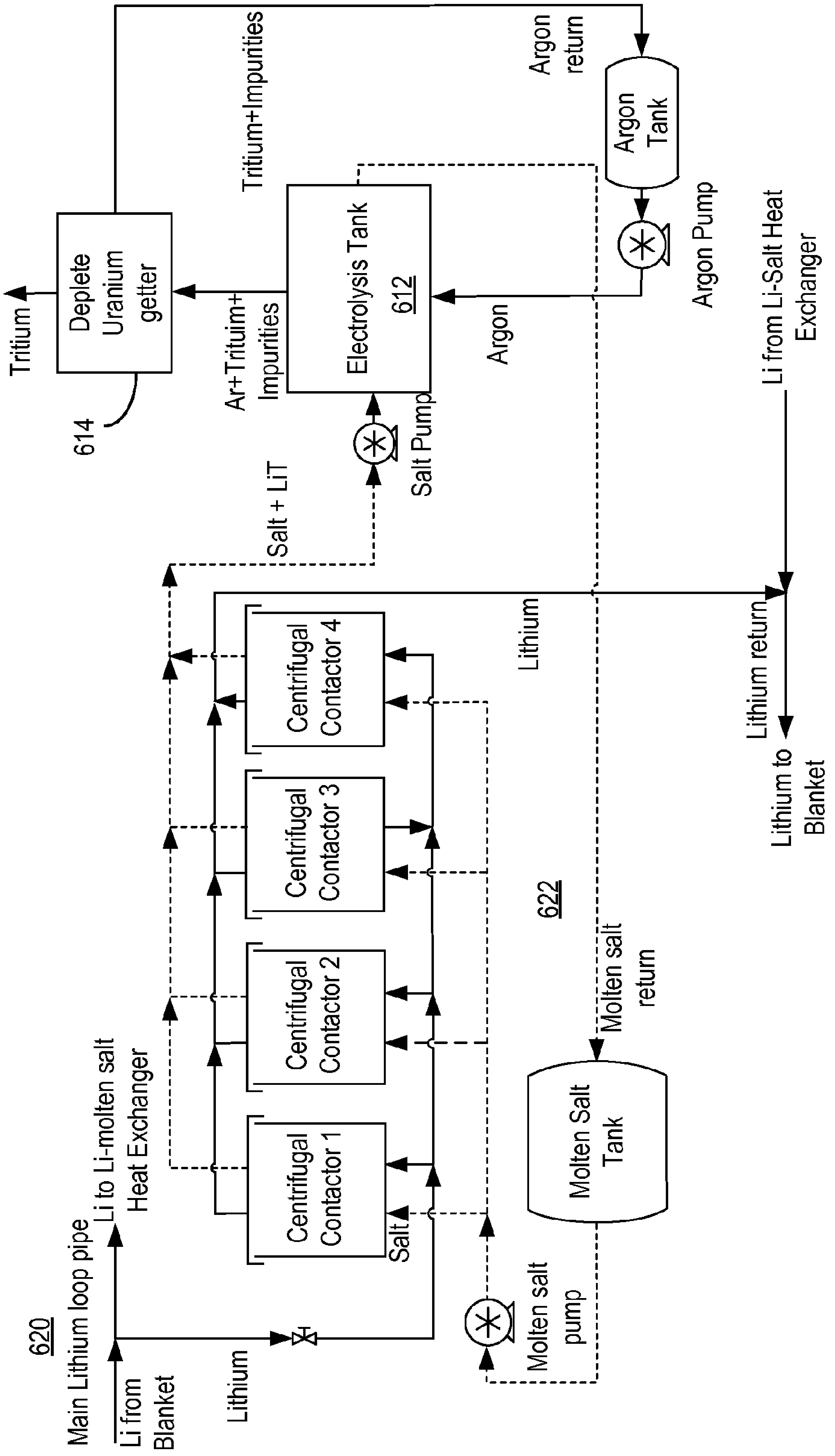

Tritium
Deplete Uranium getter
614
Tritium+Impurities
Argon return
Argon Tank
Ar+Trituim+ Impurities
Electrolysis Tank
612
Argon
Argon Pump
Salt Pump
Salt + LiT
Li from Li-Salt Heat Exchanger
Lithium
Lithium return
Lithium to Blanket
Centrifugal Contactor 4
Centrifugal Contactor 3
Centrifugal Contactor 2
Centrifugal Contactor 1
622
Molten salt return
Molten Salt Tank
Li to Li-molten salt Heat Exchanger
Salt
620
Main Lithium loop pipe
Li from Blanket
Lithium
Molten salt pump

FIG. 6

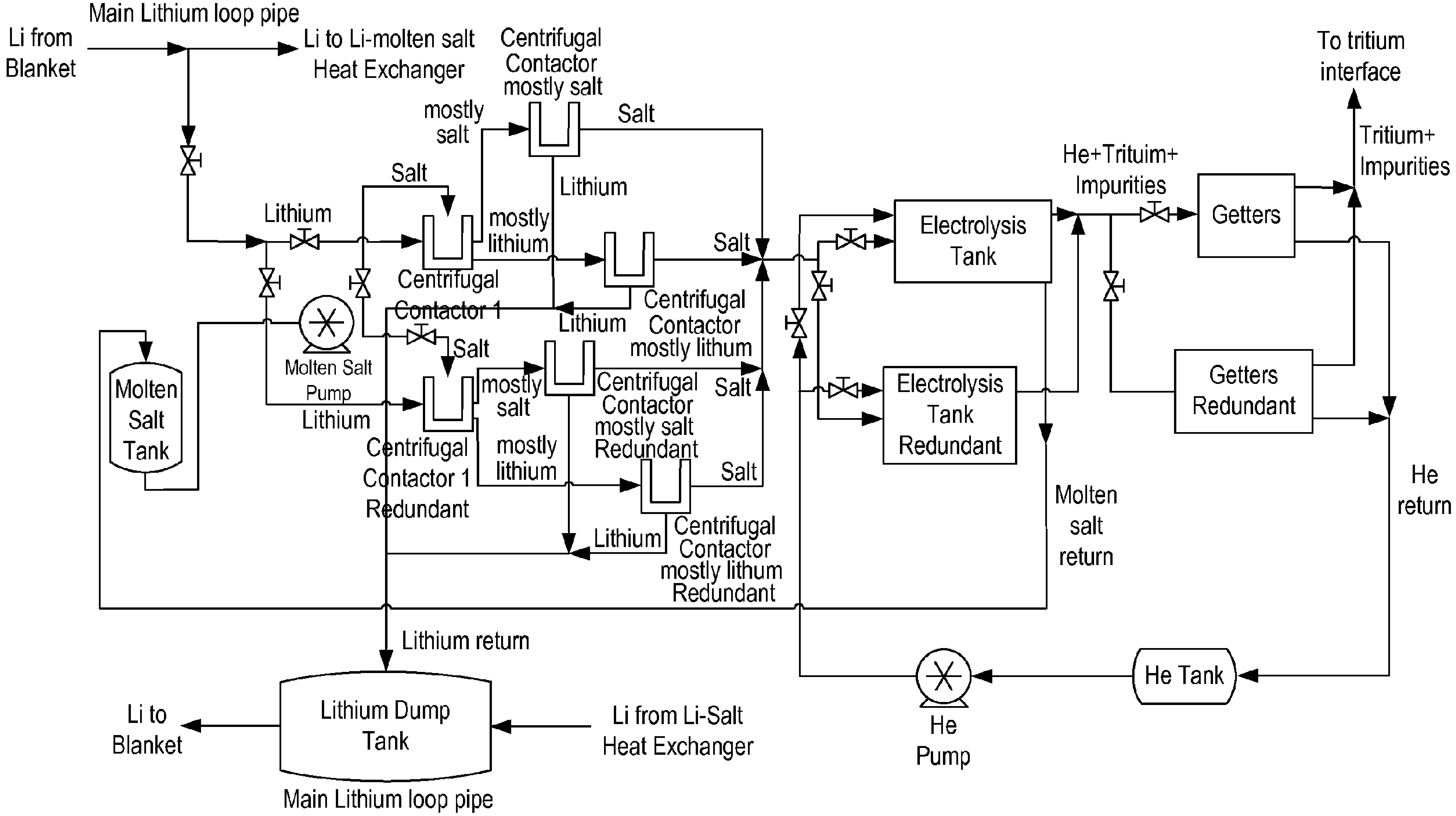

Li from Blanket
Main Lithium loop pipe
Li to Li-molten salt Heat Exchanger
Centrifugal Contactor mostly salt
mostly salt
Salt
Salt
Lithium
Lithium
mostly lithium
Salt
Centrifugal Contactor 1
Centrifugal Contactor mostly lithum
Lithium
Salt
Molten Salt Tank
Molten Salt Pump
Lithium
mostly salt
Centrifugal Contactor mostly salt Redundant
Salt
Centrifugal Contactor 1 Redundant
mostly lithium
Salt
Lithium
Centrifugal Contactor mostly lithum Redundant
Lithium return
Lithium Dump Tank
Li to Blanket
Li from Li-Salt Heat Exchanger
Main Lithium loop pipe
Electrolysis Tank
Electrolysis Tank Redundant
He+Trituim+ Impurities
Getters
Getters Redundant
To tritium interface
Tritium+ Impurities
Molten salt return
He return
He Tank
He Pump

FIG. 7

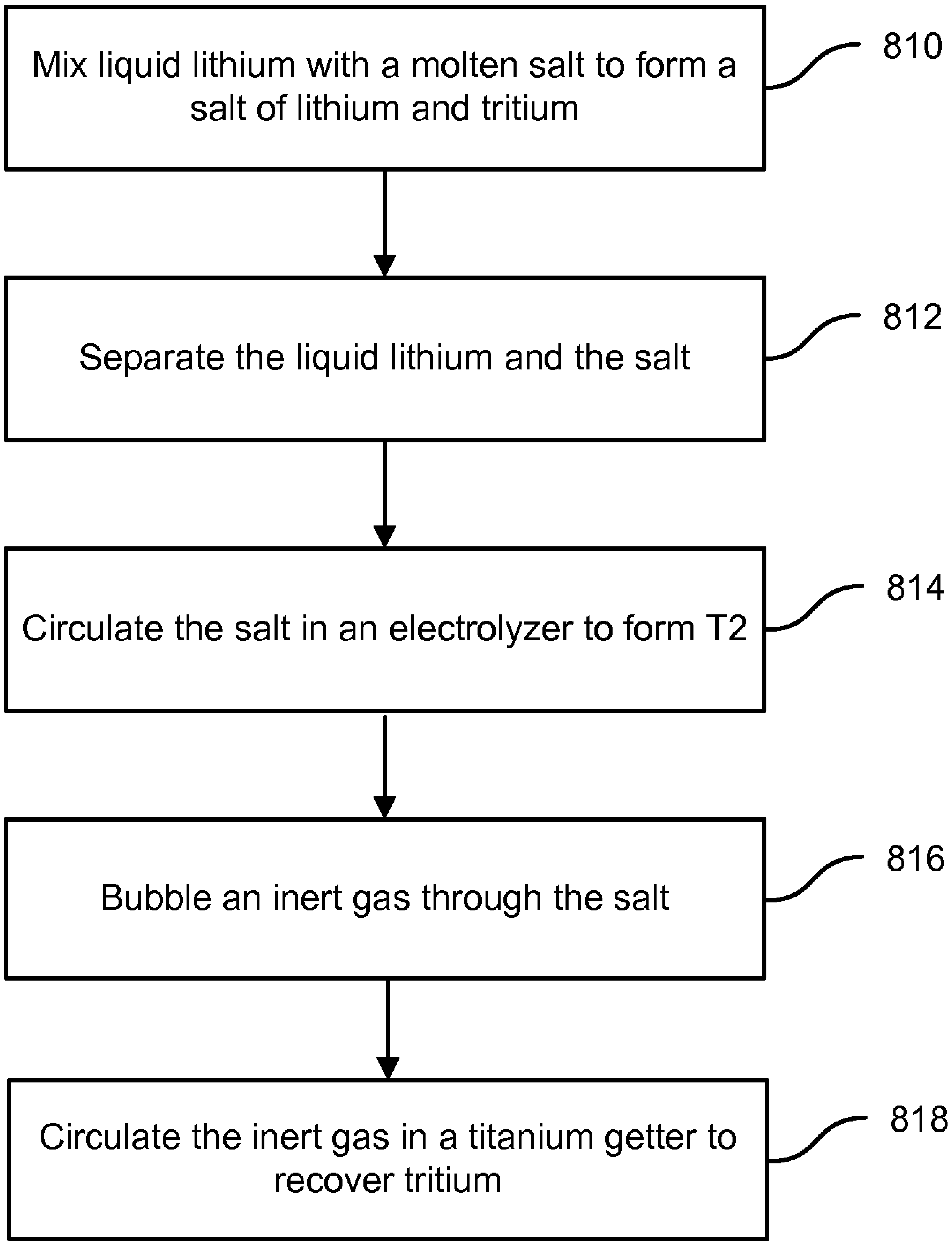

Mix liquid lithium with a molten salt to form a salt of lithium and tritium
810
Separate the liquid lithium and the salt
812
Circulate the salt in an electrolyzer to form T2
814
Bubble an inert gas through the salt
816
Circulate the inert gas in a titanium getter to recover tritium
818

FIG. 8

SYSTEM FOR EXTRACTION OF TRITIUM FROM LIQUID METAL COOLANTS

CROSS-REFERENCES TO RELATED APPLICATIONS

[0001] This application claims priority to U.S. Provisional Patent Application No. 61/722,569, filed Nov. 5, 2012, and entitled: "System for Extraction of Tritium from Liquid Metal Coolants," the disclosure of which is hereby incorporated by reference in its entirety for all purposes.

STATEMENT AS TO RIGHTS TO INVENTIONS MADE UNDER FEDERALLY SPONSORED RESEARCH OR DEVELOPMENT

[0002] The United States Government has rights in this invention pursuant to Contract No. DE-AC52-07NA27344 between the United States Department of Energy and Lawrence Livermore National Security, LLC for the operation of Lawrence Livermore National Laboratory.

BACKGROUND OF THE INVENTION

[0003] The National Ignition Facility (NIF), the world's largest and most energetic laser system, is operational at Lawrence Livermore National Laboratory (LLNL) in Livermore, Calif. One goal of operation of the NIF is to demonstrate fusion ignition for the first time in the laboratory. Initial experiments are calculated to produce yields of the order of 20 MJ from an ignited, self-propagating fusion burn wave. The capability of the facility is such that yields of up to 150-200 MJ could ultimately be obtained. The NIF is designed as a research instrument, one in which single "shots" on deuterium-tritium containing targets are performed for research. A description of the NIF can be found in Moses et al, *Fusion Science and Technology,* volume 60, pp 11-16 (2011) and references therein.

[0004] There is a rapidly growing need for power, and especially for clean power. At LLNL a project known as Laser Inertial-confinement Fusion Energy, (often referred to herein as "LIFE") is working toward introduction of fusion based electric power plants into the U.S. economy before 2030, and in a pre-commercial plant format before that. LIFE technology offers a pathway for the expansion of carbon-free power around the world. It will provide clean carbon-free energy in a safe and sustainable manner without risk of nuclear proliferation.

[0005] One challenge with respect to LIFE, as well as any technology for generating electrical power to be distributed to large numbers of consumers, is reliability. Consumers expect to have extraordinarily high reliability in their electric power supply. The result is that utilities that provide that electrical power maintain their facilities to assure the required high reliability. Thus, among the challenges with respect to fusion power, is to provide mechanisms by which a reliable long-lived fusion chamber can be provided in which the fusion reactions occur, yet which can be maintained or replaced when necessary with minimal downtime for the overall power plant.

[0006] Despite the progress made in the design of fusion based electric power plants, there is a need in the art for improved methods of extracting tritium from liquid coolants.

SUMMARY OF THE INVENTION

[0007] The present invention relates generally to methods and systems for extracting tritium from lithium-based coolants. More particularly, embodiments of the present invention provide methods and systems for extracting tritium from lithium liquids that includes trace amounts of lithium. Although embodiments of the present invention are discussed in terms of lithium-based coolants, other liquid metal coolants are included within the scope of the present invention.

[0008] According to an embodiment of the present invention, a method for removing tritium from liquid lithium is provided. The method includes mixing the liquid lithium containing trace amounts of tritium with a molten salt and forming a salt of lithium and tritium. The method also includes separating the liquid lithium from the salt of lithium and tritium and circulating the molten salt in an electrolyzer to form molecular tritium. The method further includes bubbling an inert gas through the electrolyzer to remove the molecular tritium and circulating the argon and removed molecular tritium in a titanium getter to recover the tritium.

[0009] According to another embodiment of the present invention, a system for recovering tritium from a lithium coolant is provided. The system includes a lithium coolant circuit. The lithium coolant in the circuit includes trace amounts of tritium. The system also includes a plurality of contactors coupled to the lithium coolant circuit. The plurality of contactors are operable to mix the lithium coolant with a molten salt and form a salt of lithium and tritium. The system further includes an electrolysis unit coupled to one or more of the plurality of contactors, an inert gas source operable to bubble an inert gas through the electrolysis unit, and a getter coupled to the electrolysis unit and operable to recover the tritium.

[0010] Numerous benefits are achieved by way of the present invention over conventional techniques. For example, embodiments of the present invention provide methods and systems for extracting tritium from lithium coolant fluids that are more efficient than conventional techniques. These and other embodiments of the invention along with many of its advantages and features are described in more detail in conjunction with the text below and attached figures.

BRIEF DESCRIPTION OF THE DRAWINGS

[0011] FIG. **1** is a simplified schematic diagram illustrating a 1.2 GW (thermal) power plant based on the LIFE engine.

[0012] FIG. **2** is a simplified schematic diagram of centrifugal contactors connected in series.

[0013] FIG. **3** is a simplified plot of the liquid lithium flow processed to recover tritium as a function of extraction efficiency of centrifugal contactors.

[0014] FIG. **4** is a simplified plot showing the liquid lithium fraction that is processed to maintain 0.1 wppm steady state tritium concentration.

[0015] FIG. **5** is a plot showing the number of contactors as a function of the centrifugal contactor extraction efficiency.

[0016] FIG. **6** is a simplified schematic diagram of a tritium recovery system by molten salt extraction according to an embodiment of the present invention.

[0017] FIG. **7** is a simplified schematic diagram of a tritium recovery system utilizing redundancy.

[0018] FIG. **8** is a simplified flowchart illustrating a method of extracting tritium according to an embodiment of the present invention.

DETAILED DESCRIPTION OF SPECIFIC EMBODIMENTS

[0019] Liquid lithium is considered as the primary coolant and breeder material in the design of the Laser Inertial Fusion Energy (LIFE) engine. This work presents the analysis of a tritium recovery system from the blanket liquid lithium for the LIFE engine and is based on a molten salt extraction technology. The goal is to size the primary components of the system. The dimensions of the centrifugal contactors, electrolyzer, and getter are estimated as a function of the desired tritium steady state inventory and the component efficiencies. Results show that a relatively compact system can recover tritium at the needed rate to maintain a low tritium inventory.

[0020] The LIFE Engine is a laser-based energy system. Liquid lithium is being considered as the blanket cooling and breeding material for the LIFE engine due to its many virtues: low density, high thermal capacity, good thermal conductivity, high potential to breed tritium due to its high neutron capture cross-section, and high affinity for hydrogen isotopes that minimizes tritium permeability. High hydrogen affinity, however, complicates tritium recovery while operational safety demands a low tritium inventory. Contact with water is avoided at all times.

[0021] The tritium recovery system is designed to keep the tritium steady state inventory of the LIFE engine power plant at less than about 50 g. The amount of liquid lithium in LIFE pilot plant is ~1000 tons. If we have ~1000 m^3 of liquid lithium in our 1.2 GW(th) LIFE plant we need a steady sate tritium concentration of 0.1 wppm. If our tritium recovery system has 90% efficiency then we need a system that can process ~1% of the lithium flow to decrease tritium concentration from 0.1 wppm to 0.01 wppm.

[0022] FIG. **1** is a simplified schematic diagram illustrating a 1.2 GW (thermal) power plant based on the LIFE engine. The design of the pilot LIFE engine power plant includes four loops, each with an intermediate heat exchanger, steam generator, and reheater. Thus one tritium recovery system processes ~7 kg/s of lithium flow to reduce tritium concentration from 0.1 wppm to 0.01 wppm.

[0023] FIG. **1** illustrates two of the four loops. First, liquid lithium transfers energy from the fusion chamber **110** to the intermediate heat exchanger **112** where the thermal energy of the liquid lithium is transferred to the molten salt. The molten salt in the secondary loop (also referred to as an intermediate loop) carries thermal energy from the intermediate heat exchanger **112** to the steam generator and reheater **114**. The steam then goes to the Rankin cycle turbines. Each of the four loops will also have a tritium recovery system by molten salt extraction (labeled "Lithium Processing" in FIG. **1**).

[0024] Several options have been proposed for tritium recovery from liquid lithium. These include use of a permeable window, a gettering process, a cold trap, distillation, molten salt, and gettering and molten salt combined. Because the molten salt method is based on liquid diffusion at relatively low temperature (~500° C.) it results in a relatively compact system with relatively low energy consumption. For that reason, and others discussed below, the molten salt extraction is utilized in the embodiments described herein.

[0025] FIG. **8** is a simplified flowchart illustrating a method of extracting tritium according to an embodiment of the present invention. As illustrated in FIG. **8**, the molten salt extraction method includes four steps in this embodiment. The method includes mixing a lithium fluid containing trace amounts of tritium with a molten salt (e.g., lithium halides, for example, including LiF, LiCl, LiBr, or combinations thereof) in a centrifugal contactor (**810**). In other embodiments, one or more sodium salts can be utilized as the molten salt. The intimate contact between the molten salt and the trace amounts of tritium in the lithium fluid preferentially extracts the LiT into the salt phase (i.e., a salt of lithium and tritium). The lithium fluid and the salt of lithium and tritium are separated in the centrifugal contactor (**812**). The salt of lithium and tritium is circulated in an electrolyzer and the LiT is oxidized to form T2 (**814**), which is swept from the salt of lithium and tritium by bubbling an inert gas (e.g., argon) through the salt (**816**). Finally, the inert gas is circulated in a titanium getter which recovers the tritium from the inert gas (**818**).

[0026] The most important characteristic of the molten salt extraction method is being free from solid-state diffusion. Liquid phase diffusion is often faster than solid-state diffusion. However, distillation requires very high temperatures. Maroni et al. were the first to propose the use of molten salt extraction for tritium recovery from liquid lithium. In previous experiments molten salt had been used to remove impurities from liquid metals. They observed that some experiments with lithium/hydrogen galvanic cells showed that the lithium hydride formed during discharge was preferentially extracted into the electrolyte.

[0027] As described above, the liquid lithium (also referred to as lithium liquid) that contains tritium is mixed with lithium halide salts in a centrifugal contactor/separator. It has been shown that the volumetric distribution coefficient, Dv (defined as the ratio of tritium content per unit volume in the salt to tritium content per unit volume in the lithium) was between 2 and 4, verifying that the LiT moves preferentially from the liquid lithium to the salt when they are mixed. Then, the salt of lithium and tritium is circulated to an electrolyzer where the LiT is oxidized to form T2, which is swept from the salt phase by bubbling an inert (i.e., noble) gas. The noble gas circulates through a getter that recovers the tritium. It has been estimated that a 20% electrolyzer recovery efficiency is used to capture tritium from the molten salt by measuring the hydrogen isotopes added to the molten salt and the hydrogen concentration in the noble gas coming out of the electrolyzer.

[0028] Embodiments of the present invention provide a molten salt extraction method that can be used in a full-scale fusion system. Important issues that are addressed are: 1) The need to reduce the halide impurities in the primary lithium blanket to safe levels from an activation perspective. This depends on how well the centrifugal contactors separate the lithium and molten salt after LiT is transferred. 2) The need to minimize impurities in the salt that can decrease electrolyzer efficiency. Once again, this depends on how well the contactor-separator works.

[0029] The following provides an analysis of a complete molten salt extraction system to recover tritium from the liquid lithium of the LIFE engine power plant. We estimate the size of the major components and calculate system energy consumption.

[0030] Centrifugal Contactors

[0031] LiF—LiCl—LiBr (22-31-47 mol %) is the primary choice for molten salt in an embodiment although other mixtures can be utilized. The selection of this primary choice was made primarily on melting point consideration: i.e., <450° C. is required. LiF—LiCl—LiBr (22-31-47 mol %) also showed good stability during testing, demonstrating that the particular ratio discussed as the primary choice is not required by the present invention. We consider that due to similar properties of the molten halides the analysis presented here will be valid if another halide is utilized. Most research has been done with 1:1 lithium to salt volume ratio and no issues have been reported, thus the 1:1 ratio is recommended in the interest of reducing overall salt inventory in the plant.

[0032] FIG. **2** is a simplified schematic diagram of centrifugal contactors connected in series. Lithium and salt flow counter-current, mixing and separating in every centrifugal contactor. Lithium and molten salt flows are in counter-current.

[0033] The centrifugal contactor 1 **210** receives salt with no tritium and lithium with low tritium concentration (most tritium in liquid lithium is found as LiT) because tritium is previously extracted in centrifugal contactor 2 **220** and contactor 3 **230**. The two flows mix and separate with the lithium leaving with less tritium and the salt leaving contactor 1 **210** with a small amount of tritium and entering contactor 2 **220** where it mixes with lithium with higher tritium concentration, and so on. Increasing liquid lithium purity utilizes more contactors in series. It is also possible to connect the centrifugal contactors in parallel. In that case, the fraction of lithium flow that needs to be processed increases because the concentration of tritium will be higher after the single contactor stage.

[0034] There is a trade-off between the contacting step (centrifugal contactors) and the salt processing step (electrolyzer). If the contacting step is larger with high energy consumption then the electrolyzer can be smaller and use less energy because less salt with higher tritium concentration will flow from the centrifugal contactors to the electrolyzer. Recovery efficiency determines the number of contactors, and electrolyzer volume is determined by the average current density. The more contactors in series, the less salt volume flows into the electrolyzer, enabling compact electrolyzer design. Some experiments demonstrated **100%** hydrogen recuperation efficiency from molten salt electrolysis. More experimental work is needed, but with the available information it seems better to maximize electrolysis because the centrifugal contactors are more complex and demand more energy. Thus, in some embodiments, parallel centrifugal contactors are utilized, for example, for the pilot LIFE engine plant.

[0035] Electrolyzer

[0036] The electrolysis unit is the least studied component of the system. Recovery efficiency, defined as the weight of the hydrogen recovered from the salt divided by the weight of hydrogen added to the salt, is believed to be the most important electrolyzer parameter. Electrolysis experiments have been conducted to separate hydrogen from molten salt, reporting recovery efficiency ranging from 20% to 100%. The higher efficiency is explained by the use of an argon bubbler that was also the anode of the cell that removed the hydrogen before back mixing occurred. The kinetics of tritium recovery in the contactor units has been studied but did not include a study of electrolyzer kinetics. Typically, an assumption of 90% electrolysis process efficiency is used. It has been shown that applying less than 0.6 V does not release any tritium. Recovery efficiency subsequently increased until reaching a maximum at 0.9 V. After that, recovery efficiency decreases for higher voltage. Voltage should not be increased beyond 1.5 V to avoid molten salt decomposition that occurs at ~2 V.

[0037] In the electrolyzer the LiT in the salt phase is oxidized to become molecular tritium (T2). The T2 collection electrode must be designed to recover tritium before it can react back into the salt phase. In an particular embodiment, a porous electrode enabling surface oxidation of T2 with simultaneous argon flow through the small electrode orifices is utilized to carry away the tritium before back reaction can take place.

[0038] Getter

[0039] The noble gas (e.g., Ar)-T2 mixture flows from the electrolyzer to a getter. Even though there might not be many getters specifically designed to extract tritium from argon, this component is more conventional because there are many hydrogen getters, and argon/helium is unreactive. Several getter materials can be used: titanium, yttrium, and depleted uranium, which is likely the best option.

[0040] System Analysis.

[0041] For the 1.2 GW (thermal) LIFE engine pilot power plant, tritium breeding is estimated at Rb=200 g/d. The total volume of liquid lithium (blanket, pipes, pumps, and intermediate heat exchangers) is estimated to be 100 m^3. At density pLi=480 kg/m^3 at T=500° C., the total lithium mass is 480 tons. The LIFE engine power plant has the goal of keeping the steady state tritium inventory <50 g. Tritium steady state concentration in the lithium loop is therefore ~0.1 wppm. If we used the values of DV=2.9 (measured) and ϵ=90%, we can plot the liquid lithium flow that needs to be processed to recover the tritium as a function of the extraction efficiency of the centrifugal contactors (ii). FIG. **3** is a simplified plot of the liquid lithium flow processed to recover tritium as a function of extraction efficiency of centrifugal contactors. Extraction efficiency is difficult to calculate or estimate because it depends on specific geometry and operating conditions of the centrifugal contactors.

[0042] In one implementation, one volume of liquid lithium is mixed with three volumes of molten salt, resulting in a total mixed flow of 210 m_3/h. If we use the 45 m3/h (200 gpm) centrifugal contactor we need 5 sets of three contactor per loop, or 15 contactor per loop, 60 contactors total.

[0043] FIG. **4** is a simplified plot showing the liquid lithium fraction that is processed to maintain 0.1 wppm steady state tritium concentration. To estimate the number of centrifugal contactors we selected a commercial product that has a maximum throughput of 757 liters per minute (200 gpm), with a footprint of 152 cm×152 cm (60"×60") and height 163 cm (64"). Using an estimate for the extraction efficiency, it is possible to calculate the number of contactors needed for the whole LIFE engine plant as a function of the centrifugal contactor extraction efficiency. FIG. **5** is a plot showing the number of contactors as a function of the centrifugal contactor extraction efficiency.

[0044] If we conservatively assume n=0.4, the total number of centrifugal contactors is 16, or 4 contactors in parallel per loop. FIG. **6** is a simplified schematic diagram of a tritium recovery system by molten salt extraction according to an embodiment of the present invention. As illustrated in FIG. **6**, with four centrifugal contactors (Centrifugal Contactors 1-4) in parallel, the electrolyzer **612**, the getter **614**, and the lithium fluid circuit **620** and the molten salt circuit **622**. In this example, a depleted uranium getter is utilized. Additionally, although argon is used as the noble gas, this is not required by the present invention. As previously described, the molten salt flows from each of the centrifugal contactors to an electrolyzer **612** where the T− is oxidized to form T2, and a stream of argon sweeps the molecules of T2 before back reaction occurs. The argon stream with tritium (and possibly other impurities) goes through a getter unit **614** where depleted uranium absorbs the tritium, which is later released by heating during the regeneration stage.

[0045] The tritium recovery system by molten salt extraction illustrated in FIG. **6** is suitable for use with the 1.2 GW (thermal) LIFE engine power plant or other suitable sources that generate tritium.

[0046] FIG. **7** is a simplified schematic diagram of a tritium recovery system utilizing redundancy. Examples of the redundant systems include the redundant contactors (Centrifugal Contactor 1 Redundant, Centrifugal Contactor mostly salt Redundant, and Centrifugal Contactor mostly lithium Redundant), redundant electrolysis tank (Electrolysis Tank Redundant) and the redundant getters (Getters Redundant). One of ordinary skill in the art would recognize many variations, modifications, and alternatives.

[0047] It is also understood that the examples and embodiments described herein are for illustrative purposes only and that various modifications or changes in light thereof will be suggested to persons skilled in the art and are to be included within the spirit and purview of this application and scope of the appended claims.

What is claimed is:

1. A method for removing tritium from liquid lithium, the method comprising:

mixing the liquid lithium containing trace amounts of tritium with a molten salt;

forming a salt of lithium and tritium;

separating the liquid lithium from the salt of lithium and tritium;

circulating the molten salt in an electrolyzer to form molecular tritium;

bubbling an inert gas through the electrolyzer to remove the molecular tritium; and

circulating the argon and removed molecular tritium in a titanium getter to recover the tritium.

2. The method of claim **1** wherein the molten salt includes lithium halides.

3. The method of claim **2** wherein the halides comprise at least one of fluorine, chlorine, or bromine.

4. The method of claim **1** wherein the liquid lithium contains trace amounts of tritium.

5. The method of claim **1** wherein mixing the liquid lithium with the molten salt and separating the liquid lithium from the molten salt are performed in a centrifugal contactor.

6. The method of claim **1** wherein the inert gas comprises at least one of helium or argon.

7. A system for recovering tritium from a lithium coolant, the system comprising:

a lithium coolant circuit, wherein the lithium coolant in the circuit includes trace amounts of tritium;

a plurality of contactors coupled to the lithium coolant circuit, wherein the plurality of contactors are operable to mix the lithium coolant with a molten salt and form a salt of lithium and tritium;

an electrolysis unit coupled to one or more of the plurality of contactors;

an inert gas source operable to bubble an inert gas through the electrolysis unit; and

a getter coupled to the electrolysis unit and operable to recover the tritium.

8. The system of claim **7** wherein the molten salt comprises at least one of LiF, LiCL, or LiBr.

9. The system of claim **7** wherein the plurality of contactors comprise centrifugal contactors arranged in parallel.

10. The system of claim **7** wherein the getter comprises at least one of a titanium or depleted uranium.

11. The system of claim **7** wherein the inert gas comprises at least one of helium or argon.

* * * * *