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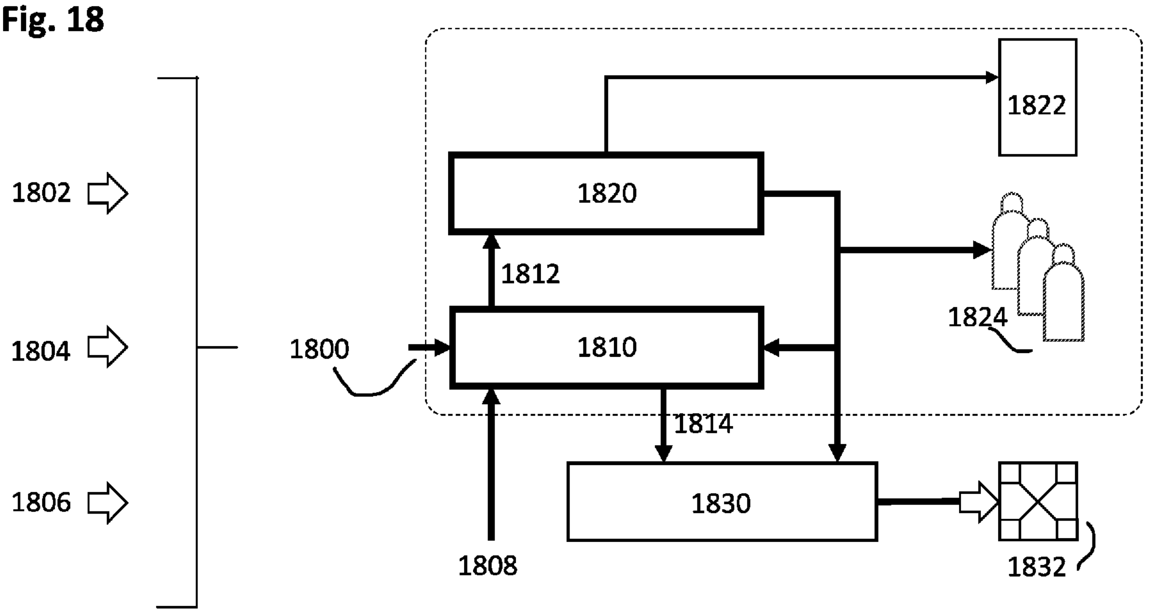
(54) Title: SYSTEMS AND METHODS FOR FAST MOLTEN SALT REACTOR FUEL-SALT PREPARATION

Fig. 18



(57) Abstract: The present disclosure provides systems and methods for fast molten salt reactor fuel- salt preparation. In one implementation, the method may comprise providing fuel assemblies having fuel pellets, removing the fuel pellets and spent fuel constituents from the fuel assemblies, granulating the removed fuel pellets or process feed to a chlorination process, processing the granular spent fuel salt into chloride salt by ultimate reduction and chlorination of the uranium and associated fuel constituents chloride salt solution, enriching the granular spent fuel salt, chlorinating the enriched granular spent fuel salt to yield molten chloride salt fuel, analyzing, adjusting, and certifying the molten chloride salt fuel for end use in a molten salt reactor, pumping the molten chloride salt fuel and cooling the molten chloride salt fuel, and milling the solidified molten chloride salt fuel to predetermined specifications.

[Continued on next page]

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SYSTEMS AND METHODS FOR FAST MOLTEN SALT REACTOR FUEL-SALT PREPARATION

CROSS-REFERENCE TO RELATED APPLICATIONS

[1] This application incorporates by reference in their entirety: U.S. Provisional Patent Appl. No. 63/605,135, filed on December 1, 2023; U.S. Provisional Patent Appl. No. 63/578,044, filed on August 22, 2023; U.S. Provisional Patent Application No. 63/495,416, filed April 11, 2023; U.S. Patent Application No. 18/102,344, filed January 27, 2023, which is a continuation of U.S. Patent Application No. 17/527,862, filed November 16, 2021, which is a continuation-in-part (CIP) of International Application No. PCT/US2020/032902, filed May 14, 2020, which claims priority to, and the benefit of, U.S. Patent Application No. 16/415,668, filed May 17, 2019, and U.S. Patent Application No. 16/415,692, filed May 17, 2019.

TECHNOLOGICAL FIELD

[2] The present disclosure relates generally to methods and systems for processing pelletized-form light water reactor spent nuclear fuel into fluoride-based or chloride-based molten salt reactor fuel.

BACKGROUND

[3] Nuclear fuel that has been irradiated in a nuclear reactor is generally referred to as spent nuclear fuel. Such spent nuclear fuel is generally not reused or recycled but instead is most often stored onsite in specially-design pools in the vicinity of the nuclear reactor in which such nuclear fuel was used. Aged spent nuclear fuel, having undergone significant decay so that it doesn’t produce significant heat, may also be stored in cannisters placed within dry casks on pads at the reactor site in which it was produced, at decommissioned reactor sites, and/or at other approved sites pending disposal at a permanent disposal facility.

[4] According to the U.S. Energy Information Administration, as of 2013, there were more than 70,000 metric tons of spent nuclear fuel stored at sites within the United States (https://www.eia.gov/nuclear/spent_fuel/). Such spent nuclear fuel will be lethal to humans for thousands of years, requiring its storage meet stringent requirements and close monitoring. Further, while spent nuclear fuel storage has proven to be reasonably safe to date, the risk remains that a large fire, explosion, terrorist attack, plane crash, or accident could occur that damages a spent fuel pool and/or dry cask storage of such spent fuel.

[5] Therefore, it would be desirable to have a system, apparatus and/or method that takes into account at least some of the issues discussed above, as well as possibly other issues, and yields an improved solution.

BRIEF SUMMARY

[6] Exemplary implementations of the present disclosure are generally directed to methods and processes involving light water power reactor spent nuclear pellet-form fuel, extracted from fuel assemblies, or, rods containing such spent nuclear pellet-form fuel, separating fuel rod cladding from the pellets, retaining substantially all spent fuel pellets and fragments thereof, and processing to halide salt powder suitable for use, perhaps with enrichment, to fuel for a molten salt reactor.

[7] Exemplary implementations of the present disclosure are generally directed to methods and systems resulting in a substantially complete automated process for making new fuel for a molten salt reactor and which, consequently, reduce the inventory of spent fuel at commercial power reactor sites. In certain of such implementations, virtually none of the contents of the spent fuel are removed, virtually all of the spent fuel and contents are converted to molten salt fuel, and essentially all reprocessed/reprocessed fuel will be capable of generating power, substantially without the generation of additional waste, resulting in what could potentially be near-continuous reduction of currently-stored spent fuel inventory over time, with the consequent volumetric reduction of highly radioactive waste to stable or low-level forms. In certain implementations, during such conversion to molten salt fuel, noble gases Krypton and Xenon may be removed for storage or other uses.

[8] In exemplary implementations of the present disclosure, processes are discussed for converting uranium oxides and plutonium oxides (as used) to uranium and plutonium chlorides. In certain implementations, processes are discussed for converting thorium oxide to thorium chlorides.

[9] Implementations of the present disclosure are generally directed to methods and systems for preparation of a fluoride-based salt fuel for a thermal molten salt reactor (TMSR).

[10] Further implementations of the present disclosure are generally directed to methods and systems for preparation of a chloride-based salt fuel for a fast molten salt reactor (FMSR).

[11] Implementations of the present disclosure are generally directed to methods and systems for retaining essentially all fuel materials in a spent fuel recycling process to substantially fully close the nuclear fuel cycle, thereby virtually eliminating a nuclear waste stream of rejected material. Highly radioactive fission waste and trans-uranium elements, e.g., Actinides in the spent fuel are retained in the process and new fuel product.

[12] Implementations of the present disclosure are generally directed to methods and systems for allowing fission waste products and actinides in spent nuclear fuel to be burned to stable forms and incorporate salt fuel for enrichment in order to overcome the negative power effects of the fission product waste.

[13] Certain implementations of the present disclosure are generally directed to methods and systems for reducing spent nuclear fuel and constituents to fluoride salt fuel, including calculated enrichment (U235 or Pu239). Uranium or plutonium is added to the spent fuel before or during fluoridation. Fertile thorium oxide may be added and fluorinated to thorium fluoride salt in certain implementations. The final product is a dry fuel salt powder.

[14] Other implementations of the present disclosure are generally directed to methods and systems for producing chloride fuel salt by molten chloride reduction of spent nuclear fuel and constituents, including calculated enrichment (U235 or Pu239). Uranium or plutonium is added during chlorination. While not a fuel, fertile thorium oxide may be added and fluorinated to thorium fluoride salt in certain implementations. The final product is a dry salt fuel powder. In a variation of this implementation, molten salt fuel product may be poured into canisters, stored, and later inductively heated to liquid and used in the molten salt reactor.

[15] Implementations of the present disclosure are generally directed to methods and systems that avoid chemical separation of spent fuel constituents into separate streams (which would generally result in more storage of highly radioactive waste) or additional chemical processes to rejoin spent fuel groups.

[16] According to one example implementation, a facility and process design of the present disclosure allows operators to process spent nuclear fuel stored at nuclear power plants. The process, once put into practice, essentially closes the nuclear fuel cycle, by reducing the amount of nuclear spent fuel stored at plant sites, and molten salt reactor fuel produced in accordance with this disclosure is intended to be used in advanced molten salt reactors to ultimately generate electricity, hot water, etc. According to another exemplary implementation, a process facility is provided that is a hardened, secure, limited-access facility, configured to accept and contain highly radioactive material spent nuclear fuel, and specifically designed to house apparatus machinery and attendant support systems for continuous spent fuel processes, including spent fuel container ingress and processing spent fuel to a final packaged product, and egress of such final product. This includes shielding against radiation, remote robotic operations and safe handling, and the exclusion of personnel from processing area.

[17] In another example implementation, a facility for conducting a process of the present disclosure is located on site within a secured perimeter, together with a spent nuclear fuel storage facility and a molten salt reactor.

[18] According to a further exemplary implementation of the present disclosure, processes are disclosed capable of utilizing virtually all light water-reactor ceramic spent nuclear fuel consisting of uranium oxide, lanthanide series elements, fission product metals and non-metals, actinide series elements, i.e., substantially all of the material found in spent nuclear fuel, for the production of halide salt fuel. These processes avoid “wet” chemical separation, since element (nuclide) constituents are not chemically separated. Production of fluoride salt is by a dry process, whereas, the process for the production of chloride salt is a liquid emersion conversion to salt. There are effectively no left-over waste products or waste streams.

[19] According to one further exemplary implementation of the present disclosure, fuel assemblies containing an array of fuel tubes are aligned horizontally on a rod puller disassembly table, and spent fuel pellets are removed from tubes, “rods,” or, “pins,” (collectively referred to herein as “rods”) by laser slitting of the fuel tubes, opening the tubes, and mechanically removing any spent fuel pellets and fragments that remain adhered to tubes. Cleaned fuel tubes, channels, and assembly end pieces (non-fuel), are set aside for recycling.

[20] According to one further exemplary implementation of the present disclosure, spent fuel pellets and fuel pieces are processed for a specific molten salt reactor type, i.e., fluorinated salt for a “thermal” reactor and chlorinated salt for a “fast” reactor. Specifically, fuel for a thermal molten salt reactor is processed to a fluoride salt fuel by ultimate reduction and fluorination of uranium and its associated fuel constituents, and fuel for a fast molten salt reactor is processed to a chloride salt fuel by ultimate reduction and chlorination of uranium and its associated fuel constituents. Fertile thorium oxide may be added and fluorinated to thorium fluoride salt in certain implementations for a thermal molten salt reactor, or chlorinated to thorium chloride salt, for a fast molten salt reactor.

[21] According to one further exemplary implementation of the present disclosure, spent fuel pellet removal from fuel pins is performed in a closed atmosphere to prevent release of dust and to capture gases back into the process. In such implementations, fuel for a molten salt reactor proceeds to one of two parallel path, specifically: (a) a path wherein at least one rotating calciner is used in a fuel fluorination preparation process, which receives extracted spent fuel pellets directly (“Option A”), or (b) a path wherein chlorinated fuel preparation spent fuel pellets are ball-milled in an enclosed atmosphere to collect dust and gases, which are recycled back into the process, and spent fuel pellets are milled to coarse granular feed to the molten salt uranium/plutonium oxide reduction tank (“Option B”). Fertile thorium oxide may be added and fluorinated to thorium fluoride salt using the calciner method, or chlorinated to thorium chloride salt by addition of the thorium oxide to the uranium/plutonium oxide reduction tank.

[22] According to a further exemplary implementation of the present disclosure, spent fuel pellets undergoing Option A, i.e., the fluorination process, may be enriched by adding U235 powder to the spent fuel before fluorination, to predetermined specifications, to provide sufficient fissionable material in the final product. Non-fuel, but fertile thorium oxide, may be added to the calciner, and fluorinated to thorium fluoride salt. This enrichment could be low enriched uranium including high assay-low enriched uranium (e.g., HA-LEU, < 20% enrichment). In one implementation, spent fuel and constituents are reduced to fluoride salt powder in a rotary calciner, and the powder may be enriched as necessary with fluorinated uranium-235 or plutonium-239 in order to support molten salt reactor operation.

[23] According to yet a further exemplary implementation of the present disclosure, spent fuel undergoing Option B, a chlorination process, may be enriched with U235 or Pu239 powder added to spent fuel salt at a uranium/plutonium oxide reduction tank, to the desired specifications, to thereby provide enough fissionable material in the final product. Fertile thorium oxide, may be added to the calciner, and fluorinated to thorium fluoride salt. This enrichment could be low enriched uranium including high assay-low enriched uranium (e.g., HA-LEU, < 20% enrichment) or plutonium, or mixed oxide (MOX) fuel. In one implementation, spent fuel and constituents are reduced to chloride salt fuel by immersion in a molten chloride salt bath. Molten chloride salt fuel may then be enriched, in the event initial and subsequent enrichments will be required, with uranium-235, plutonium-239, or MOX (uranium and plutonium), added to the oxide reduction tanks, in order to support molten salt reactor operation.

[24] Proper sizing of tanks and equipment, including, without limitation, multiple oxide reduction tanks being in parallel, and multiple mixing and adjustment tanks being in parallel, allows for enrichment and chemical analysis, and required tank separation and reactivity monitoring. Proper sizing ensures ample volume for mole-fraction concentration specifications. Mixing tanks provide for sampling, adjustment, and content certification. Separation, and partitioning, by high-density boron impregnated slabs between all tanks, of both sets of tanks, a first group (oxide reduction tanks) and a second group (mixing and adjustment tanks), ensures sub-criticality during the entire process. Molten chloride salt fuel is solidified and milled to powder in the process. Additional chemical processes, such as fluidized beds, small chemical reactors, or ion exchange columns (not shown), may be employed to process fuel dust or volatile constituents to stable form and to subsequently rejoin spent fuel process streams. Final-product molten salt fuels are provided in powder form, sample-tested, and certified.

[25] According to a further exemplary implementation of the present disclosure, in the case of chlorinated molten salt, initial calculations determine how much granulated spent fuel will be added to each oxide reduction tank of molten alkali or alkali earth chloride by mass and concentration of free chloride. The number and size of molten chloride reaction tanks necessary for continuous process operation to produce chlorinated salt-fuel will be determined, in part, by the rate of spent fuel pellet production and milling, spent fuel chlorination to salt, and safety considerations.

[26] According to a further exemplary implementation of the present disclosure, spent fuel gases evolved during the Options A and C fluorination and Option B chlorination processes, are collected by a fluidized bed, converted to fluorinated and chlorinated fuel salts, respectively, and returned to their separate fuel salts.

[27] According to yet another exemplary implementation of the present disclosure, chlorinated fuel salt is pumped from at least one reacting tank, by its own pump when it and its discharge isolation valve are selected. Fuel salt is pumped to a common header, with a selector valve for each to admit fuel salt to at least one cooling tray. Piping and valves from reacting tanks to cooling trays are maintained above salt melt temperature by redundant and remotely replaceable heating element jackets, to prevent salt fuel solidification.

[28] According to a still further exemplary implementation of the present disclosure, chlorinated fuel salt in the molten state is introduced into cooling trays, cooled by chilled water, designed with multiple parallel, but separate rows, each surrounded by cooling coils to remove heat from the molten salt and cause it to solidify. The cooling trays are in a stacked array, with adequate space between trays for addition of molten salt and removal of solidified. Actual configuration and groupings of a number of trays into one array, depends on the: (a) movement of ganged-arrays; (b) tray loading from the pump-out of the mixing and adjustment tanks; (c) stacked array movement to cooling stations; (d) stacked array movement to the ball mill feed table (not shown); (e) solidified salt fuel removal and deposit on the ball mill feed table; and (f) salt fuel milling.

[29] According to a yet further exemplary implementation of the present disclosure, solidified and cooled chlorinated fuel salt bars, or "sticks," are removed from their molds, after solidification. These sticks are collected and fed to a ball mill and fine mill, which is enclosed to retain process dust, for processing to powder, and milled to specifications. The number of cooling trays will be sufficient to support continuous feed to the milling operation and chlorinated fuel salt powder system demand.

[30] According to a yet another exemplary implementation of the present disclosure, fuel salt product from Option A, is collected from the calciner and milled to powder product specifications, and fuel salt product from Option B is collected from fuel salt mold, ball-milled, and further milled to a fine powder. An alternate implementation of Option B method provides certified fuel salt directly to individual storage canisters, which are inductively heated to liquid for use in a molten salt reactor.

[31] According to a yet another exemplary implementation of the present disclosure, end products of the fluorination process (Option A), and end products of the chlorination process (Option B) include their respective salt fuels and are milled to specifications and then collected in containers on carts, sealed, and transported by cart for direct use or storage. Container geometry and amount of salt fuel product are sized to prevent criticality in stored arrays. Fission product nuclides and other neutron absorbing barriers help assure adequate margin to criticality in all potential concentrations.

[32] Another exemplary implementation of the present disclosure in the production of fuel for a thermal molten salt reactor by: providing spent fuel pellets; processing the spent fuel pellets and fuel pieces into a fluoride fuel salt by ultimate oxidation, reduction, and fluorination of uranium and its associated fuel constituents in a generally continuous progression, wherein the processing produces water vapor; and filtering, condensing, and removing the water vapor produced during the reduction and fluorination operations. While not a fuel, fertile thorium oxide, may be added to the calciner, and fluorinated to thorium fluoride salt in certain implementations.

[33] Another exemplary implementation of the present disclosure provides a method for producing fuel for a thermal molten salt reactor including: (a) providing fuel assemblies containing an array of fuel tubes aligned horizontally on a rod puller disassembly table and removing fuel pellets from the tubes; (b) processing the spent fuel pellets and fuel pieces into a fluoride salt by ultimate oxidation, reduction, and fluorination of uranium and its associated fuel constituents; and (c) filtering (including condensing and removing) the water vapor formed during the reduction and fluorination operations. In an additional exemplary implementation, the method could further include enriching the granular spent fuel salt with U235, and if further desired, fluorinating the U235-enriched granular spent fuel salt or plutonium in a calciner rotary kiln or fluidized bed.

[34] In some exemplary implementations, a method for producing fuel for a fast molten salt reactor is provided which includes providing fuel assemblies removing fuel pellets containing uranium from the fuel assemblies and granulating the fuel pellets into granular spent fuel salt for processing feed in a semi-voided atmosphere using a ball mill, roller mill, or chopping mill, and processing the granular spent fuel salt into chloride salt by ultimate reduction and chlorination of the uranium and associated fuel constituents of the uranium. The method further includes enriching the granular spent fuel salt with U235, Pu239, or MOX, chlorinating the enriched granular spent fuel salt to yield molten chloride salt fuel using anhydrous HCl and halide salt reduction, and then analyzing, adjusting, and certifying the molten chloride salt fuel for end use in a molten salt reactor. A non-fuel, fertile thorium oxide, may be added to the uranium/plutonium oxide reduction tank and chlorinated to thorium chloride salt. Additionally, the method includes pumping the molten chloride salt fuel to stacked arrays of cooling trays or canisters and cooling the molten chloride salt fuel to yield solid salt fuel bars, sticks, or canister solid forms and milling the solidified molten chloride salt fuel to predetermined specifications for the fast molten salt reactor.

[35] A method for producing fuel for a fast molten salt reactor, the method including providing fuel assemblies and removing fuel pellets containing uranium and all spent fuel constituents, from the fuel assemblies and granulating the fuel pellets in a semi-voided atmosphere using a ball mill, roller mill, or chopping mill, for process feed to the chlorination process. The granular spent fuel salt is processed into chloride salt by ultimate reduction and chlorination of the uranium and associated fuel constituents chloride salt solution. Fertile thorium oxide, may be added to the uranium/plutonium oxide reduction tank and chlorinated to thorium chloride salt. Reduction may occur using a strong reducing agent, preferably a chloride-containing reducing agent, such as anhydrous hydrogen chloride (AHCl). The granular spent fuel salt is enriched with U235, Pu239, or MOX, and the enriched granular spent fuel salt is chlorinated to yield molten chloride salt fuel using AHCl halide salt reduction. The molten chloride salt fuel is analyzed, adjusted, and certified for end use in a molten salt reactor. This implementation also includes pumping the molten chloride salt fuel to stacked arrays of cooling trays or canisters and cooling the molten chloride salt fuel to yield solid salt fuel bars, sticks, or canister solid form, and milling the solidified molten chloride salt fuel to predetermined specifications for the fast molten salt reactor.

[36] Non-limiting example approximate temperatures, times, gas concentrations, materials used to construct the apparatus, and other parameters which are expected to be used are shown in the drawings.

[37] More specifically, implementations of the present contemplate a full-size facility enclosing the methods and processes for processing commercial light water reactor spent nuclear fuel to final product fuel salt product for a molten salt power reactor. Equipment and machinery in the facility receive spent fuel assemblies and deliver them to a rod pulling table, and disassemble them from their support elements, namely, into separated, individual cladding fuel rods containing raw spent fuel. The fuel rods are slit and/or sliced axially along substantially their entire length by laser. Care is taken to prevent the laser from cutting through the spent fuel, burning, or fusing pellets. In one implementation, the laser simultaneously cuts opposite sides of each rod into semi-cylindrical halves, thereby exposing fuel pellets when the two halves of the fuel rod are separated. Prior to the fuel rod cladding sections being removed from the process, mechanical brushes sweep the inside surfaces of such sections lengthwise in order to recover all of the spent fuel pellets and pieces thereof, given such fuel pellets and pieces may exhibit various forms, from being generally intact, i.e., cylindrical, to broken and deformed shapes, indicated by previous cycles of operating history and subsequent handling. Gaseous constituents of the spent fuel are collected during disassembly and the conversion processes discussed herein and are processed within a fluidized bed into halides for recovery.

[38] In one exemplary implementation, thermal reactor salt fuel requires low neutron energy (thermal energy) for thermal fission to occur, whereby neutrons immediately begin to lose energy quickly after they are produced from fission to continue the process of thermal fission. This is achieved by conversion of spent fuel to salt fuel of light mass elemental salt, whereby light mass elemental metals of beryllium or lithium, for example, and fluorine, form salt fuel effecting thermal fission.

[39] In another exemplary implementation, during fluorination, light mass elemental metal hydrides of beryllium or lithium, for example, form salt fuel effecting thermal fission by reduction of spent fuel to salt fuel and oxidation of hydride light mass metals to salts, whereby during operation, thermal molten salt reactor neutrons quickly lose energy to thermal energy, after they are produced from fission, to continue the process of thermal fission.

[40] In one exemplary implementation, fast reactor salt fuel requires high neutron energy for fast fission to occur, and such energy is desired to be greater than the threshold for fast neutron energy, whereby neutrons retain enough energy after they are produced from fission to continue the process of fast fission. This is achieved by conversion of spent fuel to salt fuel of heavier mass elemental salt, whereby heavy mass elemental metals of potassium, zirconium, or zinc, for example, and halides of chlorine, bromine or iodine, form salt fuel effecting fast fission.

[41] In another implementation, during halogenation, heavy mass elemental metal hydrides of zirconium, molybdenum, or tin, for example, form salt fuel effecting fast fission by reduction of spent fuel to salt fuel and oxidation of hydride heavy mass metals to salts, whereby during operation, fast molten salt reactor neutrons retain energy well above fast neutron threshold energy after they are produced from fission to continue the process of fast fission.

[42] According to a further exemplary implementation of the present disclosure, processes are disclosed capable of utilizing virtually all water-reactor ceramic spent nuclear fuel consisting of uranium oxide, lanthanide rare-earth series elements, fission products, actinide series elements, i.e., substantially all of the material found in spent nuclear fuel, for the production of halide salt fuel. These processes avoid “aqueous wet” chemical separation, since fission product waste and actinide constituents are not chemically separated from nuclear fuel material. Production of fluoride salt fuel and chloride salt fuel, and waste fission products and actinides, is a liquid immersion conversion in molten halide salt fuel. There are effectively no left-over waste products or waste streams.

[43] According to another exemplary implementation of the present disclosure, fuel assemblies containing an array of fuel tubes are aligned horizontally in a disassembly table box for breakdown of the fuel assembly into individual fuel rods, and relocation of fuel rods to a pellet extraction grid table. Spent fuel pellets are removed from tubes, by horizontally laser cutting the fuel tube on both sides the entire length of the tube, opening the tubes, and mechanically removing any spent fuel pellets and fragments that remain adhered to tubes. Cleaned fuel tubes, channels, and assembly grid spacers and end pieces, are set aside for recycling.

[44] In a further exemplary implementation of the present disclosure, spent fuel pellet removal from fuel pins or tubes, is performed in a closed atmosphere to prevent release of dust and gases, which are recovered, converted to salt fuel and returned back into the process. In such implementations, fuel for a molten salt reactor proceeds to one of two parallel paths, specifically: (a) spent fuel pellets are milled in a closed atmosphere, to a powder in preparation for conversion to thermal molten salt reactor fluoride salt fuel as feed to the uranium/plutonium oxide reduction tank, or (b) spent fuel pellets are milled in a closed atmosphere, to a powder in preparation for conversion to fast molten salt reactor chloride salt fuel as feed to the uranium/plutonium oxide reduction tank.

[45] According to a further exemplary implementation of the present disclosure, spent fuel and constituents are reduced to fluoride salt fuel by immersion in a molten fluoride salt bath.

[46] In another exemplary implementation, a method further includes enriching the powdered spent fuel salt with U235, Pu239, or MOX fuel, chlorinating the enriched powdered spent salt fuel to yield molten chloride salt fuel using anhydrous hydrogen chloride (AHCl) reduction, in molten chloride salt, and then analyzing, adjusting, and certifying the molten chloride salt fuel for end use in a molten salt reactor. The method further includes ultimate reduction of uranium oxide and constituent oxides using metal hydrides. Generally, in exemplary implementations of the present disclosure processes for conversion of powdered spent nuclear fuel, used fuel, to molten salt reactor salt fuel begins with a starting base bath of molten halide salt, or a mixture of halide salts as the molten medium to dissolve all spent fuel constituents. Particular acids of the halides e.g., hydrogen – fluoride, chloride, bromide, or iodide, may be used for halogenation of uranium, plutonium, fission products and actinides by “fluorination,” “chlorination,” “bromination,” or “iodination” of powdered spent nuclear fuel, converting it to “salt fuel.” Generally, halide salt, e.g., sodium chloride, potassium chloride, cesium chloride or strontium chloride, and anhydrous hydrogen chloride are used for spent fuel conversion to chloride salt fuel. This is necessary to initialize and maintain a continuity of salt fuel physical and nuclear characteristics.

[47] Other exemplary implementations of the present disclosure include a method of processing spent nuclear fuel pellets into molten salt reactor fuel, the method comprising milling the spent nuclear fuel pellets into spent nuclear fuel powder and feeding to a halide forming process, wherein the halide includes at least one of fluoride, chloride, bromide, and iodide, and processing the spent nuclear fuel powder into halide salt by ultimate reduction; halide forming of the uranium and associated fuel constituents in a halide salt solution comprised of a bath of selected metal hydride salts; enriching the halide salt; and halogenating the enriched halide salt to yield molten halide salt fuel. In certain implementations, fertile thorium oxide, a non-fuel, may be reduced to a halide salt.

[48] Exemplary implementations of the methods may include that the step of processing the spent nuclear fuel powder into halide salt occurs by reacting the halide salt with at least one of anhydrous hydrogen halide and metal hydride, which could occur in an oxide reduction tank, and in certain exemplary implementations may include the anhydrous hydrogen halide and/or metal hydride being provided via a sparger in an oxide reduction tank. Fertile thorium oxide may be added to the uranium/plutonium oxide reduction tank and converted to halide salt. Enrichment of the halide salt may also take place in the oxide reduction tank, and hydrogen may be created, converted to water, with the water being generally continuously removed from the oxide reauction tank.

[49] Certain exemplary implementations of the present disclosure include placing the molten halide fuel salt in a canister, covering the molten halide fuel salt with argon or nitrogen gas, and sealing the canister with the molten halide fuel salt and argon or nitrogen gas therein.

[50] Other exemplary implementations of the present disclosure include a method of processing spent nuclear fuel pellets into molten salt reactor fuel, the method comprising milling the spent nuclear fuel pellets into spent nuclear fuel powder and feeding to a fluoride forming process; processing the spent nuclear fuel powder into fluoride salt by ultimate reduction; fluoride forming of the uranium and associated spent nuclear fuel powder constituents in a fluoride salt solution comprised of a bath of selected metal hydride salts; enriching the fluoride salt; and fluorinating the enriched fluoride salt to yield molten fluoride salt fuel.

[51] Certain exemplary implementations of the present disclosure may include the step of processing the spent nuclear fuel powder into fluoride salt includes reacting the fluoride salt with anhydrous hydrogen fluoride, which could be in an oxide reduction tank and potentially through use of a sparger in the oxide reduction tank. Also, the fluoride salt could be enriched in the oxide reduction tank. Fertile thorium oxide may be added and fluorinated to thorium fluoride salt in certain implementations.

[52] Exemplary implementations of the present disclosure may include placing the molten fluoride fuel salt in a canister; covering the molten fluoride fuel salt with argon gas; and sealing the canister with the molten fluoride fuel salt and argon gas therein.

[53] In some exemplary implementations, spent fuel gasses from the spent nuclear fuel powder are collected by a fluidized bed of chemical reactor and converted to fluorinated fuel salts.

[54] Other exemplary implementations of the present disclosure include a system for processing spent nuclear fuel pellets into molten salt reactor fuel, the system including a mill configured for milling the spent nuclear fuel pellets into spent nuclear fuel powder and an oxide reduction tank configured for receipt of the spent nuclear fuel powder and for containing a process for forming the spent nuclear fuel powder into halide salt by ultimate reduction; halide forming of the uranium and associated spent nuclear fuel powder constituents in a halide salt solution comprised of a bath of selected metal hydride salts; enrichment of the halide salt; and halogenating the enriched halide salt to yield molten halide salt fuel. Fertile thorium oxide powder may be added to the oxide reduction tank, by ultimate reduction to thorium halide salt.

[55] In still other exemplary implementations of the present disclosure, a system is provided for processing spent nuclear fuel pellets into molten salt reactor fuel and includes a mill configured for milling the spent nuclear fuel pellets into spent nuclear fuel powder and an oxide reduction tank configured for receipt of the spent nuclear fuel powder and for containing: a process for forming the spent nuclear fuel powder into fluoride salt by ultimate reduction; fluoride forming of the uranium and associated spent nuclear fuel powder constituents in a fluoride salt solution comprised of a bath of selected metal hydride salts; enrichment of the fluoride salt; and fluorination of the enriched fluoride salt to yield molten fluoride salt fuel. Fertile thorium oxide powder may be added and fluorinated to thorium fluoride salt.

[56] In still other exemplary implementations of the present disclosure, cover and off-gas(es) from reactions in the molten salt immersed hydrohalogenation reaction vessel are separated by reflux condensation in a reflux condenser above the molten salt, which returns to the molten salt immersed hydrohalogenation reaction vessel condensate, said condensate comprising all metal and metal salt vapors emanating as part of gas from the molten salt immersed hydrohalogenation reaction vessel returning back to the molten salt immersed hydrohalogenation reaction vessel for the metals in the condensate to be hydrohalogenated to metal salts, while allowing gaseous passage of reflux condenser effluent gas comprising cover gas and hydrohalogenation reaction off-gases and released SNF fission product gases which are then separated by distillation whose gaseous effluent is dry distillation gas effluent that contains appreciably no H_2O or hydrogen halide or hydrohalic acid and whose liquid condensate contains appreciably all of the H_2O or hydrogen halide or hydrohalic acid emanating as part of reflux condenser effluent gas; which liquid condensate is then separated to H_2O and hydrogen halide, the hydrogen halide of which is recombined with the previously separated dry distillation gas effluent to yield dry cover/off-gas (C/OG) that is recirculated and fed back to the molten salt immersed hydrohalogenation reaction vessel, so that any hydrogen halide in the dry C/OG may once more serve as hydrohalogenation reagent.

[57] In still other exemplary implementation of the present disclosure, substantially uranium dioxide (UO_2) spent nuclear fuel rod/pin contents in a sealed batch reactor are first carbochlorinated to substantially uranium pentachloride (UCl_5) under pressure and increasing temperature, with phosgene (COCl2) gas separated from carbochlorination byproduct gases recovered and held from prior carbochlorination event(s), then with liquid carbon tetrachloride (CCl_4) separated and condensed from carbochlorination byproduct gases and liquids recovered and held from prior carbochlorination event(s), then with fresh liquid CCl_4; and then reduced to substantially uranium tetrachloride (UCl_4), by heating in inert gas; all of which process steps whose product gases are recovered and separated and either bottled as salable product gas, or held or bottled for reuse in carbochlorination operations.

[58] In still other exemplary implementations of the present disclosure, substantially uranium dioxide (UO_2) spent nuclear fuel rod/pin contents are vapor phase carbochlorinated to substantially or completely to uranium tetrachloride (UCl_4) in a fluidized bed reactor with phosgene ($COCl_2$) gas separated from carbochlorination byproduct gases recovered and held from prior carbochlorination event(s), then with carbon tetrachloride (CCl_4) gas separated from carbochlorination byproduct gases recovered and held from prior carbochlorination event(s), then with fresh CCl_4 gas; and then optionally heated in the fluidized bed in inert gas charge after said vapor phase carbochlorination to reduce any product uranium pentachloride (UCL_5) resulting from the vapor phase carbochlorination to UCl_4, thus yielding a salt whose uranium salt content is appreciably all UCl_4.

[59] In still other exemplary implementations if the present disclosure, substantially uranium dioxide (UO_2) spent nuclear fuel rod/pin contents recovered from mechanical decladding are carbochlorinated to substantially or completely to uranium tetrachloride (UCl_4) by either liquid or vapor phase CCl_4 carbochlorination, followed by decomposition of any resultant UCl_5 to UCl_4; in parallel with carbochlorination of U3O8 product from voloxidation of SNF cladding to UCl_5, followed by decomposition of resultant UCl_5 to UCl_4. The UCl_4 product from the parallel carbochlorination/decomposition streams are optionally reduced to UCl_3 with hydrogen (H_2); all the while using recovery and recirculation or reuse of cover/off-gases in order to enhance reagent -charge exposure, minimize the amount of reagent necessary, and eliminate need to release byproducts as emissions or waste.

[60] Implementations of the present disclosure can also include the steps and means for halogenation of the uranium oxides to fuel salt and to byproduct at least one off-gas; separating of the off-gas into streams including a stream of halogenation agents and a stream of the balance of the off-gas; and reintroducing the stream of halogenation agents into the halogenation of the uranium oxides. Also disclosed are the steps and means for off-gas processing including separating and storing the stream of the balance of off-gas and use of a cover gas and at least one of anhydrous hydrogen chloride and anhydrous hydrogen fluoride as halogenation agent. Further implementations include the steps and means for the halogenation occurring in a bath including a molten salt and/or halogenation of spent nuclear fuel having fission products; condensing vapors of fission product metals and halogenation salts; and returning the condensed vapors to the bath. Note that the halogenation may include use of anhydrous hydrogen fluoride in at least one fluidized bed.

[61] Additional implementations include the steps and means for converting uranium oxides to fuel salts suitable for use in molten salt reactors and outputting to the off-gas processing the cover gas and the at least one off-gas and wherein the separating of the at least one off-gas further includes separating hydrohalic acid from the balance of the at least one off-gas; separating the hydrohalic acid to water and dry hydrogen halide; and recirculating the dry hydrogen halide to the halogenation or storing the dry hydrogen halide. Implementations of the systems and methods include halogenation of spent nuclear fuel having fission products; condensing vapors of fission product metals and halogenation salts; and returning the condensed vapors to a fluidized bed.

[62] Certain implementations of the present disclosure include methods and systems for converting uranium oxides to fuel salts suitable for use in molten salt reactors and include the means and steps comprising a first chlorination of the uranium oxides to fuel salts through use of phosgene recovered from a prior chlorination operation, and where subsequent to the first chlorination, a second chlorination of the uranium oxides to fuel salts through use of carbon tetrachloride recovered from a prior chlorination operation. Also, subsequent to the second chlorination, a third chlorination of uranium oxides to fuel salts through use of fresh carbon tetrachloride. Subsequent to the third chlorination, reduction of uranium pentachloride, if any, occurs to salts of substantially uranium tetrachloride by heating in inert gas; and subsequent to the reduction, addition of the uranium tetrachloride from the uranium pentachloride reduction process to the uranium tetrachloride from at least one of the first chlorination process, the second chlorination process, and the third chlorination process. Additional variations include reduction of salts of substantially uranium tetrachloride to salts of substantially uranium trichloride by heating with hydrogen and, where the uranium oxides are from spent nuclear fuel having fission products, condensing vapors of the fission product metals and chlorination salts, and returning the condensed vapors to at least one of the first chlorination, the second chlorination, and the third chlorination. Further, the separating of the off-gas may include separating carbon tetrachloride and phosgene from the balance of the at least one off-gas, and recirculating or storing the carbon tetrachloride and the phosgene.

[63] Other implementations may also include steps and means for separation of balance of at least one off-gas includes a cryogenic distillation process that removes oxygen and ozone from the balance of the at least one off-gas, and, where the inert cover gas of the halogenation process is carbon dioxide, the process for separation of the balance of the off-gases can involve use of absorption in liquid carbon dioxide.

[64] Moreover, implementations of the present disclosure may include steps and means for packing the fuel salts in sealed, dry-cover-gas-charged storage containers for later processing, and in certain implementations, blending the fuel salts with a dry carrier and dilutant salt and storing the fuel salts after blending in at least one of granular form, powder form, and solid form, where the solid form is configured to be in countable units and includes at least one of countable ingots, blocks, bricks, ingots, cylinders, semi-cylinders, rods, and spheres, and combinations of one or more of the blocks, said bricks, said ingots, said cylinders, said semi-cylinders, said rods, or said spheres. Additionally, implementations can include steps and means for milling, grinding, shredding, or reducing the solid form of the fuel salts into powder form and/or granular form, and if desired, formation of the solid form into countable unitary solid form blocks by selective laser melting of the fuel salts.

[65] Implementations of the present disclosure may also include steps and means for, in the case where the uranium oxides are from spent nuclear fuel removed from spent nuclear fuel storage containers or casks, and packaging of the blocks in sealed, inert-gas-charged compartments in the spent nuclear fuel storage containers or casks. Other implementations may include transporting the countable units to a molten salt reactor, and reducing the countable units into powder form and/or granular form milling, grinding, and/or shredding, and introducing the powder and/or granular form of the countable units into the molten salt reactor.//Furthermore, implementations of the present disclosure may include steps and means for enriching the fuel salts, and also, halogenation of thorium oxide or adding thorium halide to the fuel salts to affect the addition of fertile salt.

[66] The features, functions and advantages discussed herein may be achieved independently in various exemplary implementations or may be combined in yet other exemplary implementations further details of which may be seen with reference to the following description and drawings.

BRIEF DESCRIPTION OF THE DRAWINGS

[67] Having thus described exemplary implementations of the disclosure in general terms, reference will now be made to the accompanying drawings, which are not necessarily drawn to scale, and wherein: FIG. 1 schematically illustrates methods and systems according to exemplary implementations of the present disclosure for use in processing spent nuclear fuel into molten salt reactor fuel, and more specifically, a fluoride-based salt fuel for a thermal molten salt reactor (TMSR);

[68] FIG. 2 schematically illustrates methods and systems according to an exemplary implementation of the present disclosure for use of a molten salt preparation in processing spent nuclear fuel into chloride or flouride fuel salt, and more specifically, the receiving of pellet-form spent fuel, which has been milled to pulverized powdered form and uranium/plutonium oxide reduction tank, wherein the purpose is to remove oxygen and prevent production of other oxides, results in the removal of oxygen as water and conversion of generated hydrogen to water, obviating the need for using costly catalysts or chemicals not readily available, and upon oxygen and hydrogen being removed from the fuel salt, substantially the only byproduct is water;

[69] FIG. 3 schematically illustrates the mixing and adjustment tank, illustrates methods and systems according to exemplary implementations of the present disclosure for use of a molten salt sampling, adjustment, and certification, in processing spent nuclear fuel into chloride or flouride fuel salt;

[70] FIG. 4 schematically illustrates in plan view an exemplary methods and implementations of the present disclosure for use of a molten salt preparation in processing spent nuclear fuel into chloride or flouride fuel salt, and more specifically, typical separation for operation and critical-safe parallel arrangements of molten salt spent fuel oxide reduction and mixing tanks, including oxide reduction tanks and mixing and adjustment tanks and their relative orientation and physical separation, and physical separation by neutron absorbing panels, generally boron composites, to ensure subcriticality between parallel arrangements of oxide reduction tanks, and the same neutron absorbing panels between parallel arrangements of mixing and adjustment tanks to ensure subcriticality, and including an oxide reduction tank discharge header discharges to the mixing and adjustment tank without using nozzles;

[71] FIG. 5 schematically illustrates methods and systems according to exemplary implementations of the present disclosure for use of a molten salt preparation in processing spent nuclear fuel into chloride or flouride fuel salt, and more specifically, salt mold cooling trays;

[72] FIG. 6 schematically illustrates methods and systems according to exemplary implementations of the present disclosure for use of a molten salt preparation in processing spent nuclear fuel into chloride or flouride fuel salt, and more specifically, a salt mold cooling tray, including a top cover, cooling molds, and heating and cooling coils;

[73] FIG. 7 schematically illustrates a method and system according to an exemplary implementation of the present disclosure, including, a process for thermal molten salt reactor (TMSR) fuel-salt preparation;

[74] FIG. 8 schematically illustrates a method and system according to an exemplary implementation of the present disclosure, including, processes for fast molten salt reactor (FMSR) and thermal molten salt reactor (TMSR) fuel-salt preparation;

[75] FIG. 9 schematically illustrates methods and systems according to an exemplary implementation of the present disclosure for use of a molten salt preparation in processing spent nuclear fuel into chloride or flouride fuel salt;

[76] FIG. 10 schematically illustrates a site on which components of a system according to an example implementation of the present disclosure may be located;

[77] FIG. 11 schematically illustrates a process for converting commercial light water reactor spent nuclear fuel (SNF) to molten salt reactor halide salt fuel;

[78] FIG. 12 schematically illustrates a molten salt immersed hydrohalogenation process;

[79] FIG. 13 schematically illustrates an exemplary implementation of a reaction vessel tailored for process function;

[80] FIG. 14 schematically illustrates a hydrofluorination process block diagram;

[81] FIG. 15 schematically illustrates a process where pentachloride, UCl_5, is produced;

[82] FIG. 16 schematically illustrates a fluidized bed reactor that may be configured to operate in either batch or continuous operation;

[83] FIG. 17 schematically illustrates a decladding process;

[84] FIG. 18 schematically illustrates a process for hydrohalogenation;

[85] FIG. 19 schematically illustrates a process of halogenation, including molten salt hydrohalogenation and off-gas processing;

[86] FIG. 20 schematically illustrates an off-gas processing section comprising a condenser;

[87] FIG. 21 schematically illustrates a condenser/cryogenic off-gas process and includes circulation of post-cryogenic process recirculable gases;

[88] FIG. 22 schematically illustrates an alternate implementation of a condenser/cryogenic off-gas process and includes circulation of post-cryogenic process recirculable gases;

[89] FIG. 23 schematically illustrates an alternate implementation of a condenser/cryogenic off-gas process and includes circulation of post-cryogenic process recirculable gases;

[90] FIG. 24 schematically illustrates another alternate implementation of a condenser/cryogenic off-gas process and includes circulation of post-cryogenic process recirculable gases;

[91] FIG. 25 schematically illustrates an alternate implementation of an off-gas that shows circulation of dry off-gases in counter-flow to a condenser and then to a molten salt immersion reaction vessel;

[92] FIG. 26 schematically illustrates the relationship between major process sections discussed above and also conveys an overall physical “U-path” of material flow;

[93] FIG. 27 schematically illustrates an overview of a halogenation and gas processing material/process; and

[94] FIG. 28 schematically illustrates an exemplary implementation of dismantling and decladding fuel rods.

DETAILED DESCRIPTION

[95] Some implementations of the present disclosure will now be described more fully hereinafter with reference to the accompanying drawings, in which some, but not all variations of the disclosure are shown. Indeed, variations of the disclosure may be embodied in many different forms and should not be construed as limited to the examples set forth herein; rather, these are provided so that this disclosure will be thorough and complete and will fully convey the scope of the disclosure to those skilled in the art.

[96] As used herein, “and/or” means any one or more of the items in the list joined by “and/or.” As an example, “x and/or” means an element of the three-element set, e.g., [(x), (y), (x, y)]. Additionally, as used herein, the terms “exemplary” and “example” mean in context as serving as a non-limiting example, instance, illustration, or circumstance. Moreover, as used herein, the term “for example,” or, “e.g.,” introduces a list of one or more non-limiting examples, instances, illustrations, or circumstances.

[97] Exemplary implementations in accordance with the present disclosure are described with reference to systems and/or methods, such as in the context of processing spent nuclear fuel. Further, for example, reference is made herein to values of or relationships between components, parameters, properties, variables or the like. These and other similar values or relationships are absolute or approximate to account for variations that may occur, such as those due to engineering tolerances or the like. Like reference numerals refer to like elements throughout.

[98] Of note, the disclosures set forth in *Conversion of Oxide to Metal or Chloride*, by Sakamura, et al., Organization Central Research Institute of the Electric Power Industry (CRIEPI), Japan, and *Effect of Melt Composition on the Reaction of Uranium Dioxide with Hydrogen Chloride in Molten Alkali Chlorides*, by Volkovich, et al., Ural State Technical University, Russia and the entirety of both of the foregoing documents are incorporated herein by reference.

[99] Further incorporated by reference in their entirety are the following documents: *Processing of Used Nuclear Fuel*, World Nuclear Association, (updated June 2018), https://world-nuclear.org/information-library/nuclear-fuel-cycle/fuel-recycling/processing-of-used-nuclear-fuel.aspx); *Recycling Nuclear Fuel: The French Do It, Why Can't Oui?*, December 28, 2007, The Heritage Foundation (https://www.heritage.org/environment/commentary/recycling-nuclear-fuel-the-french-do-it-why-cant-oui; *Recycling Process of Defective Aged Uranium Dioxide Pellets*, Fatah Mernache, et al, published online Aug.12 , 2015, Journal of Nuclear Science and Technology, Vol 53, Issue 6; *Engineering Design of a Voloxidizer with a Double Reactor for the Hull Separation of Spent Nuclear Fuel Rods*, Young-Hwan Kim, et al Korea Atomic Energy Research Institute, Science and Technology of Nuclear Installations, Vol 2017, Article ID 985; *Oxidation of UO2 Fuel Pellets in Air At 503 and 543 K Studied Using X Ray Photoelectron Spectroscopy and X Ray Diffraction*, P.A. Tempest et al, Journal of Nuclear Materials Feb 1988; *The High Burnup Structure in Nuclear Fuel*, Vincenzo V. Rondinella et al, European Commission, Joint Research Centre, Institute for Transuranium Elements Germany, Materials Today, Dec 2010, Vol 13, No 12; *Uranium Tetrafluoride*, IBILABS International Bio-Analytical Industries, Inc. August 7, 2016; *Uranium Tetrafluoride*, Wikipedia Ref Journal of the American Chemical Society, 1969; *Hydrofluoric Acid Corrosion Study of High-Alloy Materials*, P.E. Osborne et al, ORNL, UT Battelle, LLC for DOE, August 2002; and "Inconel 600", Spec sheet FSA, Shanghai Fengqu Superalloy Co, Ltd. March 13, 2019.

[100] Additionally, incorporated by reference in their entirety are the following patent documents: GB 803258; GB 1171257; GB 2536857; JP 11231091; KR 20060035917A; KR 20090089091A; KR 2009010 109237A; KR 20090109238A; KR 20110034347A; US

2013/0266112A1; WO 2017/158335A1; US 2011/0286570A1; US 9767926; US 4062923; and US 6251310. Further incorporated by reference in its entirety are the documents: "Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors – I: DUPIC Fuel Fabrication Cost, by Hangbox Choi, Won Li Ko, and Myung Seung Yang, Korea Atomic Energy Research Institute, *Nuclear Technology*, Vol. 134, May 2001; Proceedings of the 16th International Conference on Nuclear Engineering ICONE 16, 2006/2008 "Second Generation Experimental Equipment Design to Support Veloxidation Testing At INL"; World Journal of Nuclear Science and Technology , 2015 "Reduction Kinetics of Uranium Trioxide to Uranium Dioxide Using Hydrogen."

[101] Briefly, FIGs. 1 and 7 illustrate an exemplary implementation of a system including one or more processes for thermal molten salt reactor (TMSR) fuel-salt preparation, and FIG. 8 illustrates another exemplary implementation of the present disclosure, namely, a system including one or more processes for fast molten salt reactor (FMSR) fuel-salt preparation.

Methods and Systems for Calciner Fluoride Fuel Salt Preparation ("Option A")

[102] FIGs. 1 and 7 illustrate an exemplary implementation of a system, generally 100, according to one exemplary implementation of the present disclosure for calciner fluoride fuel salt preparation in the production of thermal molten salt reactor fuel salt. An externally heated and cooled calciner apparatus provides for a continuous process for converting spent fuel UO_2 pellets (which have been previously removed from the fuel cladding) at elevated temperatures to UF_4 crystals/powder using a rotating cylinder through which the pellets advance, and using a counter-flow of oxygen (via the center of an axial support tube of an integrated helical auger) for oxidizing, then reducing in a concurrent flow of hydrogen gas, then fluorinating in a concurrent flow of HF gas (the gasses being introduced via the central pipes contained in the axis of the calciner.) The calciner apparatus has sealing mechanisms at both ends to prevent any of the gases or particulates from entering the surrounding facility's atmosphere. The pellets are loaded through these sealing mechanisms at one end of the calciner apparatus, and the UF_4 continuously exits through such sealing mechanisms at the other end of the calciner apparatus.

[103] In some aspects, for example, the system 100 process begins with spent fuel pellets being recovered from fuel rod cladding (not shown) and fed into a rotating calciner, generally, 106. In an exemplary implementation, two calciners, one for each of two lines, could be used. As shown in FIG. 1, an axial cross-section of a calciner 106 depicts the construction thereof and the gas flow there-through, shown by arrows 112. The direction of process flow through the calciner, is from left to right as indicated by arrows, in the axial-cross section. Sealed entries and exits to and from the calciner prevent gases escaping outside the system, while allowing entry of spent fuel pellets and exit of fluoride salt. (See U.S. Patent No. 7,824,640, to Pitts, incorporated herein in its entirety by reference). All gases used and recycled in the calciner will be filtered to exclude unwanted elements and particles from exiting with the product. (See U.S. Patent No. 4,666,684, to Pitts, incorporated herein in its entirety by reference). Proceeding through the calciner 106, there are three process subdivisions designated by three radial cross-sections, or zones, generally 106A, 106B, and 106C, (FIG. 1) showing the particular gas flow of each section and desired product.

[104] Excess gases leave the calciner 106 by negative pressure to external filters (not shown). The externally-heated calciner rotates slowly, heating pellets to approximately 500°C for a period of time, which in one non-limiting example could be approximately 1 to 3 hours. Section 106A includes a fixed integral helical auger, the direction of rotation being indicated, as viewed in the direction of gas flow from right to left in FIG. 1.

[105] Calciner 106 dimensions, in one non-limiting example, could be approximately 15 to 30 inches in outside diameter, and axial section A could be approximately 10 to 20 feet in length. Axial sections 106B and 106C, in one non-limiting example, could be approximately 5 to 10 feet in length each. Sensors, which in some non-limiting examples may be embedded or attached wireless micro-sensors, generally 114, are shown in the calciner casing 116 and serve to monitor process parameters such as temperatures, pressures, material and added constituents flow rates, radiation, gases, and/or other measurable process details.

[106] One center conduit, or pipe, 118 extends the entire length of the calciner 106, which has a plug in the mid length of the pipe to prevent the mixing of the oxygen and hydrogen gasses. Oxygen (an oxidizing agent) enters at the left (as shown in FIG. 1), as pellets enter the calciner 106, and the oxygen exits from the pipe 118 into the calciner interior 120, starting the oxidation of pellets. Curved arrows 112 on the axial section 106A indicate oxygen flow from the pipe 118 to the calciner interior 120. Spent fuel pellets, generally 124, are indicated in section 106A at the helical auger 126 as are also mixing vanes 128. During this part of the process in calciner 106, UO_2 (uranium dioxide) spent fuel pellets are oxidized to various oxides of uranium, which causes the pellets to disintegrate because of expansion during oxidation. All other constituents of spent fuel are contained and oxidized in this section.

[107] A smaller center pipe 130 enters from the right end of calciner 106 and does not penetrate the full length of the calciner, but instead terminates at the start of axial section B.

A baffle 131 will be used to reduce the mixing of the oxidizing gas and the reducing gas, at the appropriate spot axially, in the calciner, but will still allow advancement of the product through the calciner. (See U.S. Patent No. 3,969,477, incorporated herein in its entirety by reference). One non-limiting exemplary location of baffle 131 is shown in FIG. 1. Baffle 131 is between the oxygen flow and the hydrogen gas flow in the calciner 106. The center pipe 130 supplies hydrogen gas for the second part of the process that takes place in the calciner 106, as shown by curved arrows 112 indicating outflow into the calciner main volume. The hydrogen gas is a reducing agent and flows from the center pipe 130 into the calciner interior 120, in section 106B. During this part of the process, various oxides of uranium are reduced to UO_2, and the consistency of the spent fuel pellets has been changed from a generally pellet form to coarse powder, shown in section 106B at the helical auger 126 and mixing vanes 128. Virtually the only effluent is water vapor, which is condensed during filtration of the recirculating gasses. All other constituents of spent fuel are contained and reduced during this part of the process.

[108] The final process converts UO_2 to UF_4 (uranium tetrafluoride). Hydrogen gas continues to flow through the smaller center pipe exiting into the calciner section 106B, as described previously; then, HF (hydrogen fluoride) gas enters into the larger annular pipe 118 at the right end of the calciner 106 as shown in FIG. 1, and exits into the main interior, or, body, 120 of the calciner 106 at the beginning of axial section 106C, with arrows 132 indicating direction of flow. During this part of the process, uranium and virtually all other constituents in the spent fuel, fission products, rare earths, and actinides are all substantially fluoridated. The resultant coarse powder product is shown in cross-section 106C at the helical auger and mixing vanes 128.

[109] Design and construction of the calciner apparatus 106 may include any suitable manufacturing techniques, including without limitation, application of 3D printing in order to use heat and corrosion resistant materials to create a durable internal design of calciner 106.

[110] Calciner 106 includes instruments and sensors for the measurement of pressure, temperature, gas concentration, gas flow, and material flow, which can be accomplished by many, perhaps hundreds, of wireless imbedded micro sensors 114, which are monitored in real-time by computer systems and artificial intelligence applications to maintain safety of operation and to provide continuous improvement of the process. The sensors 114 may be built into the calciner apparatus 106 during the 3D printing process.

[111] The calciner apparatus 106 keeps radioactive particles contained to prevent contamination of the surrounding facility, and calciner apparatus 106 generally produces only relatively small volumes of condensed liquid waste water, which will require specialized disposal. Operation of calciner apparatus 106 is more easily automated for operation on a 24/7 basis and is potentially less-expensive to operate over its lifetime than other types of processing. The design of the process using calciner apparatus 106 is scalable for increased capacity, as well as lending itself to be standardized for replication, so that multiple units can be used for backup purposes and/or to increase facility capacity.

[112] The conversion gases used in calciner apparatus 106 are carried by inert gases such as helium or argon, which are recycled. Water vapor generated during processing is condensed and removed from the process on a continuous basis. Gases exit the calciner apparatus 106 at each of the sealed ends to the filtering and replenishment equipment. The recirculated gases are filtered to remove elements not desired in the end product.

[113] During the first stage of this process, as shown in section 106A of FIG.1, the pellets 124 are exposed to a counter-flow of oxidizing gas, shown by arrow 125, such as oxygen, to covert the UO2 to various combinations of higher oxides of uranium, which increases the volume of the pellets up to 30%, potentially causing them to fracture. As shown in the section 106A, the motion of the rotating cylinder 138, integral helical auger 128 blades, with small shelves, or ledges, 140 to lift the pellets 124, provides friction between the pellets, and small impact forces experienced by the pellets hasten the oxidation process, which itself expands and fragments the pellets 124 further. This ultimately results in powdered oxides of uranium. The diameter of the calciner 106, in one exemplary implementation, could be in the range of approximately 15-30 inches.

[114] The second stage 106B of the calciner 106 process is to the right of a closure 127 in conduit 118 (FIG. 1) and exposes the oxide powders of uranium to a flow of reducing gas such as hydrogen, converting the various uranium oxide powders to UO_2. FIG. 1 shows the designs of section 106B, helical auger 128, and the axial gas supply channel 130.

[115] The third stage 106C of the calciner 106 process shown in section 106C exposes the UO_2 powder to fluoridizing gaseous HF, which produces UF_4 in a crystalline/powder form for use in lithium fluoride molten salt-based reactors, for example. Section 106C shows the auger 128 and central path of the HF gas. Such H_2 and HF gases then exit conduit 119 to filtering, which includes condensing and removing of the water vapor formed during the reduction and fluorination operations.

[116] The UF_4 exits the process in a manner which prevents leaking of gases to the atmosphere. A mechanism will be provided to seal the end of the calciner, so that the gases generated will be contained, and the product will exit cooled and ready for the next operation.

The product is sampled, tested, and certified for shipment. The UF_4 is automatically placed in containers, which are automatically sealed and cooled, and then stored for delivery to the customer. (In order to provide more uniform particle sizes than can perhaps be produced in the calciner 106, as the product exits the calciner 106, a subsequent milling operation for milling to powder to desired specifications may be used.) The third stage of the process shown in section 106C exposes the UO_2 powder to fluoridizing gaseous HF, which produces UF_4 in a crystalline/powder form for use in lithium fluoride salt-based reactors, for example. Section 106C shows the auger 128 and central path of the HF gas.

[117] In an exemplary implementation of the present disclosure, a method is illustrated in FIG. 7 for producing fuel for a thermal molten salt reactor, the implementation of the method including:

a. providing fuel assemblies containing an array of fuel tubes are aligned horizontally on the rod puller disassembly table 270 (FIG. 9), and fuel pellets are removed from tubes;

b. processing the spent fuel pellets and fuel pieces into a fluoride salt by ultimate oxidation, reduction and fluorination of uranium and its associated fuel constituents; and

c. filtering (including condensing and removing) the water vapor formed during the reduction and fluorination operations.

Another exemplary implementation of such method could include, if desired and as shown in FIG. 7, fluorinating the U235-enriched granular spent fuel salt or plutonium in a calciner rotary kiln or fluidized bed, and if additionally desired, enriching the granular spent fuel salt with U235.

[118] In an exemplary implementation, because both the reduction of the oxides of uranium to uranium dioxide and the conversion of uranium dioxide to uranium tetrafluoride are exothermic, the calciner includes both external heating and cooling apparatus (not shown) over most of its length.

[119] In an exemplary implementation, the temperature of conversion of uranium dioxide to uranium tetrafluoride in HF gas is to be maintained above 400 deg C during and after the conversion is completed, to prevent the undesired formation of volatile uranium hexafluoride, which will occur if it is cooled below 400 deg C in the presence of HF gas. Therefore, the uranium tetrafluoride must exit through the sealed end of the calciner above 400 deg C and then cooled to ambient temperature. This requires a counter flow of argon in the exit sealing mechanism of the calciner as cooling proceeds. Toward this end, the sealing and transfer mechanism for the fuel product is to be configured with sufficient cooling capacity. The calciner is configured to reduce the likelihood of the oxygen and hydrogen used in processing from being too close together in the oxidation and reduction steps in the calciner. Although at least one baffle 131 is used, it may be desirable to use multiple baffles, with the introduction of positive pressure inert gasses such as argon, between them, to prevent the mixing of oxygen and hydrogen during the process. Such inert gas can be introduced into the calciner through a pipe (not shown) placed axially in the auger 126, extending from the entrance end of the calciner to the baffle area.

[120] In exemplary implementation, Option A may include, if desired, the spent nuclear fuel being generally permanently stored, then processed into spent fuel salt, and the spent fuel salt used in a thermal molten salt reactor, all on a single site having a secured perimeter.

[121] Non-limiting example approximate temperatures, times, gas concentrations, materials used to construct the apparatus, and other parameters which are expected to be used are shown in the drawings.

Methods and Systems for Chloride Fuel Salt Preparation ("Option B")

[122] FIGs. 2-7 and 9 illustrate an exemplary implementation of a system, generally 200, or portions thereof, according to one example of the present disclosure for chloride fuel salt preparation in the production of fast molten salt reactor fuel salt.

[123] The process 200 begins after the spent fuel pellets 124 recovered from cladding in a manner as discussed above, being fed into a ball mill 202 (FIG. 9), and pulverized to a granular, preferably powder, form. Gases are recovered from the initial disassembly, from the ball mill 202, and from one or more enclosed conveyors (not shown), routing granulated spent fuel to the (FIG. 2) oxide reduction tanks 210. Noble gases Krypton and Xenon may be removed for storage or other uses.

[124] The oxide reduction tanks are the first tanks in line of the process 200 to treat granular/pulverized spent nuclear fuel. Spent fuel is reduced using a strong reducing agent, preferably a chloride-containing reducing agent, such as anhydrous hydrogen chloride (AHCl) addition through a tank sparger 212 at the bottom of the tank 210. A small excess of chloride with molten chloride fuel salt ensures enough free chloride to produce chloride salt fuel. The process produces water vapor and hydrogen which are continuously removed by blower extraction and condensation, and glow plugs (not shown) ensure hydrogen and oxygen gases are burned to water product. This process completes the goal of removing oxygen from all oxides in the salt fuel. Automated and dip sampling configuration, and density probes, while provided, are not shown. Gases are collected into a fluidized bed (not shown) or ion exchange column (not shown) for chlorination and recycling back into the main process. Raw granulated spent fuel is routed from the ball mills 202 by the enclosed conveyor to parallel oxide reduction tanks 210 containing molten salt. Granulated spent fuel is conveyed in a closed system, to the oxide reduction tank hopper 216.

[125] A tank 220 containing molten chloride salt maintained, in one non-limiting example, at approximately (30-50) degrees C (80-120 degrees F) above the melting point of the halide salt (molten alkali or alkali earth chloride) melting point estimated to be 500 C (930 F). The melting point of the molten salt is variable with the amount and consistency of alkali and alkali-earth chlorides, and with the amount of spent fuel added to the mix. Nominal density of spent fuel salt chloride is expected to be 3.0 g/cc, depending on concentration (mol%). It is anticipated salt fuel for the fast molten salt reactor will require significant enrichment. This enrichment will be performed with addition of U235, Pu239, or MOX fuel. At an estimated beginning 30 mol% uranium chloride and plutonium-chloride, the balance being fission product chlorides and actinide chlorides (5-10) mol%, the remaining mix contains free molten salt at (60-65) mole%.

[126] FIG. 3 shows the fuel salt mixing and adjustment tank 220, second in line of an exemplary implementation of the process, receives fuel salt in a hopper 223 from the oxide reduction tank 210. Both tanks 210, 220 (FIG. 2 and 3) have automated sampling, and pump recirculation distribution headers (not shown) internal to the tanks. Tanks 210, 220 (FIG. 2 and 3) are sized and configured to prevent a criticality (critical-safe) in the tank as pulverized spent fuel is added and enriched with U235, Pu239, or MOX fuel, to high assay low enriched uranium (HA-LEU) at less than approximately 20% enrichment. Both tanks 210, 220 have the capability to receive salt, spent fuel, or enrichments; however, tank 220 will normally receive only salt replenishment as needed. Fertile-thorium oxide may be added to the uranium/plutonium oxide reduction tank 210 and chlorinated to thorium chloride salt. The enrichment is necessitated in fueling and operation of a fast molten salt reactor. Tanks 210 and 220, in one non-limiting example, have approximate estimated dimensions of 10 feet in height by 16 feet front to back and 10 inches wide and is capable of processing approximately 1000 gallons. Tanks 220, in one exemplary implementation, are constructed integrally with an outside tank (not shown) having leak detection between the inside and outside tanks. Outside tank dimensions allow for insulation, multiple electric heater access points, recessed instrument enclosures, and accesses to each.

[127] The tanks 210, 220 are instrumented with dip sample points (not shown) for automatic and/or manual sampling and analysis. This capability confirms independent on-line sampling that a receiving-mixing tank’s contents are fully chlorinated to the extent possible (uranium, fission products, lanthanides, and actinides), i.e., substantially the entire inventory of spent nuclear fuel nuclides. A density probe 221and manual liquid density measurement generated therefrom confirm whether the spent fuel salt density is at the expected density nominally (3.0-4.0) g/cm^3 (kg/m^3), molten alkali or alkali earth chloride density, no other content, is approximately (1.6 g/cm^3). The contents of the oxide reduction tank 210 (FIG. 2), and mixing and adjustment tank 220 will be processed further when sample analyses are confirmed. It is estimated reaction processing time is in an exemplary implementation 8 hours, including enrichment and sample confirmation, for the oxide reduction tank 210, and 4 hours for the mixing and adjustment tank 220. Full range gamma and neutron nuclear instruments, generally 224, provide continuous monitoring, trending, and alarming (counts/second) and rate of change. In one implementation, oxide reduction tank 210 size and configuration require four equally spaced instruments over the height and depth of each tank. A blower and chiller 226 combination removes water from tank 210. An anhydrous hydrogen chloride cylinder and compressor, generally 228, supply sparger arrangement 212. Salt mixers (e.g., screw-type mixers) 222 set at different depths, and front to back of the tank, ensure sufficient mixing of each tank. Additionally, FIG. 2 is an exemplary implementation wherein tank screw pump 218 is shown. Tank screw pump 218 is connected to tank 210 via conduit 210A, and an inlet valve 218A is provided proximate the inlet of pump, and an outlet valve 218B is provided proximate the outlet of pump 218. A conduit 218C connects valve 218B to a discharge valve 219 and to a tank pump recirculation isolation valve 229 connected to molten fuel salt mixing tank 220.

[128] In an exemplary implementation shown in FIG. 3, a tank screw pump 218 is connected to tank 220 via conduit 220A, and an inlet valve 218A is provided proximate the inlet of pump, and an outlet valve 218B is provided proximate the outlet of pump 218. A conduit 218C connects valve 218B to a tank header valve 217 and to a tank pump recirculation isolation valve 225.

[129] In FIG.4, a representation of tanks 220 containing molten salt spent fuel are shown in a plan view in a side-by-side relationship, and, as in the case of all the drawings herein, are not shown to scale. This view represents the general size and configuration for both the oxide reduction tanks 210 (side by side) and mixing and adjustment tanks 220 (side by side). More specifically, FIG. 4 shows an implementation wherein six tanks 220 are shown. Accompanying apparatus and equipment and configurations used in connection with the tanks 220, are not shown.

[130] In the basic process flow (FIG. 9), it could be considered that the first oxide reduction tank 210 would pump out to the first mixing and adjustment tank, the second oxide tank, the second mixing tank, and continue this sequence until all oxide reduction tanks have pumped out to their respective mixing and adjustment tanks. The tanks, FIG. 4, also include the tank pump discharge header and nozzles, which are only located on the mixing and adjustment tank 220 pump out header. Spaced between tanks are encased boron slabs, or, dividers, or encasements, 230. Boron encasements 230 positioned between all fuel salt tanks 220 prevent nuclear criticality communication between the array of oxide reduction tanks in close proximity, and between the array of mixing and adjustment tanks in close proximity. Each group is considered herein as one subcritical assembly group, generally 266. Boron dividers 230 are backup defense in depth against possible criticality.

[131] In an exemplary implementation, equipment is selected for durability and reliability. Two channels of electric “jacketed heaters” (not shown) are fitted to piping, valves and pumps ensure salt fuel in piping and equipment is of a high enough temperature to remain liquid and will flow. The heater channels are monitored, alarmed, and component failure identified if such a failure occurs. If sections of piping are allowed to cool where molten salt is solidified, heaters can be activated to re-melt the fuel salt. Instrumentation and automated functions are fully alarmed and continuously communicated to a control center. Diagnostic protocols help operators identify system interruptions or points requiring repair. All components on tanks and transfer piping are preferably accessible and capable of remote repair after steps are taken to isolate failed components from the system. Multiple independent receiving-mixing tanks and transfer equipment ensures a continuous supply of fuel salt in operation, including in the event of a system failure.

[132] In an exemplary implementation, fuel salt preparation is begun with introduction of chloride salts of alkali metals or alkaline earth metals (e.g., $NaCl$, KCl, $CsCl$, $SrCl_2$, $MgCl_2$, $CaCl_2$), typically in crystalline form, and usually a mixture of two or more salts to a tank. Heaters (e.g., electrical heating elements) 231 are energized to melt the salt to molten state and maintain temperature well above melting point. Pulverized-granulated spent nuclear fuel is taken from the ball mill 202 and carried by enclosed conveyor to the tank hopper 223 and deposited via hopper isolation valve 227 into the oxide reduction tank, and open isolation valve 227 (FIG. 2). Spent fuel addition, regulated by limiting size and speed of the enclosed conveyor, ensures tank temperature is maintained within predetermined specifications and sufficient mixing and reaction of tank contents occurs. All mixers and the tank pump are turned on to initiate mixing, recirculation by pumping, and consistency of fuel salt in the tank and pump discharge lines. In an exemplary implementation, tank size and screw pump capacity are regulated to allow 4-6 hours for mixing and sampling, density recording and analysis before a tank is ready to be discharged.

[133] Figures 5 and 6 show an exemplary implementation in plan and elevational views of a molten fuel salt collection tray, generally 240, consisting of the tray cover 254, fuel salt collection molds 256 atop heating and cooling elements 242, in a generally checkerboard pattern of heating elements 234A and cooling elements 234B. Insulation between elements along the sides and bottom of the tray prevents solidification during pouring and minimizes cooling time after solidification. More specifically, FIG. 5 is a plan view of salt mold cooling tray 240 with the tray cover and cooling molds removed.

[134] In an exemplary implementation, salt mold cooling trays 240 (FIG. 9) are positioned and held in a stacked array of 8-10 trays, with spacing between the trays being sufficient to allow for removal of the cooling molds and cover as one assembly. Stacked arrays may be each contained within mobile cabinets (not shown) and tracked together by a revolving drive (not shown) which moves one stacked and cooled array group, to the ball mill feed table 250. At the table 250, the one-piece molds of each tray, in a particular group, are removed and upended or overturned to deposit solid “bars” of fuel salt. Each tray’s mold is removed and returned to its position before the next mold is removed. After an entire array group has been emptied, it is returned in turn to be refilled with molten salt. Tray molds can be a non-stick surface, with salt fuel contraction during cooling, thereby facilitating solid salt fuel removal. The metal molds may be connected side-to-side and laterally supported to ensure tray strength and versatility. Solid salt “bars” are gathered to the side of the turning table and are generally organized lengthwise on a moving conveyor and fed into 2-3 coarse ball mills 202A (FIG. 9). Product fuel salt from the ball mill is further conveyed to a fine mill, such as a Fitz mill 252 for sizing, sampling, certification, and packaging for protection against environmental conditions. More specifically, FIG. 6 illustrates a salt mold cooling tray front view, including top cover 254, cooling molds 256, heating and cooling elements 234A, 234B, which, in one implementation, could be coils.

[135] In an alternate implementation, molten salt fuel may be stored as a contiguous solid in canisters and subcritical arrays. This process involves preparation of chloride fuel salt in the aforementioned receiving and mixing tanks 220, sampling and certification of tanks, and transfer by screw pump to a “critical safe” steel canister (not shown), set aside for cooling. Canisters are transported and stored, in “critical safe” arrays. Facilities using “solid salt” canisters are equipped to remotely handle and inductively heat each canister to form liquid fuel salt for addition to their molten salt reactors.

[136] In FIG. 4, molten salt spent fuel receiving and mixing tanks 220 are shown in a side-by-side representative top view, but not to scale. More specifically, FIG. 4 shows an implementation wherein six receiving-mixing tanks 220 are side-by-side, including a tank pump discharge header, generally 260, and encased boron encasements 230 (sized for inner tank side dimension area). Boron encasements 230 provide structural stability and protection from damage and are positioned between fuel salt tanks 220 to prevent nuclear criticality communication between tanks in a given array of tanks in close proximity, such array being considered herein as one subcritical assembly group, generally 266. Boron dividers 230 are backup defense, in depth, against possible criticality.

[137] As shown in an exemplary implementation in FIG. 2, the oxide reduction tank is the first tank in the process, wherein a system and process are illustrated which reduces uranium and plutonium oxides to chlorides. Fertile-thorium oxide may be added to the uranium/plutonium oxide reduction tank 210 and chlorinated to thorium chloride salt. After this process, the contents of the reduction tank 210 are pumped over to the mixing and adjustment tank. In one implementation, an isolation valve is provided on the loading hopper. In an exemplary implementation wherein only chloride salts are desired, the salts are prepared in the oxide reduction tank, and, then pulverized, granulated spent fuel is added to the oxide reduction tank, the temperature increase is noted, and oxide reduction is begun by the sparging action of anhydrous hydrogen chloride. Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used. After an allowance of time necessary for mixing and water and hydrogen gas removal, toward complete reduction, the contents are pumped to the mixing and adjustment tank for final analysis, certification, and then pumped over to cooling trays. The teaching of the present disclosure includes reducing all oxides, removing oxygen entirely and preventing production of other oxides, ensures an authentic chloride salt fuel, when oxygen is removed from the fuel salt. The result is that substantially the only byproduct from this implementation of the present disclosure is water, which is collected for sampling and released.

[138] In an exemplary implementation of the present disclosure, a method is illustrated in FIG. 8 for producing fuel for a fast molten salt reactor, the implementation of the method including:

a. providing fuel assemblies, removing fuel pellets containing uranium and all spent fuel constituents, from the fuel assemblies;

b. granulating the fuel pellets in a semi-voided atmosphere using a ball mill, roller mill, or chopping mill, for process feed to the chlorination process;

c. processing the granular spent fuel salt into chloride salt by ultimate reduction and chlorination of the uranium and associated fuel constituents chloride salt solution, by anhydrous hydrogen chloride (AHCl);

d. enriching the granular spent fuel salt with U235, Pu239, or MOX;

e. chlorinating the enriched granular spent fuel salt to yield molten chloride salt fuel using AHCl halide salt reduction;

f. analyzing, adjusting, and certifying the molten chloride salt fuel for end use in a molten salt reactor;

g. pumping the molten chloride salt fuel to stacked arrays of cooling trays or canisters and cooling the molten chloride salt fuel to yield solid salt fuel bars, sticks, or canister solid form;

h. milling the solidified molten chloride salt fuel to predetermined specifications for the fast molten salt reactor; and

i. optionally, fertile-thorium oxide may be added to the uranium/plutonium oxide reduction tank 210 and chlorinated to thorium chloride salt.

[139] In exemplary implementations, Option B may include, if desired, the spent nuclear fuel being generally permanently stored, then processed into spent fuel salt, and the spent fuel salt used in a fast molten salt reactor, all on a single site having a secured perimeter.

[140] Non-limiting example approximate temperatures, times, gas concentrations, materials used to construct the apparatus, and other parameters which are expected to be used are shown in the drawings.

[141] FIG. 10 schematically illustrates a site on which components of a system 1000 according to an example implementation of the present disclosure may be located. The system 1000 may include a site 1002 within a secured perimeter 1004, and a limited-access facility 1006 on the site 1002. As described above, the system 1000 may include a number of components 1008, and at least some of these components 1008 of the system 1000 may be located within the limited-access facility 1006. The system 1000 may also include a spent nuclear fuel storage facility 1010 located on the site, and a molten salt reactor 1012 located on the site 1002.

[142] In other exemplary implementations of producing chloride salt fuel using system 200, additional reduction may be achieved by addition of metal hydrides. Generally, in exemplary implementations of the present disclosure the processes follow that described above for Option B processes for conversion of powdered spent nuclear fuel, used fuel, to molten salt reactor salt fuel begins with a starting base bath of molten halide salt, or a mixture of halide salts as the molten medium to dissolve all spent fuel constituents. Particular acids of the halides e.g., hydrogen – fluoride, chloride, bromide, or iodide, may be used for halogenation of uranium, plutonium, fission products and actinides by “fluorination,” “chlorination,” “bromination,” or “iodination” of powdered spent nuclear fuel, converting it to “salt fuel.” Generally, halide salt e.g., sodium chloride, potassium chloride, cesium chloride, or strontium chloride, and anhydrous hydrogen chloride are used for spent fuel conversion to chloride salt fuel. This is necessary to initialize and maintain a continuity of salt fuel physical and nuclear characteristics.

[143] As discussed above, the oxide reduction tanks are the first tanks in line of the process to treat pulverized/powdered spent nuclear fuel. Spent fuel is reduced using a strong reducing agent, preferably a chloride containing reducing agent, such as anhydrous hydrogen chloride (AHCl) addition through a tank sparger 212 at the bottom of the tank 210. Additional reduction may be achieved by addition of metal hydrides. A small excess of chloride with molten chloride fuel salt ensures enough free chloride to produce chloride salt fuel. The reduction of uranium oxide, plutonium oxide and substantially all spent nuclear fuel constituents produces hydrogen and oxygen forming water vapor and are continuously removed by blower extraction and condensation. Generally continuous removal of water during oxide reduction is essential to maintain an acidic balance continuously, during spent fuel reduction. And, in another exemplary implementation, numerous glow plugs (not shown) ensure hydrogen gas and oxygen are burned to water product, are placed near the top of the tank interior, have redundant power supplies and glow plug failure monitoring. All ancillary equipment for production of water vapor from hydrogen and oxygen, and removal of water from the tanks, is designed with significant margin in excess of the maximum expected process generation rate. This process completes the goal of removing oxygen from all oxides or hydroxides, in the salt fuel. Automated and dip sampling configuration, and density probes are also provided (not shown). Gases are collected into a fluidized bed or small chemical reactor (not shown) for chlorination and recycling back into the main process. Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used. Raw powdered spent fuel is routed from the ball mills 202 by the enclosed conveyor to parallel oxide reduction tanks 210 containing molten salt. Powdered spent fuel is conveyed in a closed system, to the oxide reduction tank hopper 216. Tank 220 containing molten chloride salt is maintained, in one non-limiting example, at approximately (30-50) degrees C (80-120 degrees F) above the melting point of the halide salt (molten alkali chloride) melting point estimated to be 600 C (1048 F). The melting point of the molten salt may be adjusted by the addition of zirconium chloride after complete removal of oxygen, and with the amount of spent fuel added to the mix. Fertile-thorium oxide may be added to the uranium/plutonium oxide reduction tank 210 and chlorinated to thorium chloride salt.

[144] An additional exemplary implementation of oxidation may be further achieved by the addition of metal hydrides e.g., aluminum hydride or stannane (tin hydride) to enhance fast molten salt reactor nuclear properties. Nominal density of spent fuel salt chloride is expected to be 3.0 g/cc, depending on its (Mole %) concentration. It is anticipated salt fuel for the fast molten salt reactor will initially require significant enrichment. This enrichment will be performed by the addition of U235, Pu239, or MOX fuel. At an estimated beginning (30 mole%) uranium chloride and plutonium-chloride, the balance being fission product chlorides and actinide chlorides (5-10) mole%, the remaining mix contains free molten salt at (60-65) mole%. Tanks 210, 220 are in exemplary implementations instrumented with dip sample points (not shown) for automatic and/or manual sampling and analysis. This capability confirms independent on-line sampling that a processing tank's contents are fully mixed and chlorinated to the maximum extent possible, substantially the entire inventory of spent nuclear fuel.

[145] Fast reactor salt fuel requires high neutron energy for fast fission to occur, and such energy is desired to be greater than the threshold for fast neutron energy, whereby neutrons retain enough energy after they are produced from fission to continue the process of fast fission. This is achieved by conversion of spent fuel to salt fuel of heavier mass elemental salt, whereby heavy mass elemental metals of potassium, zirconium, or zinc, for example, and halides of chlorine, bromine or iodine, form salt fuel effecting fast fission. Heavy mass elemental metal hydrides of zirconium, molybdenum, or tin, for example, form salt fuel effecting fast fission by reduction of spent fuel to salt fuel and oxidation of hydride heavy mass metals to salts, whereby neutrons retain energy well above fast neutron threshold energy after they are produced from fission to continue the process of fast fission.

[146] In an exemplary implementation of the present disclosure, a method is illustrated in FIG. 8 for producing fuel for a fast molten salt reactor, the implementation of the method including:

a. providing fuel assemblies, removing fuel pellets containing uranium and all spent fuel constituents, from the fuel assemblies;

b. reducing to powder, the fuel pellets in a semi-voided atmosphere using a ball mill, and fine mill, for process feed to the chlorination process;

c. processing the granular spent salt fuel into chloride salt by ultimate reduction and chlorination of the uranium and associated fuel constituents chloride salt solution, by anhydrous hydrogen chloride (AHCl); following the additions of AHCl, salt fuel reduction may be further enhanced by addition of metal hydrides;

d. enriching the spent salt fuel with U235, Pu239, or MOX;

e. chlorinating the enriched powdered spent salt fuel to yield molten chloride salt fuel using AHCl halide salt reduction;

f. analyzing, adjusting, and certifying the molten chloride salt fuel for end use in a molten salt reactor;

g. pumping the molten chloride salt fuel to stacked arrays of cooling trays or canisters and cooling the molten chloride salt fuel to yield solid salt fuel bars, sticks, or canister solid form;

h. milling the solidified molten chloride salt fuel to predetermined specifications for the fast molten salt reactor; and

i. optionally, fertile-thorium oxide may be added to the uranium/plutonium oxide reduction tank 210 and chlorinated to thorium chloride salt.

Methods and Systems for Fluoride Fuel Salt Preparation ("Option C")

[147] In another exemplary implementation of the present disclosure, generally, thermal molten salt reactor flouride salt fuel can be produced using the same equipment apparatus (shown in FIGs. 2-6, 8, and 9) as that used for fast molten salt reactor salt fuel in Option B discussed above. As discussed above, spent fuel pellets 124 are extracted from the cladding and passed them through the ball mill 202 (FIG. 9) and pulverized them into granular, or powder, form, where gases are recovered from the initial disassembly, from the ball mill 202, and from one or more enclosed conveyors (not shown). Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used. The powdered spent fuel is routed to the oxide reduction tanks 210 (FIG. 2). The spent fuel powder is ready to be processed to either thermal or fast reactor salt fuel. Using portions of the process and an implementation of system 200 discussed above, including the removal of water from the system 200, recovery of gases and particulates back into the process, proceed with reduction of spent fuel constituents, uranium oxide, fission products, and actinides, into molten salt, in a generally continuous manner.

[148] Thermal reactor salt fuel is prepared by addition of the powdered spent nuclear fuel to a molten salt bath of lithium and/or beryllium fluoride salts in an oxide reduction tank 210.

Quantities of fluoride molten salt contained in the oxide reduction tanks 210, powdered spent fuel, required enrichment, and anhydrous hydrogen fluoride are determined before beginning any additions. Calculations of quantities are determined for a specific end product Mole% of salt fuel in Mole% of molten salt. Partial additions of all reactants are performed with adequate time allowed for mixing and reactions, sampling and confirmation, before further additions. Fuel salt is thoroughly mixed before anhydrous hydrogen fluoride (AHF) is admitted through the tank sparger arrangement 212. Salt fuel properties for a thermal salt fuel preparation, are discussed below.

[149] More specifically, the process begins after the spent fuel pellets 124 recovered from cladding in a manner as discussed above, being fed into a ball mill and fine mill 202 (FIG. 9), and pulverized to a powder form. Gases are recovered from the initial disassembly, from the ball mill 202, and from one or more enclosed conveyors (not shown), routing granulated spent fuel to the (FIG. 2) oxide reduction tanks 210. The oxide reduction tanks which, include uranium/plutonium, are the first tanks in line of the process to treat pulverized/powdered spent nuclear fuel. Spent fuel is reduced using a strong reducing agent, a fluoride containing reducing agent, such as anhydrous hydrogen fluoride (AHF) addition through a tank sparger 212 at the bottom of the tank 210. Additional reduction may be achieved by addition of metal hydrides. A small excess of fluoride with molten fluoride fuel salt ensures enough free fluoride to produce fluoride salt fuel. The reduction of uranium oxide, plutonium and generally all spent nuclear fuel constituents produces hydrogen and oxygen forming water vapor and which are continuously removed by blower extraction and condensation. Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used.

[150] In another exemplary implementation, numerous glow plugs (not shown) ensure hydrogen gas and oxygen are burned to water product, are placed near the top of the tank interior, have redundant power supplies and glow plug failure monitoring. All ancillary equipment for production of water vapor from hydrogen and oxygen, and continuous removal of water from the tanks, is designed with significant margin in excess of the maximum expected process generation rate. This process completes the goal of removing oxygen from all oxides and hydroxides in the salt fuel. Automated and dip sampling configuration, and density probes, while provided, are not shown. Gases are collected into a fluidized bed or small chemical reactor (not shown) or ion exchange column (not shown) for chlorination and recycling back into the main process. Raw powdered spent fuel is routed from the ball mills 202 by the enclosed conveyor to parallel oxide reduction tanks 210 containing molten salt. Powdered spent fuel is conveyed in a closed system, to the oxide reduction tank hopper 216.

[151] A tank 220 containing molten fluoride salt maintained, in one non-limiting example, at approximately (30-50) degrees C (80-120 degrees F) above the melting point of the halide salt (molten alkali fluoride) melting point estimated to be 600 C (1048 F). The melting point of the molten salt may be adjusted by the addition of zirconium chloride after complete removal of oxygen, and with the amount of spent fuel added to the mix. An additional exemplary implementation of oxidation may be further achieved by the addition of metal hydrides e.g., beryllium hydride, or lithium hydride to enhance nuclear properties for a thermal molten salt reactor. Nominal density of spent fuel salt fluoride is expected to be 3.0 g/cc, depending on its (Mole %) concentration. It is anticipated salt fuel for the thermal molten salt reactor will initially require enrichment. This enrichment will be performed by the addition of U235, Pu239, or MOX fuel. At an estimated beginning (30 mole%) uranium fluoride and plutonium-fluoride, the balance being fission product fluorides lanthanide fluorides, and actinide fluorides (5-10) mole%, the remaining mix contains free molten salt at (60-65) mole%. Non-fuel fertile-thorium oxide, may be added to the uranium/plutonium oxide reduction tank 210 and fluorinated to thorium fluoride salt.

[152] FIG. 3 shows the fuel salt mixing and adjustment tank 220, second in line of an exemplary implementation of the process, receives salt fuel in a hopper 223 from the oxide reduction tank 210. Both tanks 210, 220 (FIG. 2 and 3) have automated sampling, and pump recirculation distribution headers (not shown) internal to the tanks. Tanks 210, 220 (FIG. 3) are sized and configured to maintain subcriticality (critical-safe) in the tank as powdered spent fuel is added and enriched with U235, Pu239, or MOX fuel, to high-assay low enriched uranium (HALEU) at less than 20% enrichment. Both tanks 210, 220 have the capability to receive salt, spent fuel, or enrichments; however, tank 220 will normally receive only salt replenishment as needed. The enrichment is necessitated in fueling and operation of a thermal molten salt reactor. Tanks 210 and 220, in one non-limiting example, have approximate estimated dimensions of 10 feet in height by 16 feet front to back and 10 inches wide and is capable of processing approximately 600 gallons to allow the remaining free volume (head-space) for processing gases. Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used. Tanks 220, in one exemplary implementation, are constructed integrally with an outside tank (not shown) having leak detection between the inside and outside tanks. Outside tank dimensions allow for insulation, multiple electric heater access points, recessed instrument enclosures, and accesses to each.

[153] The tanks 210, 220 are instrumented with dip sample points (not shown) for automatic and/or manual sampling and analysis. This capability confirms independent on-line sampling that a processing tank's contents are fully mixed and fluorinated to the extent possible, substantially the entire inventory of spent nuclear fuel. A density probe 221 and manual liquid density measurement generated therefrom confirm whether the spent fuel salt density is at the expected density nominally (3.0-4.0) g/cm^3 (kg/m^3), molten alkali fluoride density, is approximately (1.6 g/cm^3). The contents of the oxide reduction tank 210 (FIG. 2), and mixing and adjustment tank 220 will be processed further when sample analyses are confirmed. Estimated processing time is in an exemplary implementation 8 hours, including enrichment and sample confirmation, for one oxide reduction tank 210, and 4 hours for the mixing and adjustment tank 220. The oxide reduction tanks 210 and mixing and adjustment tanks 220 are paired, may be step-wise staggered; therefore, full use would mean 4-8 hours overlap time between the first oxide reduction tank and mixing and adjustment tank pair, and the second oxide reduction tank and mixing and adjustment tank pair. Full range gamma and neutron nuclear instruments, generally 224, provide continuous monitoring, trending, and alarming (counts/second) and rate of change. In one implementation, oxide reduction tank 210 size and configuration require four equally spaced instruments over the height and depth of each tank. A blower and chiller 226 combination removes water from tank 210. An anhydrous hydrogen fluoride cylinder and compressor, generally 228, supply in-tank sparger arrangement 212. Salt mixers 222 are set at alternate depths, and front to back of the tank, ensure sufficient mixing of each tank. Additionally, FIG. 2 is an exemplary implementation wherein a tank transfer screw pump 218 is shown. A medium to high-volume tank screw pump 218 is connected to tank 210 via conduit 210A, and an inlet valve 218A is provided proximate the inlet of pump, and an outlet valve 218B is provided proximate the outlet of pump 218. A conduit 218C connects valve 218B to a discharge valve 219 connected to molten fuel salt mixing and adjustment tank and to a tank pump recirculation isolation valve 229 for mixing oxide reduction tank contents.

[154] In an exemplary implementation shown in FIG. 3, a medium to high-volume tank screw pump 218 is connected to tank 220 via conduit 220A, and an inlet valve 218A is provided proximate the inlet of pump, and an outlet valve 218B is provided proximate the outlet of pump 218. A conduit 218C connects valve 218B to a tank header valve 217 mixing and adjustment tank pump out connection, and to a tank pump recirculation isolation valve 225.

[155] In FIG.4, a representation of tanks 220 containing molten salt spent fuel are shown in a plan view in a side-by-side relationship, and, as in the case of all the drawings herein, are not shown to scale. This view represents the general size and configuration for both the oxide reduction tanks 210 (side by side) and mixing and adjustment tanks 220 (side by side). More specifically, FIG. 4 shows an implementation wherein six tanks 220 are shown.

[156] Accompanying tank support systems, apparatus and equipment and configurations used in connection with the tanks 220, are not shown.

[157] In the basic process flow (FIG. 9), the first oxide reduction tank 210 would pump out to the first mixing and adjustment tank, the second oxide reduction tank to the second mixing and adjustment tank, and continue this sequence until all oxide reduction tanks have pumped out to their respective mixing and adjustment tanks. The tanks, FIG. 4, also include the tank pump discharge header and nozzles, which are only located on the mixing and adjustment tank 220 pump out header. Spaced between tanks are encased boron slabs, or, dividers, or encasements, 230. Boron encasements 230 positioned between all oxide reduction tanks 210 and mixing and adjustment tanks 220 prevent nuclear criticality when all tanks together act as one, and maintain adequate margin of sub-criticality communication between the array of oxide reduction tanks in close proximity, and between the array of mixing and adjustment tanks in close proximity. Each group is considered herein as one subcritical assembly group, generally 266. Boron dividers 230 are backup defense in depth against possible criticality.

[158] In an exemplary implementation, equipment is selected for durability and reliability. Two channels of electric “jacketed heaters” 231 (FIG. 2) are fitted to tanks, piping, valves and pumps ensure salt fuel in piping and equipment is of a high enough temperature to remain liquid and will flow in the event that one set of monitored heating elements fail. The heater channels are monitored, alarmed, and component failure identified if such a failure occurs. If sections of piping are allowed to cool where molten salt is solidified, heaters can be activated to re-melt the fuel salt. Instrumentation and automated functions are fully alarmed and continuously monitored and displayed at the control center. Diagnostic protocols identify and locate system failures and inform operator system status interruptions or points requiring repair. All components on tanks and transfer piping must be accessible and capable of remote repair after steps are taken to isolate failed components from the system. Multiple independent oxide reduction tanks and mixing and adjustment tanks and transfer equipment ensures a continuous supply of fuel salt in operation, including in the event of a system failure.

[159] In an exemplary implementation, salt fuel preparation is begun with introduction of fluoride salts of alkali and alkali earth metal fluorides (LiF, BeF_2), typically in crystalline form, and usually a mixture of two or more salts to a tank. Heaters (electrical heating elements) 231 are energized to melt the salt to molten state and maintain temperature well above melting point. Pulverized powdered spent nuclear fuel is taken from the ball mill 202 and carried by enclosed conveyor to the tank hopper 216 and deposited via hopper isolation valve 227 into the oxide reduction tank, and open isolation valve 217 (FIG. 2). Spent fuel addition, regulated by size and speed of the enclosed conveyor, known reaction rates derived from tests, feed limiters, ensures tank temperature is maintained within predetermined limits and sufficient mixing and reaction of tank contents occurs. All mixers and the tank pump are turned on to initiate mixing, recirculation by pumping, and consistency of salt fuel in the tank and pump discharge lines. In an exemplary implementation, tank size and screw pump capacity are regulated to allow 4-6 hours for complete mixing and sampling, density, sample enrichment and Mole% salt fuel are recorded and a second sample analysis completed and confirmed before a tank is ready to be discharged.

[160] Figures 5 and 6 show an exemplary implementation in plan and elevation views of a molten salt fuel collection tray, generally 240, consisting of the tray cover 254, salt fuel collection molds 256 atop heating and cooling elements 242, in a generally checkerboard pattern of heating elements 234A and cooling elements 234B. Insulation between elements along the sides and bottom of the tray prevents solidification during pouring and minimizes cooling time after solidification. More specifically, FIG. 5 is a plan view of salt mold cooling tray 240 with the tray cover and cooling molds removed.

[161] In an exemplary implementation, salt mold cooling trays 240 (FIG. 9) are positioned and held in a stacked array of 8-10 trays, with spacing between the trays being sufficient to allow for removal of the cooling molds and cover as one assembly. Stacked arrays are tracked together by a revolving drive (not shown) which moves one stacked and cooled array group, to the ball mill feed table 250. At the table 250, the one-piece molds of each tray, in a particular group, are removed and upended or overturned to deposit solid “bars” of salt fuel. Each tray’s mold is removed and returned to its position before the next mold is removed. After an entire array group has been emptied, it is returned in turn to be refilled with molten salt.

[162] An additional exemplary implementation accounts for the hygroscopic property of salt and salt fuel, so that each stacked array of molds is enclosed by a shroud and nitrogen inerting system (not shown) for the short time stacked arrays are being cooled, and such stacked array and enclosed cooling system ensures cooled nitrogen is recirculated around the stacked array and cooling compressor driven heat removal system. Such cooling and inerting is maintained until the stacked array solid salt, still at high temperature, but entirely solidified, is provided to the ball mill and fine mill, hot powdered salt fuel is put into standard containers or canisters, filled with argon, or cover gas and sealed.

[163] Tray molds are a non-stick surface, with salt fuel contraction during cooling, thereby facilitating solid salt fuel removal. The metal molds may be connected side-to-side and laterally supported to ensure tray strength and versatility. Solid salt “bars” are gathered to the side of the turning table and are generally organized lengthwise on a moving conveyor and fed into coarse ball mills 202A (FIG. 9). Product salt fuel from the ball mill is further conveyed to a fine mill, such as a Fitz mill 252 for sizing, sampling, certification, and packaging for protection against environmental conditions. More specifically, FIG. 6 illustrates a salt mold cooling tray front view, including top cover 254, cooling molds 256, heating and cooling elements 234A, 234B, which, in one implementation, could be coils.

[164] In an alternate implementation, molten salt fuel may be stored as a contiguous solid in canisters and subcritical arrays. This process involves preparation of fluoride salt fuel in the aforementioned receiving and mixing tanks 220, sampling and certification of tanks, and transfer by screw pump to a “critical safe” standard steel canister (not shown), filled with a cover gas, and sealed, and set aside for cooling. Canisters are transported and stored, in “critical safe” arrays. Facilities using “solid salt” canisters are equipped to remotely handle and inductively heat each canister to form liquid salt fuel for addition to their molten salt reactors.

[165] In FIG. 4, molten salt spent fuel receiving and mixing tanks 220 are shown in a side-by-side representative top view, but not to scale. More specifically, FIG. 4 shows an implementation wherein six receiving-mixing tanks 220 are side-by-side, including a tank pump discharge header, generally 260, and encased boron encasements 230 (sized for inner tank side dimension area). Boron encasements 230 provide structural stability and protection from damage and are positioned between salt fuel tanks 220 to maintain a sub-critical process to prevent nuclear criticality communication between tanks in a given array of tanks in close proximity, such array being considered herein as one subcritical assembly group, generally 266. Boron dividers 230 are backup defense, in depth, against possible criticality.

[166] As shown in an exemplary implementation in FIG. 2, the oxide reduction tank is the first tank in the process, wherein a system and process are illustrated which reduces uranium and plutonium oxides to fluorides. Non-fuel fertile-thorium oxide, may be added to the uranium/plutonium oxide reduction tank 210 and fluorinated to thorium fluoride salt After this process, the contents of the reduction tank 210 are pumped over to the mixing and adjustment tank. In one implementation, an isolation valve is provided on the loading hopper.

In an exemplary implementation wherein only fluoride salts are desired, the salts are prepared in the oxide reduction tank, and, then pulverized, powdered spent fuel is added to the oxide reduction tank, the temperature increase is noted, and oxide reduction is begun by the sparging action of anhydrous hydrogen fluoride. After an allowance of time necessary for mixing and water and hydrogen gas removal, toward complete reduction, the contents are pumped to the mixing and adjustment tank for final analysis, certification, and then pumped over to cooling trays. The teaching of the present disclosure includes reducing all oxides and hydroxides , removing oxygen entirely and preventing production of other oxides, ensures an authentic fluoride salt fuel, when oxygen is removed from the salt fuel. The result is that substantially the only byproduct from this implementation of the present disclosure is water, which is collected for sampling and released. Noble gases Krypton and Xenon may be removed from the process, and stored or otherwise used.

[167] In an exemplary implementation of the present disclosure, a method is illustrated in FIG. 8 for producing fuel for a thermal molten salt reactor, the implementation of the method including:

a. providing fuel assemblies, removing fuel pellets containing uranium and all spent fuel constituents, from the fuel assemblies;

b. reducing to powder, the fuel pellets in a semi-voided atmosphere using a ball mill, and fine mill, for process feed to the fluorination process;

c. processing the powdered spent salt fuel into fluoride salt by ultimate reduction and fluorination of the uranium and associated fuel constituents fluoride salt solution, by anhydrous hydrogen fluoride (AHF); following the additions of AHF, salt fuel reduction may be further enhanced by addition of metal hydrides;

d. enriching the spent salt fuel with U235, Pu239, or MOX;

e. fluorinating the enriched powdered spent salt fuel to yield molten fluoride salt fuel using AHF halide salt reduction;

f. analyzing, adjusting, and certifying the molten fluoride salt fuel for end use in a molten salt reactor;

g. pumping the molten fluoride salt fuel to stacked arrays of cooling trays or canisters and cooling the molten fluoride salt fuel to yield solid salt fuel bars, sticks, or canister solid form;

h. milling the solidified molten fluoride salt fuel to predetermined specifications for the thermal molten salt reactor; and

i. fertile-thorium oxide may optionally be added to the uranium/plutonium oxide reduction tank 210 and fluorinated to thorium fluoride salt.

Additional Exemplary Implementations

Briefly, additional exemplary implementations of the present disclosure include molten salt reactor salt fuel production by immersed oxide reduction processing. Various types of molten salt reactors (MSRs) are receiving government and industry attention due to the promise of greater efficiency of fuel use and the potential for use of reprocessed spent nuclear fuel (SNF). While most developmental attention has focused on the MSRs themselves, cost-effective production of fuel for the MSRs will likely be an important part of overall MSR acceptability. The present disclosure includes processes for production of MSR salt fuel based principally around acid halide reduction of oxide fuel reagents in molten carrier/dilutent salt. The process is relatively simple, requires minimal waste streams, and is inherently proliferation-resistant. Viability of the acid halide reduction in molten salt points to simplification and resultant reduction in cost of MSR salt fuel production facilities and thus lower cost of MSR salt fuel product.

Potential users of the present disclosure include electric utilities which operate light water power reactors and/or those entities which now have considerable inventories of spent fuel, and who may see need for more electric power generation capacity. These potential customers are also highly likely to be the owners and operators of anticipated MSRs, since MSRs operating on salt fuel derived from SNF may offer the opportunity to reduce inventories of spent fuel, with minimal waste streams.

Salt Fuel Production Processing

Processing spent nuclear fuel to salt fuel may be performed under U.S. Patents Nos. 10,685,753; 11,062,813; and 11,57,7968, each of which are incorporated herein in their entirety by reference. The process block diagram that appears in FIG. 11 represents the overall process for conversion of spent fuel from a spent nuclear fuel (SNF) assembly, 1100, or non-irradiated uranium dioxide to salt fuel. If starting with a SNF assembly, 1100, a dismantling of the SNF assembly and decladding of the SNF solids, 1101, will be necessary for access to the SNF solids, 1102. The dismantling and decladding operation generates assembly hardware metal for recycling, 1103; and off-gas, 1104. The conversion process in the molten salt processing function block, 1110, from oxide-based fuel, 1102, to halide salt fuel, 1114, can be performed by immersing, in a reaction vessel with agitation or pumped circulation and sparger, uranium oxide and/or plutonium oxide in a bath of molten carrier/dilutent halide salt (i.e., the carrier/diluent salt is heated to liquid and kept above its melting point), 1105.

The conversion process begins with the addition of granular or powdered oxide fuel, 1102, to the molten carrier/dilutent halide salt bath in a process vessel of the molten salt immersion process, 1110. Under continuous agitation or pumped circulation that keeps denser unreacted spent fuel in suspension, anhydrous hydrogen halide (e.g., hydrogen chloride or hydrogen fluoride), 1106, is sparged into the molten salt immersion process vessel, 1110, during the conversion of UO_2, PuO_2, fission products, and actinides to their respective halides (chlorides or fluorides corresponding to the anhydrous hydrogen halide chosen for sparged introduction), while resultant water vapor is removed as part of molten salt processing off-gas, 1112, and held in a tank, 1122, as product of off-gas processing, 1120, for later treatment and release. The chemical equations for fast molten salt reactor (FMSR) and thermal molten salt reactor (TMSR) salt fuel production are provided here:

The chemical equation for production of chloride salt fuel suitable for a fast molten salt reactor (FMSR) would be

$$2UO_2\ (s) + (\text{Anhydrous})\ 8HCl\ (g) \xrightarrow{\Delta 700\ C} UCl_3\ (l) + UCl_4\ (l) + 2H_2O\ (g) + Cl^- \qquad (1)$$

The chemical equation for production of fluoride salt fuel suitable for a thermal molten salt reactor (TMSR) would be

$$2UO_2\ (s) + (\text{Anhydrous})\ 8HF\ (g) \xrightarrow{\Delta 700\ C} UF_3\ (l) + UF_4\ (l) + 2H_2O\ (g) + F^- \qquad (2)$$

Sparging under agitation/circulation of vessel contents of the molten salt immersion process, 1110, accompanied by water vapor removal, 1112, continues through reduction of all oxides. The molten mixture in the molten salt immersion process vessel must be maintained slightly acidic throughout the entire process, so that the reaction continues uninterrupted. In this way, all oxygen is removed from the final product. All salt fuel requires enrichment via addition of U235 oxide ($^{235}UO_2$) or Pu239 oxide ($^{239}PuO_2$), 1108, to the vessel of the molten salt immersion process, 1110. FMSRs require the highest enrichment of just under 20%, provided with high-assay low enriched uranium (HALEU) for the initial and some subsequent fueling cycles. Enrichments for some fueling cycles of a FMSR may be lower as the Fast MSR generates (breeds) fissile fuel. After sampling and adjustment of salt fuel mixture composition, plus sampling of effluent off-gases, 1112, for confirmation of completion of reactions, the mixture may be certified as salt fuel product, released for solidification and packaging, 1116, and then released as packaged product, 1118.

Further Exemplary Implementations

Further exemplary implementations of the present disclosure include systems and processes as discussed below:

As shown in FIG. 12, an exemplary implementation includes: a reaction vessel top-mounted dry reagent inlet chute, 1200; a reaction vessel top-mounted stirrer motor, stirring shaft, and vessel-bottom agitator, 1202; a set of distributed internal, vessel-bottom spargers, 1204, fed by a valved AnhHCl line, 1206, from an AnhHCl tank, 120; and a vessel-internal dispersion plate, 1210, with multiple glow plugs wired in parallel, 1212. The implementation also includes external heaters, 1214; a condenser for off-gas water removal, 1216; a packed bed for trapping and removal of off-gas chlorides and other gases, 1218; an exhaust blower, 1220, to motivate venting, 1222; and battery back-up power, 1224, for instrumentation and controls.

FIG. 13 shows a sketch on an exemplary implementation of a reaction vessel tailored for process function. This implementation includes reaction vessel system with top-mounted dry reagent inlet chute, 1302; a reaction vessel top-mounted baffle rotation actuator (not shown: could be a hydraulic, pneumatic or electric), baffle rotation shaft, 1304, and; an immersed fluidized-bed-top baffle system with rotational interleaves, 1306 that allows, when in one position of interleaves, passage of solid reagent from the top-mounted dry reagent inlet chute down to the interior of an immersed fluidized bed below without interference of immersed baffle system components/interleaves; and directs, when in another (as-depicted) position of interleaves, passage of fluidized bed reaction byproduct gases against each of successive immersed interleaves (H_2 and O_2 gases rising through molten salt gather at each of the successive interleaves – since the interleaves are at molten salt temperature of at least 600C, comingling of H_2 and O_2 gases at any interleaf with result in auto-ignition and conversion to H_2O); an immersed oxide bed, 1308, atop of which is the immersed fluidized-bed-top baffle system, and the bottom of which is a sintered, fluidized-bed-bottom sparger system, 1310, fed by a valve-controlled AnhHX line from an AnhHX tank (where “X” may be “Cl” or “F”) and a controlled cover-/off-gas recirculation line, which may combine to form a common sparger inlet line upstream of the sparger; or may enter the sparger as separate inlet lines; or may comprise separate fluidized-bed-bottom spargers, fed jointly or separately by AnhHCl and cover-/off-gas lines; and a siphon line that extends from the reaction vessel top/cover/lid down into the molten salt and down into the bottom of the interior of the fluidized bed, thus serving as a molten salt sump line, 1312; a seamless reaction vessel [bottom], 1314, which may be of single-layer construction from corrosion resistant metal, suitable for mounting of contacting heating elements, or multi-layer construction, with corrosion-resistant interior layer, bonding layer, radiation-resistant structural layer (probably 316 SS), optional secondary bonding layer, optional induction-heater-compatible (ferritic) stainless steel exterior layer, reaction vessel heating elements (H - conductive or inductive) and cooling elements (C), 1316; and reaction vessel thermal insulation, 1318. FIG. 13 does not include off-gas processing system components, aside from a molten salt immersion reaction vessel cover-/off-gas (C/OG) outlet, 1320, that feeds into the off-gas process; and a common sparger inlet/feed line, 1322, fed by upstream combination of fresh AnHX and recirculated dry cover-/off-gas and extending from the molten salt immersion reaction vessel top/cover/lid down to the fluidized-bed-bottom sparger.

In operation, dry reagents enter via the top/cover/lid chute. Initial carrier/dilutent salt load will fill and overflow the fluidized bed shell. Upon heating to liquification of carrier/dilutent salt, the fluidized bed shell will be filled with liquid carrier dilutent. Subsequent entry of fuel oxide solids result in gathering of said solid in the bottom of the fluidized bed shell, whose bottom is shaped to keep fuel oxide solids over the sintered sparger at the bottom. After loading of fuel oxides, the rotational baffle is rotated to block vertical passage straight through the baffle system. Introduction of anhydrous hydrogen halide gas then begins. Fuel oxide solids at the bottom of the fluidized bed convert to molten fuel halide salt, through which solid oxide solids above descend, toward the sparger and toward sparging anhydrous hydrogen halide gas. Byproduct H_2 and O_2, meanwhile, percolate upward through the slowly converting-and-descending column of fuel oxides and background molten salt. Intermingling of H_2 and O_2 within the bed at molten salt temperatures results in auto-ignition of H_2 and O_2 and the production of superheated water vapor, which continues to percolate upward. Any H_2 and O_2 that do not auto-ignite within the fuel oxide bed are forced by the baffle system's interleaves to travel through a circuitous bath that presents many opportunities for gathering and intermingling of H_2 and O_2 gas underneath successive baffle interleaves, which, of course are at molten salt temperature and thus at above H_2O auto-ignition temperature by the time gases reach the surface of the molten salt, all H_2 and O_2 gas should have converted to H_2O gas/vapor.

Meanwhile (not shown), pumping moves cover gas and byproduct off-gas, including H_2O vapor, out of the reaction vessel and through a condenser, where H_2O condenses and is removed. The remaining, dry cover/off-gas (C/OG) is pumped back into the sparger feed line, so that any excess hydrogen halide or halide gas recirculates underneath and into the fluidized bed. The voluminous sparging of cover gas imparts an agitation action within the fluidized bed, so no further stirring should be necessary.

Upon completion of fuel oxide hydrohalogenation to fuel salt, the closed nature of the fluidized bed leads to a high concentration of molten fuel salt within the fluidized bed shell: this is drawn out by siphon to a lower-positioned receiving vessel or solidification mold, which eliminates the need for a molten salt pump.

The material flow of another exemplary implementation appears in the hydrofluorination process diagram in FIG. 14. This architecture Includes two, in-series fluidized beds, 1402 and 1404, which could be like those of a dry hydrofluorination process used at the Honeywell Uranium Hexafluoride Processing Facility, operated by Honeywell Specialty Chemicals in Metropolis, Illinois, with hydrohalogenation gas, 1410 and 1412 driven in counter-flow to forward-moving fluidized bed solids, 1400 and 1404. In operation, Solid reagents, 1400 enter the first hydrohalogenating fluidized bed, 1402, as fuel oxides or enrichment oxides, and exit 1402 as a mix of fuel/enrichment tetrachloride(s) and yet-to-be-reacted fuel/enrichment oxide(s), 1404. The mix of fuel/enrichment tetrachloride(S) and yet-to-be-reacted fuel/enrichment oxide(s), 1404, enters the second hydrohalogenating fluidized bed, 1406, and exits 1406 as all fuel/enrichment tetrachloride(s), 1408. The inventive process augments the dual, series fluidized beds with a third step of fuel, 1414, enrichment, 1416, and carrier, 1418, salt storage and retrieval, carrier salt receiving, 1420, and blending and melting of the fuel, enrichment, and carrier salts, 1422, to molten salt fuel blend, 1424. Melting of the blend "locks in" fuel components that, if separated, could be subject to proliferation. FIG. 14 does not show/specify the reagent transfer mechanism between fluidized beds 1402 and 1406. FIG. 14 also does not show/specific the heating device for the very hot anhydrous hydrogen halide gas that first enters the second fluidized bed, 1406. That heater may be a separately excited heater, scavenging/recirculating heat exchanger that draws heat from the exothermic reactions in the fluidized beds, or a combination of separate heating and scavenging/recirculation. FIG. 14 as shown uses a series pair of fluidized beds to hydrohalogenate either fuel oxides or enrichment oxides, but it is obvious that separate pairs of fluidized beds could separately hydrohalogenate fuel oxides apart from enrichment oxides. The hot off-gas, 1426, in Fig. 14 is handled by any of the off-gas processes for hydrohalogenation whose description follow.

FIG. 15 depicts, a process is similar to that used in by others in that "the pentachloride, UCl_5, is first produced and then decomposed in a second step." In its first step, the present implementation of the process carbochlorinates dry, largely-UO_2 SNF (instead of "pure" UO_2 used by others), 1500, in a sealed batch reaction vessel, 1502, under increasing temperature (through the range of approximately 115°C to 165°C) while allowing pressure to rise (through a range of approximately 118psi-300psi). Unlike prior techniques, the method of the present implementation first uses, if available from a prior carbochlorination step, dry phosgene ($COCl_2$), dry cover gas, and carbon tetrachloride (CCl_4) recovered from said prior carbochlorination step; upon reuse consumption of all previously recovered $COCl_2$ and all previously recovered CCl_4, the method of the present implementation then proceeds, in known manner, with fresh, dry liquid CCl_4. The process then releases batch reaction vessel pressure through a recovery/removal of byproduct gases and residual reagents and cover gas(es), 1504, which reagents and cover gas(es) are held, 1506, optionally separated, 1508, and stored, 1510, for later selection, 1512, and use through controlled release, 1514, in subsequent carbochlorination operations; and removes residual CCl_4 through dry inert gas sweeping at less than 115°C, with recovery and storage and separation of CCl_4 from the sweep gas, again, for later use in subsequent carbochlorination operations. In its second step, the present implementation of two-step process decomposes UCl_5 in the reaction vessel to UCl_4 product, 1516, by sweeping with dry inert gas at 350°C-500°C, but advances the state of the art in the present implementation's recovery and separation of dry inert gas, for reuse, and byproduct chlorine gas (Cl_2) for sale and/or for production of CCl_4.

Note that the following references are incorporated herein in their entirety: C. E. Larsen, "The Role of Chemistry in the Oak Ridge Electromagnetic Project," *Bulletin of*

Historical Chemistry, vol. 28, no. 2, 2003, (accessible at < http://acshist.scs.illinois.edu/bulletin_open_access/v28-2/v28-2%20p101-109.pdf >); E. L. Wagner, *The Methods of Preparing Anhydrous Tuballoy Tetrachloride*, Report 0.350.0 (later: AECD-4125), February 1946, (accessible at <https://www.osti.gov/servlets/ purl/4366153>); J. J. Katz and Eugene Rabinowitch, *The Chemistry of Uranium – Part I, The Element, Its Binary, and Related Compounds*, First edition. New York: McGraw-Hill, 1951 (accessible at <https://babel.hathitrust.org/cgi/pt?id= mdp.39015017271118&seq=7>); and A. Michael & A. Murphy, Jr., "On the action of Chlorine in a Solution of Carbon Tetrachloride and of Carbon Tetrachloride on Metallic Oxides," American Chemical Journal, Vol. XLIV, July-December 1910, pp. 365-384 (accessible at <

FIG. 16 depicts the architecture of another implementation of the present disclosure, including in this case use of a fluidized bed reactor that may be configured to operate in either batch or continuous operation. As typical with fluidized beds, solid granular, largely-UO_2 SNF, as charge, 1600, enters the fluidized bed reactor 1602, from above and is suspended in counter-/upward-flowing chlorinating reagent gases and sweeping/suspending inert cover gas, 1604, fed at the bottom of the reaction vessel. Unlike other processes, however, which require purity of reagent gas, the chlorinating gas/vapor feed of the present implementation is initially any phosgene ($COCl_2$), 1612, recovered and held, 1608, separated, 1612, from other off-gases, 1614, of prior carbochlorination operations, followed by, initially, addition of pure chlorinating reagent; in this case, CCl_4. All reaction gases that emanate from the top of the fluidized bed reaction vessel, 1604, are collected and pumped/compressed to a holding vessel, 1608, which could be a tank or large pipe, from which the collected gases pass through a temperature control system 1618, before recirculating to reintroduction to the bottom of the fluidized bed reaction vessel, 1602. The CO_2 product of reaction is known to have been a cover gas historically used for early proof of chemistry experiments, and is present during batch liquid-phase processes, so its presence will not impede forward progress of reaction. Reintroduction of CCl_4 from reaction vessel off-gas will appear to the reaction vessel to be introduction of chlorinating reagent. $COCl_2$ (phosgene) is both an intermediate by-product of the chlorinating process and a chlorinating agent for the remainder of the intended process, so its reintroduction to the reaction vessel will simply appear as that of a second chlorinating agent. It too, is present in batch liquid-phase processes, so, like CO_2, its presence will not impede forward progress of reaction. Reintroduction of CCl_4 and $COCl_2$, in fact, increases the reagent-charge gas-solid exposure/contact time for both gases, thus eliminating the need for a second, series fluidized bed that is necessary in a single-pass process like that used by others for hydrofluorination.

After initial chlorination of the UO_2 and fission product solids in the granular SNF, reaction vessel chlorination process gases are evacuated to the holding vessel, 1608, for introduction in a subsequent carbochlorination operation. The fluidized bed reaction vessel is charged with clean, dry inert gas. The fluidized bed contents can be optionally heated to 350°C-500°C while swept with recirculating inert gas, in an inert gas circulation loop, 1620, to decompose any UCl_5 in the reaction vessel to UCl_4, 1622. It is understood that known operations switched chlorination process from a liquid phase to vapor phase process in order to eliminate the time spent on the decomposition process step. Elimination of the decomposition step, however, results in the need for very tight variability of incoming charge and reagent feed materials. Given the inherent variability in SNF composition, it is advantageous to utilize a two-step process whose first step drives to complete chlorination and whose second step ensures consistency of overall process outcome. If an optional decomposition step is/was included for overall process robustness, that step is followed by evacuation of decomposition gases and replacement of the decomposition gases with clean inert cover gas. A gas selection manifold, 1616, facilitates selection of which gases to recirculate back to the reaction vessel to support potentially changing operations in the reaction vessel. The decomposition gases, containing Cl_2, undergo separation and bottling for subsequent sale or reuse. Reuse could, in fact, be for production of fresh CCl_4.

Note that the following references are incorporated herein in their entirety: E. L. Wagner, *The Methods of Preparing Anhydrous Tuballoy Tetrachloride*, Report 0.350.0 (later: AECD-4125), February 1946, (accessible at <https://www.osti.gov/ servlets/purl/4366153>); J. J. Katz and Eugene Rabinowitch, *The Chemistry of Uranium – Part I, The Element, Its Binary, and Related Compounds*, First edition. New York: McGraw-Hill, 1951 (accessible at <https://babel.hathitrust.org/cgi/pt?id= mdp.39015017271118&seq=7>); and Hubbard, J V, Perlmutter, H A, Goren, H L, and Clewett, G H. *THE PREPARATION OF TUBALLOY TETRACHLORIDE BY FLUIDIZATION*. United States: N. p., 1945. Web. (Note that this report has a USDOE OSTI listing at <https://www.osti.gov/biblio/4359404>, but that listing may not connect to an actual document).

FIG. 17 depicts the architecture of a process that assumes use of largely UO_2 SNF rod/pin contents from mechanical decladding, 1600, in parallel with carbochlorination of largely U_3O8 from voloxidation extraction of residual materials entrained or infused in SNF cladding, 1700. The reason for the parallel processes is that mechanical decladding will more-quickly make available largely UO_2 SNF rod/pin contents, but is unlikely to extract SNF rod/pin contents that may be bonded, infused, or otherwise entrained in the SNF cladding material. Voloxidation, on the other hand, is known to reduce UO_2 to fine powder more easily extracted from the SNF cladding material, but is a heating process that is slower to extract material than mechanical decladding. Use of a parallel process allows quick extraction of most SNF rod/pin contents by mechanical decladding, thus eliminating time necessary for voloxidation of said easily-removal contents and allowing voloxidation operations to focus on the much smaller amount of SNF/rod/pin contents bonded, infused, or otherwise entrained in the SNF cladding material.

The process of the present implementation carbochlorinates the retrieved loose, largely-UO_2 SNF solids to largely UCl_4 salts by either the liquid-phase two-step process described earlier/above or the vapor-phase one-or-two-step process described earlier/above; and carbochlorinates the largely-U_3O_8 output from decladding voloxidation in a separate, parallel reaction vessel, 1702, to yield largely UCl_4 salt, 1722, by either the two-step CCl_4 liquid-phase carbochlorination process described earlier/above OR the one-or-two-step CCl_4 vapor-phase carbochlorination process described earlier/above. The gas recirculation loop, 1720, for the separate, parallel reaction vessel is analogous to that for the first reaction vessel, 1620. If uranium trichloride (UCl_3) is the uranium chloride specified in a salt fuel blend specification, the process reduces the collected dry UCl_4 from the parallel carbochlorination steps to UCl_3 via exposure to purified hydrogen (H_2), optionally under pure, dry inert cover, at temperatures of initially no more than 575°C and up to 650°C toward end-of reduction (per Katz and Rabinowitch, pp 450-451). This reduction step, whose gaseous byproduct is hydrogen chloride (HCl) obviously can be skipped if the salt fuel specification specifies UCl_4. Should it be required, however, processing of the reduction process cover/of-gas may be accomplished through inclusion of an off-gas processing system loop whose architecture(s) are similar to those that follow below for hydrohalogenation processes. The difference is that relatively little H_2O should be present in the off-gas, resulting in almost all chlorine-containing off-gas being readily separable hydrogen chloride.

Note that the following references are incorporated herein in their entirety: E. L. Wagner, *The Methods of Preparing Anhydrous Tuballoy Tetrachloride*, Report 0.350.0 (later: AECD-4125), February 1946, (accessible at <https://www.osti.gov/servlets/purl/4366153>); J. J. Katz and Eugene Rabinowitch, *The Chemistry of Uranium – Part I, The Element, Its Binary,* *and Related Compounds*, First edition. New York: McGraw-Hill, 1951 (accessible at <https://babel.hathitrust.org/cgi/pt?id=mdp.39015017271118&seq=7>);Hubbard, J V, Perlmutter, H A, Goren, H L, and Clewett, G H. *THE PREPARATION OF TUBALLOY TETRACHLORIDE BY FLUIDIZATION.* United States: N. p., 1945. Web. (This report has a USDOE OSTI listing at <https://www.osti.gov/biblio/4359404>, but that listing does not connect to an actual document.); A. Michael & A. Murphy, Jr., "On the action of Chlorine in a Solution of Carbon Tetrachloride and of Carbon Tetrachloride on Metallic Oxides," American Chemical Journal, Vol. XLIV, July-December 1910, pp. 365-384 (accessible at < https://babel.hathitrust.org/cgi/pt?id= uiuo.ark%3A%2F13960%2Ft5fb5dz5g&seq=1> ; and Robert Jubin, "Spent Fuel Reprocessing," Oak Ridge National Laboratory (accessible at http://www.cresp.org/NuclearChemCourse/monographs/07_Jubin_Introduction%20to%20Nuclear%20Fuel%20Cycle%20Separations%20-%20Final%20rev%202_3_2_09.pdf.)

In an alternate implementation, a rotating drum, 1700a with helical vanes, 1702a, similar to a concrete mixer, could be used instead of or in addition to the fluidized beds discussed above. Depending on the direction of rotation of the drum about its drum axis, 1704a, the constituents therein will be moved downwardly or upwardly for discharge of such constituents. See, for example purposes, Fig. 2 of U.S. Patent No. 2,618,472, issued November 18, 1952 to Castendyck, the entirety of which is incorporated herein by reference. Dry reagents enter the opening at the upper right. Upon heating to liquification of carrier/dilutent salt, the drum will be filled with liquid carrier dilutent. Subsequent entry of fuel oxide solids result in gathering of said solid in the drum and could be rotated such that the constituents are carried downwardly towards the bottom of the drum a sparger (not shown) is positioned at the bottom of the drum, through which anhydrous hydrogen halide gas is introduced to convert to the constituents to molten fuel halide salt as the drum rotates to lift the constituents upwardly to the right by the vanes, the constituents eventually fall off of the vanes and descend back towards the bottom of the drum toward the sparger and toward sparging anhydrous hydrogen halide gas. Byproduct H_2 and O_2, meanwhile, percolate upward through the slowly converting-and-descending masses of fuel oxides and background molten salt. During this mixing phase, the outlet of the drum is closed, to force the above-mentioned recirculation of the constituents. To empty the constituents from the drum, the drum is rotated in the direction to lift the constituents to the outlet for discharge from the drum.

FIG. 18 depicts the high-level context of halogenation, 1810, including but not limited to molten salt hydrohalogenation, that sends its process off-gas, 1812, to off-gas processing, 1820, , relative to a process input stream, 1800, comprised of constituents of carrier/diluent salts, 1802, substantially uranium dioxide spent nuclear fuel, 1804, enriching fissile oxides, including but not limited to $^{235}UO_2$ and $^{239}PuO_2$, 1806, and halogenating agents, 1808; and relative to byproducts releasable to environment, 1822, such as water or CO_2, and salable by-product gases, 1824; and relative to a process , 1830, that receives halogenation process product, 1814, solidifies any molten product and packages product fuel salt for storage in an inert gas-charged fuel salt container, 1832.

FIG. 19 depicts the high-level context of halogenation, 1810, including but not limited to molten salt hydrohalogenation, and off-gas processing, 1820, in the context of process input streams comprised of constituents of carrier/diluent salts, 1802, substantially uranium dioxide spent nuclear fuel, 1804, that are outputs of dismantling and decladding, 1902, of spent nuclear fuel assemblies, 1900, and enriching fissile oxides, including but not limited to $^{235}UO_2$ and $^{239}PuO_2$, 1806; and relative to byproducts releaseable to environment, 1822, such as water or CO2, and salable byproduct gases, 1824; and relative to a process, 1830, that receives halogenation process product, solidifies any molten product and packages product fuel salt for storage in an inert gas-charged fuel salt container, 1832. FIG. 19 depicts how off-gases, 1904, from dismantling of a spent nuclear fuel assembly and decladding of the spent nuclear fuel assembly's fuel rods/pins, 1902, may be routed to the off-gas processing gas streams of the halogenation off-gas processing, 1820, in order to consolidate facilities. FIG. 19 also notes where recyclable metals from the dismantling and decladding operation, 1902, exit the overall process.

FIG. 20 depicts a refinement of the architecture of FIG. 18, in which the off-gas processing section comprises a front-end condenser, 1827, for the extraction of water vapor from halogenation off-gas, 1812, and a cryogenic separation system, 1828, that outputs salable off-gases, 1824, separate from recirculable gases, 2026, that may be stored or returned to the halogenation process, 1810, such as but not limited to cover gas. The cryogenic separation system is introduced as an alternative to the earlier packed bed off-gas trap that does not require back-driving of bed trap materials for gas recovery.

FIG. 21 depicts as refinement of the of condenser/cryogenic Off-Gas Process of FIG. 20 that shows circulation of post-cryogenic process recirculable gases, 2026, such as but not limited to argon cover gas, feeding back in counter-flow to the cryogenic process and then condenser and then to the molten salt immersion reaction vessel to pre-chill gases sent to the cryogenic system. This counter flow cooling with gas helps cool the water-removal condenser and avoid coolant phase change in that condenser; and preheats recirculating gases, such as cover argon, returning to the molten salt immersion reaction vessel, thus reducing eliminating need to separately preheat the returning cover gas for prevention of water condensation in the molten salt immersion reaction vessel.

FIG. 22 depicts the architecture of an exemplary implementation that refines the architecture of FIG. 21 by including a path, isolated by a pair of valves, for post-water-removal gases to bypass the cryogenic separation system and recirculate to the halogenation process until after completion of the fuel oxide halogenation process; and a trap for halide gas and hydrohalic acid, 2210, upstream of the cryogenic system, 2010. The bypassing eliminates the need for cryogenic process capacity to handle full, post-water-removal gas flow. This arrangement eliminates the need for cryogenic process capacity to handle full, post-water-removal gas flow, by bypassing cryogenic separation until after completion of the fuel oxide halogenation process. Cryogenic separation can occur more slowly and with a more-compact, less-expensive cryogenic separation system. The halide/hydrohalic acid trap further decreases separation load requirements of the cryogenic separation system, allowing use of a more-compact, less-expensive cryogenic separation system.

Fig 23 depicts the architecture of an exemplary implementation that replaces the front-end condenser, 2010, of FIG. 22 with a front-end H_2O separation process, 2310, and that shows circulation of all dry (post-water-removal) off-gases in counter-flow to the condenser and then to the molten salt immersion reaction vessel. The water removal system separates the H2O from hydrohalic acid, which simple distillation cannot achieve due to an H2O-HX azeotrope. This system first condenses out water as part the form of hydrohalic acid, thus yielding a cool dry off-gas effluent component, 2312c, and a hydrohalic acid effluent component. The system then breaks the azeotrope to separate the hydrohalic acid to water and anhydrous hydrogen halide. The separation means may be any of, and without limitation to, boiling, calcium chloride, sulfuric acid, and pressure swing distillation processes familiar to industry for recovery of dry hydrogen halide. Use of the cool dry off-gas, 2312c, for at least part of water separation process cooling heats the dry-off-gas, resulting in hot dry off-gas, 2312h, that is available for controlled reintroduction to the halogenation process, 1810.

FIG. 23 depicts the architecture of an exemplary implementation in which the arrangement in FIG. 22 is further augmented by inclusion of an experimentation/emergency off-gas treatment system that comprises an off-gas three-way valve, 2400, whose position defaults to routing halogenation off-gases to an emergency gas treatment trap, 2410, which, in turn outputs to environment through a vent, 2412. The default/power-off flow path of the off-gas three-way valve is to the experimental/emergency off-gas treatment trap, 2410, whose size is sufficient to treat all off-gas in case of loss or disconnection of process power. This path also may be used for low-volume experimentation/verification purposes.

FIG. 24 shows in greater detail an exemplary implementation whose high-level architecture appeared in FIG. 23. FIG. 24 includes the valve for and immersion reaction vessel effluent off-gas default path to experimentation/emergency off-gas treatment and vent, but does not show the detail of the experimentation/emergency off-gas failsafe treatment and vent. FIG. 24 includes the outlet and return from the water separation subsystem to/from the cryogenic separation subsystem, but does not include the details of that cryogenic separation subsystem.

Not diagrammed but available as an option to cryogenic separation is the technique of gas absorption in liquid carbon dioxide. Originally developed for processing off-gas from the High-Temperature Gas-cooled Reactor (HTGR), the method is potentially attractive because of “explosion hazard associated with radiolytic formation and concentration of ozone” in SNF. No similar explosion hazard has been identified for technique of gas absorption in liquid carbon dioxide. On the other hand, the technique does rely heavily on gas streams with high carbon dioxide concentration. Given that carbon dioxide has been used as cover gas for carbochlorination of uranium oxides, and that the primary gaseous by-product of carbochlorination of uranium dioxide in carbon dioxide, the off-gas from carbochlorination of uranium oxides should be suitable as feed gas for absorption in liquid carbon dioxide.

In the embodiment of FIG. 25, the molten salt immersion (or hydrohalogenation) reaction vessel, 1, accepts AnhHX from a supply tank, 2, whose output flow is controlled by a feed control valve, 3, and passes to the reaction vessel, 1, via a feed line, 4. Gaseous effluent from the molten salt immersion reaction rises through a stack, 5, to a three-way valve, 6, whose default, power-Off position diverts gas flow via a path, 7,to failsafe off-gas treatment via passive off-gas traps and follow-on catalytic converter, which are not shown. In normal operation, the three-way valve is energized and allows reaction vessel effluent gas to pass from the reaction vessel stack, 5, to a [possibly multiple-element] fractionating column, 8, positioned and cooled to effect the condensation, possible solidification, and reflux return of heavier off-gas condensates to the reaction vessel, 1, via the reaction vessel effluent stack, 5. Lighter gases pass on to a second condenser, 9, whose outputs are either dry off-gas or condensate liquid HX-H_2O (hydrohalic acid). The dry off-gas are drawn through a dry off-gas line 10 to a pump/compressor, 11, which pushes this dry off-gas to a higher pressure through a check valve, 12, onward to a dry gas pressure line, 13, and thence to a dry off-gas tank, 14. The condensate liquid HX-H2O passes through a liquid HX-H_2O line, 15, to a pressure swing distillation (PSD) subsystem, 16, whose details are not shown but which comprises a low-pressure distillation column that feeds, through a pump, a high-pressure distillation column, and whose low-pressure distillation column outputs H_2O vapor/gas at 1 atm pressure while its high-pressure distillation column outputs gaseous HX at a higher pressure (in the range of 3atm-15atm: the diagram shows a target “high” pressure of 10atm. The gaseous water from the PSD subsystem passes through a condenser, 17, which converts the gas to liquid that collects in a holding tank 18 for analysis and any further treatment prior to release. The HX from the PSD subsystem, now largely free of H_2O, passes through a dry HX output line, 19, through a protective check valve, 20, to the dry off-gas tank, 14, which serves as a storage buffer and whose combined contents are [largely] water-free.

With the H_2O removed from the off-gas stream, the dry off-gas in the dry off-gas tank, 14, passes through a dry off-gas cooler, 21, whose output is protected by a check valve, 22, and supplies a three-way, directional dry gas separation access control valve, 23. During hydrohalogenation, the dry gas separation access control valve routes dry off-gas flow to a dry off-gas return control valve, 24. The dry off-gas return control valve meters cold dry off-gas from the dry off-gas cooler, 21, and feeds cold dry off-gas through a cold dry return off-gas line, 25, to the coolant inlet of the lower temperature off-gas condenser, 9, where it enters and counter-flows to cool the warm off-gas to below the point of condensation of entrained off-gas HX-H_2O. Upon exit from the coolant circuit of the lower-temperature condenser, 9, the now-warm dry off-gas passes through a warm dry return off-gas line, 26, to the inlet of the higher-temperature fractionating column, 8, where it counter-flows to cool the hot off-gas that enters the fractionating column, 8, from the reaction vessel effluent stack, 5, with the effect of cooling to the point of condensing all heavier off-gas constituents for reflux return back to the reaction vessel, 1, via the reaction vessel effluent stack, 5. Upon leaving the fractionating column, 8, the dry off-gas is now hot and ready for return via a hot dry return off-gas line, 27, to the reaction vessel, 1, without excessive cooling of the reaction vessel contents. Should the reaction vessel contents require cooling, a dry off-gas return mix valve, 28, allows cool dry return off-gas in the cool dry return off-gas line, 25, to bypass the fractionating column, 8, and condenser, 9, and join and mix with hot dry return off-gas from the hot dry return off-gas line, 27, to produce off-gas at the temperature appropriate for introduction to the reaction vessel, 1. Proper flow of return off-gas to maintain operation of the condensers, 8 and 9, is controlled via the dry off-gas return control valve, 24. This return system thus facilitates cooling of the off-gas processing fractionating column, 8, and condenser, 9, returns all heavier fission products in the reaction vessel effluent stream back to the reaction vessel, 1, and returns any excess HCl from the reaction vessel, 1, back to that vessel. NOTE1: it should be obvious that, should the fractionating column, 8, require cooler coolant than the warm dry off-gas from the condenser, 9, a fractionating column coolant mix valve may be used upstream of the fractionating column cooling inlet to blend the warm dry return off-gas in the warm dry off-gas line, 26, with cold dry return off-gas in the cold dry return off-gas line, 25, prior to introduction to the fractionating column coolant inlet.

Once the hydrohalogenation process is complete, as indicated by significant rise/jump in PSD HX output, the feed of fresh AnhHX from the AnhHX supply tank, 2, can be terminated by closure of the AnhHX feed control valve, 3, and dry off-gases may be allowed to exit this H_2O extraction system via the dry off-gas separation access control valve, 23, which allows the dry off-gas of the system to exit via an outlet, 29, to the dry off-gas [cryogenic] separation process [of 3.ix]. Residual Argon from the dry off-gas separation process returns to the off-gas H_2O removal system via a return inlet, 30, to purge the off-gas H_2O removal system and facilitate retrieval of non-cover gases for separation and reuse.

Incorporated by reference in their entirety are the following:

Volkovich et al - <Effect of Melt Composition on the Reaction of Uranium Dioxide with Hydrogen Chloride in Molten Alkali Chlorides (degruyter.com)>

1. Warren & Ferris (ORNL) - <OXIDATION AND CHLORINATION OF UO_2-PuO_2. (osti.gov)>
2. Sakamura - <Chlorination of UO2, PuO2 and rare earth oxides using ZrCl4 in LiCl–KCl eutectic melt - ScienceDirect>.
3. Purafil Chlorosorb Ultra product bulletin - <Purafil-Chlorosorb-Ultra-Media-Bulletin.pdf>
4. USDOE Technology Readiness Assessment (TRA) / Technology Maturation Plan (TMP) Process Implementation Guide, Rev. 1. August 2013 - <Microsoft Word - g413.3-4Final10-12-09 (doe.gov)>.
5. Figueroa, Javier, & Williamson, Mark A. Uranium Dioxide Conversion. United States. https://doi.org/10.2172/1159227.

6. USNRC Technical Training Center, 3/10 Rev 4, Fuel Cycle Processes DirectedSelf-Study Course, https://www.nrc.gov/docs/ML1204/ML12045A005.pdf.

In an exemplary implementation using carbochlorination with liquid carbon tetrachloride, instead of hydrohalogenation with hydrogen halide, carbochlorination product gases are recovered and cooled, and then separated and bottled. Byproduct CO_2 is disposed of through sale or other use, such as use as carbochlorination cover gas; recovered cover gas, if not CO2, is bottled for reuse; recovered $COCl_2$ is bottled or otherwise held for use in the initial step of subsequent carbochlorination operations; recovered CCl_4 is condensed and bottled or otherwise held for use in the second step of subsequent carbochlorination operations. All other gases, including fission byproduct noble and non-noble gases, are bottled for sale. Chlorine gas from the decomposition of UCl_5 to UCl_4 is separated from the inert sweep gas used in the decomposition process and bottled for sale or other use, and the inert gas recovered and bottled or otherwise held for re-use in subsequent decomposition operations. The process of the present implementation addresses disposal through use of recovery, separation and bottling of CO_2 for sale or subsequent other use, and re-use of byproduct $COCl_2$ and excess CCl_4 in subsequent carbochlorination operations, thus eliminating the need to dispose of such chemicals, and eliminates need to vent gases as emissions. In an exemplary implementation using carbochlorination with carbon tetrachloride vapor, all carbochlorination reaction gases that emanate from the top of the fluidized bed reaction vessel are collected and pumped/compressed to a holding vessel, which could be but is not limited to being a tank or pipe, from which the collected gases pass through a temperature control system before recirculating reintroduction via control, such as but not limited to valving, to the bottom of the fluidized bed reaction vessel. The CO_2 product of reaction is known to have been a cover gas used for early proof of chemistry experiments, and is present during batch liquid-phase processes, so its presence will not impede forward progress of reaction. Reintroduction of CCl_4 from reaction vessel off-gas will appear to the reaction vessel to be introduction of chlorinating reagent. $COCl_2$ (phosgene) is both an intermediate byproduct of the chlorinating process and a chlorinating agent for the remainder of intended process, so its reintroduction to the reaction vessel will simply appear as that of a second chlorinating agent. It too, is present in batch liquid-phase processes, so, like CO_2, its presence will not impede forward progress of reaction. Reintroduction of CCl_4 and $COCl_2$, in fact, increases the gas-solid exposure/contact time, thus eliminating the need for a second, series fluidized bed that is necessary in a single-pass process like that used for hydrofluorination. At the end of carbochlorination, the fluidized bed reaction vessel is purged of reaction gases and charged with dry inert gas.

If the carbochlorination process is to be followed by a heated dry inert gas decomposition that reduces any fluidized bed reaction vessel UCl_5 to UCl_4 and yields chlorine gas (Cl_2), the foregoing dry inert gas charge is temperature controlled and circulated or otherwise swept through the fluidized bed reaction vessel and recovered with decomposition product gases of principally Cl_2, which decomposition product gas Cl_2 is separated, and bottled for sale, to allow recirculation and reuse of dry inert gas. The off-gas process of the present implementation minimizes the overall amount of reagent gases and inert gas through use of recovery and recirculation, and eliminates need to vent gases as emissions.

Note that the following reference is incorporated herein in their entirety: J. J. Katz and Eugene Rabinowitch, *The Chemistry of Uranium – Part I, The Element, Its Binary, and Related Compounds*, First edition. New York: McGraw-Hill, 1951 (accessible at <https://babel.hathitrust.org/cgi/pt?id=mdp.39015017271118&seq=7>)

Beyond carbochlorination and decomposition, the solid and not molten product of the carbochlorination processes of the present implementation may proceed through any of a number of process paths.

The dry, largely-UCl_4 salt product from carbochlorination may be removed and packaged as-is in sealed, dry-cover-gas-charged storage containers, for later processing; the dry, largely-UCl_4 salt product from carbochlorination may be (optionally removed from the carbochlorination reaction vessel and), under dry inert cover conditions, blended with dry carrier/dilutant salt and stored as a granular blend of separate solid salts, as-is; or melted in/under dry conditions and formed and cooled to countable blocks, bricks, ingots, cylinders, semi-cylinders, rods, spheres, combinations thereof, or of some other suitable shape of blended-while-molten salt blend; or melted in/under dry conditions, formed and cooled to blocks, bricks, ingots, cylinders, semi-cylinders, rods, spheres, combinations thereof, or of some other suitable shape, then milled, shredded, or otherwise sized to grains or powder of blended-while-molten salt blend; or the dry, largely-UCl_4 salt product from carbochlorination may be (optionally removed from the carbochlorination reaction vessel and), under dry inert cover conditions, blended with dry carrier/dilutant salt and further blended with enrichment salts and stored as a salt fuel, as-is; or melted in/under dry conditions and formed and cooled to countable blocks, bricks, ingots, cylinders, semi-cylinders, rods, spheres, combinations thereof, or of some other suitable shape of salt fuel; or melted in/under dry conditions, formed and cooled to blocks, bricks, ingots, cylinders, semi-cylinders, rods, spheres, combinations thereof, or of some other suitable shape, then milled, shredded, or otherwise sized to grains or powder of blended-while-molten salt fuel.

Forming the salt product into blocks, bricks, ingots, cylinders, semi-cylinders, rods, spheres, combinations thereof, or of some other suitable shape (collectively referred to herein as "solids") should facilitate accountability of such salt product, as compared to it being in a powder/granular, or "liquid," form and thereby potentially reduce the likelihood of proliferation concerns, as well as facilitating inventory control.

In one example implementation, such salt product solids could be stored prior to enrichment and then such salt product in powder/granular and/or in solid form could be enriched later as needed for Just-In-Time (JIT) supply to an MSR.

In another example implementation, the solids could be milled into powder or granular form in JIT manner as need for blending to an MSR's fuel specifications and/or for input into an MSR.

In a yet another example implementation, such powder/granular and solids could potentially be stored in the inert-gas-charged canisters/drums and casks in which the light water reactor SNF was previously stored; provided, however all such storage and handling of such powder/granular and solids would necessarily be subject to criticality safety constraints.

FIG. 26 is a high-level flow diagram that captures the relationship between major material/process sections discussed and also conveys an overall physical "U-path" of material flow. The material flow path starts from a dry transfer system (DTS), 2610, at left of the diagram, which effectively acts as a sheltered "stop" in the spent nuclear fuel (SNF) cask/canister transfer path between the nuclear power plant (NPP) SNF cooling pool, 2600, and the NPP site's independent spent fuel storage installation (ISFSI), 2670. SNF assemblies, 2612, leave a SNF canister/cask in the DTS, 2610, traverses rightward through SNF assembly dismantling and decladding operations, 2620, which yield SNF solids, 2622, that further rightward into SNF halogenation operations, 2630, as well as assembly metals and zirconium tetrachloride, 2624 for recycling. From the halogenation operations, 2630, the material flow path "crosses over" to salt solidification operations, 2650, from which solidified fuel salt, 2652 returns leftward through salt packaging operations, 2660. Salt packages for storage, 2662, are loaded back into cavities of the SNF storage canister/cask that was the source of SNF assemblies whose fuel oxides were just converted to fuel salts or salt fuel blend. The SNF storage cask leaves the DTS as a fuel salt cask, 2670. All of the above operations are connected to off-gas processing, 2640. FIG. 26 indicates presence but does not does not include details of either (i) cover/off-gas (C/OG, 2642) input to gas processing, 2640 from the dismantling and decladding, 2620, salt solidification, 2650, and salt packaging, 2660, operations; or (ii) cover gas supply from gas processing, 2640, to dismantling and decladding, 2620, salt solidification, 2650, and salt packaging, 2660 operations. Incoming C/OG, 2642) would be pumped from each operation to the gas separation system; general supply of cover gas(es) would come from the fluid selection manifold. Detailed pressure, temperature and flow control would be performed/monitored at each operation, in a fashion similar to the fluid controls for the halogenation and reflux condensation system.

The U-path and use of the SNF source canister/cask as fuel salt or salt fuel blend storage vessel eliminates the need for a separate storage facility for process output fuel salt or salt fuel blend. If the NPP SNF cooling pool facility/area has enough space to accept a full complement of the SNF-to-salt processing system disclosed herein equipment while still leaving sufficient space for SNF cask transporter maneuver, the sheltered stop mentioned above could, in fact, be in the NPP SNF cooling pool facility/area. Such a location would eliminate need for a separate DTS and thus further reduce SNF-to-salt process implementation cost.

With the context of the high-level process flow of FIG. 26 in mind, FIG. 27 is a “unified” repartitioning/repicturing of halogenation and gas processing functions, applicable to both hydrohalogenation and carbochlorination.

Included in the halogenation reaction vessel and reflux condenser block, 2710, are the halogenation reaction vessel that accepts uranium oxides, 2700, for halogenation to fuel salts, 2714, and the reflux condensation system to return metal and metal salt condensate to the halogenation reaction vessel, as well as pass on halogenation off-gas, 2712, The distillation condensation block at the front end of gas processing, 2720, is the same as that disclosed above to separate hydrohalic acid from other off-gases of hydrohalogenation; and a distillation condensation block is in the same place to separate carbon tetrachloride and phosgene from cover/product CO_2 and other gases of carbochlorination, in either case, the distillation condenser 2720 condenses excess halogenation agents, 2724, from the balance of off-gas, 2722. The gas separation system block, 2730, includes internal holding tanks and valves and comprises the cover/off-gas recirculation system combined with the cover/off-gas separation system disclosed above. The condensate separation block, 2740, in the case of hydrohalogenation, would be the pressure swing distillation system disclosed above for separating hydrohalic acid into water and anhydrous hydrogen halide; in the case of carbochlorination, it would be a pass-through or system for transfer of recovered CCl4 to a holding tank. The holding tank system, 2750, accepts and temporarily stores separated halogenation agents, 2742, and other gases, 2732, and comprises the gas bottles and liquid tank from figures shown and discussed herein. From the holding tank system, 2750, the fluid selection manifold, 2760, disclosed above, selectively accepts stored fluids, 2752, and routes halogenation agents and cover gas for recirculative reuse, 2766, as-needed, and also serves as dispensing point for outgoing fluids that are to be released to environment, 2762, or for sale , 2764. The fluid pressure and temperature control, 2770, was disclosed above, but appeared as part of the gas processing system; depending on implementation need, it may be appropriately grouped with the halogenation and reflux condensation system, instead and use the gas processing system as a fluid supply: the interface location between functional blocks may change, but the function of the fluid pressure and temperature control remains the same.

Prior disclosures incorporated above by reference discussed mechanical decladding and what is perhaps a variation of voloxidation. FIG. 28 depicts a dismantling and decladding material flow that assumes mechanical dismantling and decladding followed by chemical decladding in a

The SNF assembly dismantling may be, but is not restricted to, the methods disclosed in *Engineering Design of a Voloxidizer with a Double Reactor for the Hull Separation of Spent Nuclear Fuel Rods*, Young-Hwan Kim, et al, w, Science and Technology of Nuclear Installations, Vol 2017, Article ID 985; Economic Analysis on Direct Use of Spent Pressurized Water Reactor Fuel in CANDU Reactors – I: DUPIC Fuel Fabrication Cost, by Hangbox Choi, Won Li Ko, and Myung Seung Yang, Korea Atomic Energy Research Institute, *Nuclear Technology*, Vol. 134, May 2001 (each of the foregoing references being incorporated in their entirety herein by reference and hereinafter referred to as the “KAERI Documents”) and/or as disclosed in U.S. Provisional Patent Application No. 63/495,416, filed April 11, 2023, also incorporated in their entirety herein by reference, hereinafter referred to as the “‘416 Application.”.

As appears in block 2820 of FIG. 28, “sizing” is reduction of size of SNF rods (aka “pins”) and mechanical decladding is the mechanical freeing and collection of the bulk of SNF rod/pin contents from mechanical constraint by the SNF rod/pin tube, and implementation of these operations may be, but are not limited to, the methods disclosed in the KAERI Documents and/or the ‘416 Application. This activity frees as principal output the remains of spent nuclear fuel solids, 2822, that are largely uranium dioxide, and as by-products fuel rod/pin cladding hulls, 2824, and off-gases, 2826, all of which contain fission products.

Chemical decladding as appears in block 2830 of FIG. 28, is a chlorination process that dissolves the Zircaloy comprising a SNF rod/pin tube/cladding. Dissolution as sole means of decladding, however, would likely take more time than mechanical decladding for the freeing of the bulk of SNF rod/pin solids, and mechanical decladding exposes the interior of the Zircaloy tube and thus enhances a follow-on cladding hull dissolution step. The process of FIG. 28 assumes chemical decladding, 2830, as an augmentation to preliminary mechanical decladding, 2820. The chemical decladding completely removes zirconium from the SNF rod/pin cladding and thus, in converse, frees all SNF rod/pin contents from the SNF rod/pin Zircaloy tube by removing tube material. The preferred process may be, but is not restricted to, the chlorination process disclosed in Collins et al, 2012, "Process Development Studies for Zirconium Recovery/Recycle from Used Nuclear Fuel Cladding," *Procedia Chemistry*, 7 (2012) 72-76, https://www.sciencedirect.com/science/article/pii/S1876619612000873>; Collins et al, 2015, "R&D Progress on Recovery/Recycle of Zirconium from Used Fuel Cladding," USDOE OSTI 1239757, <https://www.osti.gov/servlets/purl/1239757>;Collins et al, 2017, "Engineering-scale Development/Demonstration of Zirconium Recovery from Used Fuel Cladding," USDOE OSTI 1766433, <https://www.osti.gov/servlets/purl/1766433>, the entirety of the foregoing references being incorporated by reference herein and being referred to collectively as the "Collins Documents." Zirconium tetrachloride ($ZrCl_4$) output from a process like that disclosed in the Collins Documents is sufficiently pure to not require special radiological handling and may be introduced and combined with other $ZrCl_4$ in a typical zirconium refining process.

While converting zircalloy from a waste material to recyclable $ZrCl_4$, the Collins et al Documents assumed that byproduct chloride salts from their process would be waste subject to disposal. The difference with the implementations of the present disclosure is the assumption that the chemical decladding byproduct chloride salt(s) are preferably to be combined with salts chlorinated from the SNF rod/pin contents freed during mechanical decladding. The material flow diagram of FIG. 28 thus shows the outputting of the heretofore waste chemical decladding byproduct chloride salts, 2834, along with mechanically-freed SNF fuel solids, 2822, to a subsequent SNF chlorination process input stream, 2840.

Most SNF dismantling and decladding operation developers assume cover and off-gases from dismantling and decladding operations to be waste product. The overall process of FIG. 26 assumes containment and packaging of all captured gases, hence the indication in

FIG. 28 that off-gases from both mechanical decladding, 2820, and chemical decladding, 2830, will be output to the input stream of off-gas processing, 2850.

Cover/Off-Gas processing, such as disclosed in the ‘416 Application and/or U.S. Patent Application No. 17/527,862, filed November 16, 2021; and U.S. Patent Application No. 16/415,692, filed May 17, 2019, all of such applications being incorporated in their entirety herein by reference.

The following references, as noted above, are incorporated in their entirety by reference herein:

Collins et al, 2012, “Process Development Studies for Zirconium Recovery/Recycle from Used Nuclear Fuel Cladding,” *Procedia Chemistry*, 7 (2012) 72-76, <https://www.sciencedirect.com/science/article/pii/S1876619612000873>

Collins et al, 2015, “R&D Progress on Recovery/Recycle of Zirconium from Used Fuel Cladding,” USDOE OSTI 1239757, <https://www.osti.gov/servlets/purl/1239757>

Collins et al, 2017, “Engineering-scale Development/Demonstration of Zirconium Recovery from Used Fuel Cladding,” USDOE OSTI 1766433, <https://www.osti.gov/servlets/purl/1766433>

Accounting for fuel salt material is an ongoing challenging to those assuming transfer of molten, thus liquid, fuel salt. Given the powder/grain nature of output of the inventive fuel salt production processes heretofore described, an exemplary implementation of countable solid fuel salt or salt fuel implementation involves use of Selective Laser Melting (SLM) system (not shown) for selectively melting carrier/dilutant salt, fuel salt, and enrichment salt grains/powder, under inert gas, to form countable fuel salt or salt fuel blend ingots. It has been shown that SLM can result in near-zero porosity formation of metal parts. With this as a basis, SLM can melt and fuse together *salt* grains at temperatures lower than those required for metals. The resultant low-porosity formation of salt inherently addresses potential "gassiness" of salt grains resulting from halogenation.

For chain of custody and accounting/countability, each ingot in one exemplary implementation may be coded with a “built-up” or recessed mechanical identifier code or possibly an identifier code pattern comprised of contrasting salts (e.g., QR code comprised of carrier/dilutant vs fuel vs fissile salt). Multi-material SLM are commercially available, so means exist to build multi-material ingots that use material variation for identification coding. Ingots obviously need to be of size sufficient to “host” or otherwise carry a code of some sort.

In another exemplary implementation of a fuel salt ingot, using of a SLM system with a multi-material feed enables precisely positioning of salt components within an ingot. This means very precisely located pockets of fuel salt and fissile salt within an overall diluent salt ingot. With ingots so-formed, issues are less likely to arise with settling during solidification, due to the extremely small molten salt puddle create by SLM and constrained by solidified salt under and/or around and/or adjacent to the material addition point, thereby ensuring the distribution of fuel and fissile salt pockets in the ingot, and thus a stack of ingots, when stored over time. Such very precise and stable three-dimensional positioning and spacing of fissile and fuel salts within each ingot results in better control of potential criticality due to better predictability of mutual proximities of fissile salt grains/pockets/etc. While fissile salt pockets are locked once solidified after deposition, the three-dimensional locations of fissile pockets could be randomized ingot-to-ingot, or those positions could be defined and linked to an ingot-unique code. Lastly, the use of SLM to form the ingots eliminates the need for high-temperature, chloride or fluoride-resistant molds for ingot formation.

An additional challenge in using molten salt is movement and dispensing of hot, molten salt. In an exemplary implementation of an alternative (not illustrated), solid fuel salt ingots may be stacked and loaded into inert gas charged storage tubes, each of which tube fits the cavity of a SNF canister previously occupied by a SNF assembly. A SNF canister loaded in such fashion would contain an array of ingot tubes (ingot tube array) and thus serve as a fuel salt canister, and may be moved to storage or to an MSR, just as canisters are moved today. If the canister is of the horizontal storage type, movement may be by a trailer similar in nature to those for movement of horizontally-stored SNF canisters. Upon need to feed an MSR, the storage cover of the canister may be removed and replaced with a dispensing cover that also contains doors or devices to selectively open inert gas charged storage tubes in the ingot tube array. Such selective opening can be facilitated by closure of the end of each inert gas charged tube by a latched door whose latch may be reached by a mechanism of the dispensing cover. For countable dispensing, if it is assumed that the tractor-trailer for movement of the canister is tilted pitch-rearward, as occurs with full tractor-trailer dump systems, gravity will motivate the ingots to slide toward/to respective tube doors that are at the aft end of the trailer. If an MSR is further assumed to have an ingot-receiving grinder-shredder-mixer, a dispensing cover residing between the ingot tube and the MSR grinder-shredder-mixer may then selectively dispense ingots to the MSR's ingot-receiving grinder-shredder-mixer under gravitational motivation. Nothing precludes the dispensing cover including an indexing mechanism at each tube door to control and count the number of ingots coming out from each selected tube.

Use of a truck dump system eliminates need for a lift on the trailer or other tube maneuvering mechanism to get dry, inert-gas-covered ingots in the ingot tubes to move/slide out of respective ingot tubes. The delivery trailer, in fact, could be a canister trailer, such as a NUHOMS canister trailer carrying a NUHOMS canister (such as provided by Orano USA of Bethesda, MD USA) fitted with a dispensing cover in place of a temporarily-removed canister cover, that a semi-tractor backs up to an MSR grinder-shredder-mixer to deliver ingots. Such a delivery system embodiment avoids removing of, lifting of, and dispensing from ingot tubes one-by-one, and includes provisions for ingot counting to facilitate material transfer accounting.

Use of a tractor-trailer dump system eliminates need for a lift on the trailer or other tube maneuvering mechanism to get dry, inert-gas-covered ingots in the ingot tubes to move/slide out of respective ingot tubes. The delivery trailer, in fact, could be a canister trailer, such as a NUHOMS canister trailer carrying a NUHOMS canister (such as provided by Orano USA of Bethesda, MD USA) fitted with a dispensing control panel in place of a temporarily-removed canister cover, that a semi-tractor backs up to an MSR grinder-shredder-mixer to deliver ingots. Such a delivery system embodiment avoids removing of, lifting of, and dispensing from ingot tubes one-by-one, and includes provisions for ingot counting to facilitate material transfer accounting.

In other aspects of the present disclosure, one overarching aspect is to facilitate the conversion of light water nuclear power plant (NPP) spent nuclear fuel (SNF) rod/pin contents to salt fuel(s) for molten salt reactors, with a primary focus on salt fuels for molten salt fast reactors that can “burn” residual actinides. Achieving the purpose involves the functionalities of fuel assembly dismantling and fuel rod/pin decladding (“dismantling and decladding”), halogenation of oxides to salts, and solidification and packaging of salts, all of which occur with some level cover gas use and off-gas processing. The cover/off-gas processing may operationally include a fractional distillation and condensate recovery/processing stage, which provides a drying function in hydrohalogenation processes and chlorinating agent recovery in carbochlorination processes; and a gas separation and bottling/tanking stage to handle gas components that have market value as salable commodities. The recovery element facilitates re-use of otherwise potentially excess reagents and thus could reduce the quantity and operational cost of fresh reagent associated with SNF halogenation. Separation and bottling of gases for sale further eliminates the need for and utility capital for and operational expense of processing these gases for release to atmosphere. Reduction of waste processing also helps reduce processing facility footprint and thus reduces footprint-related costs. Finally, a SNF-to-salt-fuel facility that uses this approach should be sufficiently compact to reside on an existing NPP site, where it can process SNF from either the NPP or from the on-site Independent Spent Fuel Storage Installation (ISFSI). The ability to process SNF on an existing NPP site could significantly reduce a utility's cost by eliminating the need to transport the SNF offsite over highways and through communities for processing and then potentially back to the NPP for use if, in the ideal case, an MSR is also located on the NPP site. Because an additional site is not required dedicated to salt fuel production using this approach, utilities will save the time, effort, and expense to otherwise procure, prepare, and permit such an additional site.

In one exemplary implementation, a cover/off-gas process includes a fractional distillation and condensate recovery/processing stage (which provides a drying function in hydrohalogenation processes and chlorinating agent recovery in carbochlorination processes) and a gas separation and bottling/tanking stage. Additionally, the overall halogenation and associated cover/off-gas process can be configured in a batch process architecture that does not necessarily require gas separation concurrent with the fractional distillation stage. With this in mind, the efficacy of the fractional distillation and condensate recovery stage using component materials and interfaces can be selected suitable for a particular application. If the context of hydrohalogenation, suitable for SNF conversion to thermal MSR fluoride salt fuels, fractional distillation separation of hydrofluoric acid from the “dry” balance of cover/off-gas and subsequent pressure swing distillation recovery-for-reuse of hydrogen fluoride from fraction distillation hydrofluoric acid condensate could be performed. In the context of carbochlorination, suitable for fast MSR chloride salt fuels, fractional distillation could be performed for recovery-for-reuse of excess carbon tetrachloride and byproduct phosgene (with separation of CO2 from the balance of cover-off-gas to be handled by the gas separation and bottling stage) and could be followed by passage of cover/off-gas and Cl2, from UCl5 reduction to UCl4, to gas separation and bottling. This could, in turn, would be followed by fractional distillation verification that the hydrogen reduction of UCl4 to UCl3 yielded dry hydrogen chloride which does not require azeotrope breaking, and thus can also be subjected to gas separation and bottling, with the foregoing potentially all using the same gas processing system, but in the different modes of operation noted above.

In another exemplary embodiment, cover/off-gas process section implementations with interfaces, materials, and process controls and communications appropriate for repeatable deployments in/on nuclear power plant sites are used. The high-level sections are those for (i) hydrohalic acid recovery from hydrohalogenation cover/off-gas, followed by (ii) pressure swing distillation for separation of water from hydrogen halide, for disposal (of water) and reuse (of hydrogen halide); (iii) condensation, recovery, bottling or tanking for reuse, and actual reuse of phosgene byproduct and excess carbon tetrachloride from carbochlorination cover/off-gas; (iv) chlorine recovery and bottling, as potentially salable product, from reduction of uranium pentachloride byproduct of carbochlorination to uranium tetrachloride; (v) dry hydrogen chloride recovery from cover/off-gas from uranium tetrachloride reduction to uranium trichloride; and (vi) separation and recovery of cover gas from off-gas, for reuse in all halogenation and off-gas processes.

Pictorial and graphic representations herein disclose aspects of the present invention including processes and systems, and alternate implementations of processes and symptoms, for producing fuel or other compositions from unirradiated uranium, unirradiated uranium dioxide fuel, spent, or “used,” nuclear fuel.

Many modifications and other implementations of the disclosure set forth herein will come to mind to one skilled in the art to which this disclosure pertains having the benefit of the teachings presented in the foregoing descriptions and the associated drawings. Therefore, it is to be understood that the disclosure is not to be limited to the specific implementations disclosed and that modifications and other implementations are intended to be included within the scope of the appended claims.

Moreover, although the foregoing descriptions and the associated drawings describe example implementations in the context of certain example combinations of elements and/or functions, it should be appreciated that different combinations of elements and/or functions may be provided by alternative implementations without departing from the scope of the appended claims. In this regard, for example, different combinations of elements and/or functions than those explicitly described above are also contemplated as may be set forth in some of the appended claims. Although specific terms are employed herein, they are used in a generic and descriptive sense only and not for purposes of limitation.

CLAIMS:

1. A method for converting uranium oxides to fuel salts suitable for use in molten salt reactors, the method comprising:

halogenation of the uranium oxides to fuel salt and to byproduct at least one off-gas;

separating of the at least one off-gas into streams including a stream of halogenation agents and a stream of the balance of the at least one off-gas; and

reintroducing the stream of halogenation agents into the halogenation of the uranium oxides.

2. The method of claim 1, further comprising off-gas processing including separating and storing the stream of the balance of the at least one off-gas.

3. The method of claim 1, wherein the halogenation includes use of a cover gas and at least one of anhydrous hydrogen chloride and anhydrous hydrogen fluoride as halogenation agent.

4. The method of claim 3, wherein the halogenation occurs in a bath including a molten salt.

5. The method of claim 4, wherein the halogenation includes:

halogenation of spent nuclear fuel having fission products;

condensing vapors of fission product metals and halogenation salts; and

returning the condensed vapors to the bath.

6. The method of claim 5, further comprising outputting to the off-gas processing the cover gas and the at least one off-gas.

7. The method of claim 4, wherein the halogenation includes:

condensing vapors of halogenation salts; and

returning the condensed vapors to the bath.

8. The method of claim 7, further comprising outputting to the off-gas processing the cover gas and the at least one off-gas.

9. The method of claim 3, wherein the separating of the at least one off-gas further comprises:

separating hydrohalic acid from the balance of the at least one off-gas;

separating the hydrohalic acid to water and dry hydrogen halide; and

recirculating the dry hydrogen halide to the halogenation or storing the dry hydrogen halide.

10. The method of claim 3, wherein the halogenation includes use of anhydrous hydrogen fluoride in at least one fluidized bed.

11. The method of claim 10, wherein the halogenation includes:

halogenation of spent nuclear fuel having fission products;

condensing vapors of fission product metals and halogenation salts; and

returning the condensed vapors to the at least one fluidized bed.

12. The method of claim 11, further comprising outputting to the off-gas processing the cover gas and the at least one off-gas.

13. The method of claim 10, wherein the halogenation includes:

condensing vapors of fission product metals and halogenation salts; and

returning the condensed vapors to the fluidized bed.

14. The method of claim 13, further comprising outputting to the off-gas processing the cover gas and the at least one off-gas.

15. The method of claim 3, wherein the separating of the at least one off-gas further comprises:

separating hydrofluoric acid from the balance of the at least one off-gas;

separating the hydrohalic acid to water and dry hydrogen fluoride; and

recirculating the dry hydrogen halide to the halogenation or storing the dry hydrogen halide.

16. The method of claim 1, wherein the halogenation process occurs under an inert gas cover and comprises:

a first chlorination of the uranium oxides to fuel salts through use of phosgene recovered from a prior chlorination operation;

subsequent to the first chlorination, a second chlorination of the uranium oxides to fuel salts through use of carbon tetrachloride recovered from a prior chlorination operation;

subsequent to the second chlorination, a third chlorination of uranium oxides to fuel salts through use of fresh carbon tetrachloride;

subsequent to the third chlorination, reduction of uranium pentachloride, if any, to salts of substantially uranium tetrachloride by heating in inert gas; and

subsequent to the reduction, addition of the uranium tetrachloride from the uranium pentachloride reduction process to the uranium tetrachloride from at least one of the first chlorination process, the second chlorination process, and the third chlorination process.

17. The method of claim 16, further comprising reduction of salts of substantially uranium tetrachloride to salts of substantially uranium trichloride by heating with hydrogen.

18. The method claim 16, wherein the uranium oxides are from spent nuclear fuel having fission products, and further comprising:

condensing vapors of the fission product metals and chlorination salts; and

returning the condensed vapors to at least one of the first chlorination, the second chlorination, and the third chlorination.

19. The method of claim 18, further comprising outputting to the off-gas processing the cover gas and the at least one off-gas.

20. The method of claim 16, wherein the separating of the at least one off-gas further comprises:

separating carbon tetrachloride and phosgene from the balance of the at least one off-gas; and

recirculating or storing the carbon tetrachloride and the phosgene.

21. The method of claim 2, wherein the process for separation of balance of at least one off-gas includes a cryogenic distillation process that removes oxygen and ozone from the balance of the at least one off-gas.

22. The method of claim 16, wherein the inert cover gas of the halogenation process is carbon dioxide and the process for separation of the balance of the at least one off-gases involves use of absorption in liquid carbon dioxide.

23. The method of claim 1, further comprising:

packing the fuel salts in sealed, dry-cover-gas-charged storage containers for later processing.

24. The method of claim 1, further comprising:

blending the fuel salts with at least one of a dry carrier and dilutant salt; and

storing the fuel salts after blending in at least one of granular form, powder form, and solid form, wherein the solid form is configured to be in countable units and includes at least one of countable ingots, blocks, bricks, ingots, cylinders, semi-cylinders, rods, and spheres, and combinations of one or more of the blocks, said bricks, said ingots, said cylinders, said semi-cylinders, said rods, or said spheres.

25. The method of claim 24, further comprising milling, grinding, shredding, or reducing the solid form of the fuel salts into at least one of the powder form and the granular form.

26. The method of claim 24, further comprising formation of the solid form into countable unitary solid form blocks by selective laser melting of the fuel salts.

27. The method of claim 26, further comprising:

the uranium oxides are from spent nuclear fuel removed from spent nuclear fuel storage containers or casks; and

packaging of the blocks in sealed, inert-gas-charged compartments in the spent nuclear fuel storage containers or casks.

28. The method of claim 1, further comprising enriching the fuel salts.

29. The method of claim 1, further comprising halogenation of thorium oxide or adding thorium halide to the fuel salts to affect the addition of fertile salt.

30. The method of claim 24, further comprising:

transporting at least one of the countable units to a molten salt reactor;

reducing the at least one of the countable units into at least one of the powder form and the granular form by at least one of milling, grinding, and shredding; and

introducing the at least one of the powder form and the granular form of the at least one countable units into the molten salt reactor.

31. A method for converting uranium oxides to fuel salts suitable for use in molten salt reactors substantially as herein described.

32. A system for converting uranium oxides to fuel salts suitable for use in molten salt reactors, the system comprising:

means for halogenation of the uranium oxides to fuel salt and to byproduct at least one off-gas;

means for separating of the at least one off-gas into streams including a stream of halogenation agents and a stream of the balance of the at least one off-gas; and

means for reintroducing the stream of halogenation agents into the halogenation of the uranium oxides.

33. The system of claim 32, further comprising:

means for off-gas processing including separating; and

means for storing the stream of the balance of the at least one off-gas.

34. The system of claim 32, wherein the means for halogenation includes a molten salt bath.

35. The system of claim 34, wherein the uranium oxides are from spent nuclear fuel and further comprising:

means for condensing vapors of fission product metals and halogenation salts; and

means for returning the condensed vapors to the molten salt bath.

36. The system of claim 34, further comprising:

means for condensing vapors of halogenation salts; and

means for returning the condensed vapors to the molten salt bath.

37. The system of claim 32, wherein the means for halogenation includes use of a cover gas and at least one of anhydrous hydrogen chloride and anhydrous hydrogen fluoride as halogenation agent, and further comprising:

means for separating hydrohalic acid from the balance of the at least one off-gas;

separating the hydrohalic acid to water and dry hydrogen halide; and

recirculating the dry hydrogen halide to the halogenation or storing the dry hydrogen halide.

38. The system of claim 32, wherein the means for halogenation includes use of a cover gas and at least one of anhydrous hydrogen chloride and anhydrous hydrogen fluoride as halogenation agent, and wherein the means for halogenation includes use of anhydrous hydrogen fluoride in at least one fluidized bed.

39. The system of claim 32, wherein:

the uranium oxides are from spent nuclear fuel having fission products;

the means for halogenation includes use of a cover gas and at least one of anhydrous hydrogen chloride and anhydrous hydrogen fluoride as halogenation agent; and

the means for halogenation includes use of anhydrous hydrogen fluoride in at least one fluidized bed.

40. The system of claim 32, wherein the means for halogenation includes use of an inert gas cover and further comprising:

means for a first chlorination of the uranium oxides to fuel salts through use of phosgene recovered from a prior chlorination operation;

means for a second chlorination of the uranium oxides to fuel salts through use of carbon tetrachloride recovered from a prior chlorination operation;

means for a third chlorination of uranium oxides to fuel salts through use of fresh carbon tetrachloride;

means for reduction of uranium pentachloride, if any, to salts of substantially uranium tetrachloride by heating in inert gas; and

means for addition of the uranium tetrachloride from the uranium pentachloride reduction process to the uranium tetrachloride from at least one of the first chlorination process, the second chlorination process, and the third chlorination process.

41. The system claim 40, wherein the uranium oxides are from spent nuclear fuel having fission products, and further comprising:

means for condensing vapors of the fission product metals and chlorination salts; and

means for returning the condensed vapors to at least one of the first chlorination, the second chlorination, and the third chlorination.

42. The system of claim 40, further comprising:

means for separating carbon tetrachloride and phosgene from the balance of the at least one off-gas; and

means for recirculating or storing the carbon tetrachloride and the phosgene.

43. A system of claim 33, further comprising means for cryogenic distillation that removes oxygen and ozone from the balance of the at least one off-gas.

44. A system of claim 40, wherein the inert cover gas of the halogenation process is carbon dioxide and the process for separation of the balance of the at least one off-gases involves use of absorption in liquid carbon dioxide.

45. The system of claim 32, further comprising:

means for packing the fuel salts in sealed, dry-cover-gas-charged storage containers for later processing.

46. The system of claim 32, further comprising:

means for blending the fuel salts with at least one of a dry carrier and dilutant salt; and

means for storing the fuel salts after blending in at least one of granular form, powder form, and solid form, wherein the solid form is configured to be in countable units and includes at least one of countable ingots, blocks, bricks, ingots, cylinders, semi-cylinders, rods, and spheres, and combinations of one or more of the blocks, said bricks, said ingots, said cylinders, said semi-cylinders, said rods, or said spheres.

47. The system of claim 46, further comprising means for milling, grinding, shredding, or reducing the solid form of the fuel salts into at least one of the powder form and the granular form.

48. The system of claim 46, further comprising formation of the solid form into countable unitary solid form blocks by selective laser melting of the fuel salts.

49. The method of claim 48, further comprising:

the uranium oxides are from spent nuclear fuel removed from spent nuclear fuel storage containers or casks; and packaging of the blocks in sealed, inert-gas-charged compartments in the spent nuclear fuel storage containers or casks.

50. The system of claim 32, further comprising means for enriching the fuel salts.

51. The system of claim 46, further comprising:

at least one molten salt reactor;

means transporting at least one of the countable units to the molten salt reactor;

means for reducing the at least one of the countable units into at least one of the powder form and the granular form by at least one of milling, grinding, and shredding; and

means for introducing the at least one of the powder form and the granular form of the at least one countable units into the molten salt reactor.

52. A system for converting uranium oxides to fuel salts suitable for use in molten salt reactors substantially as herein described.

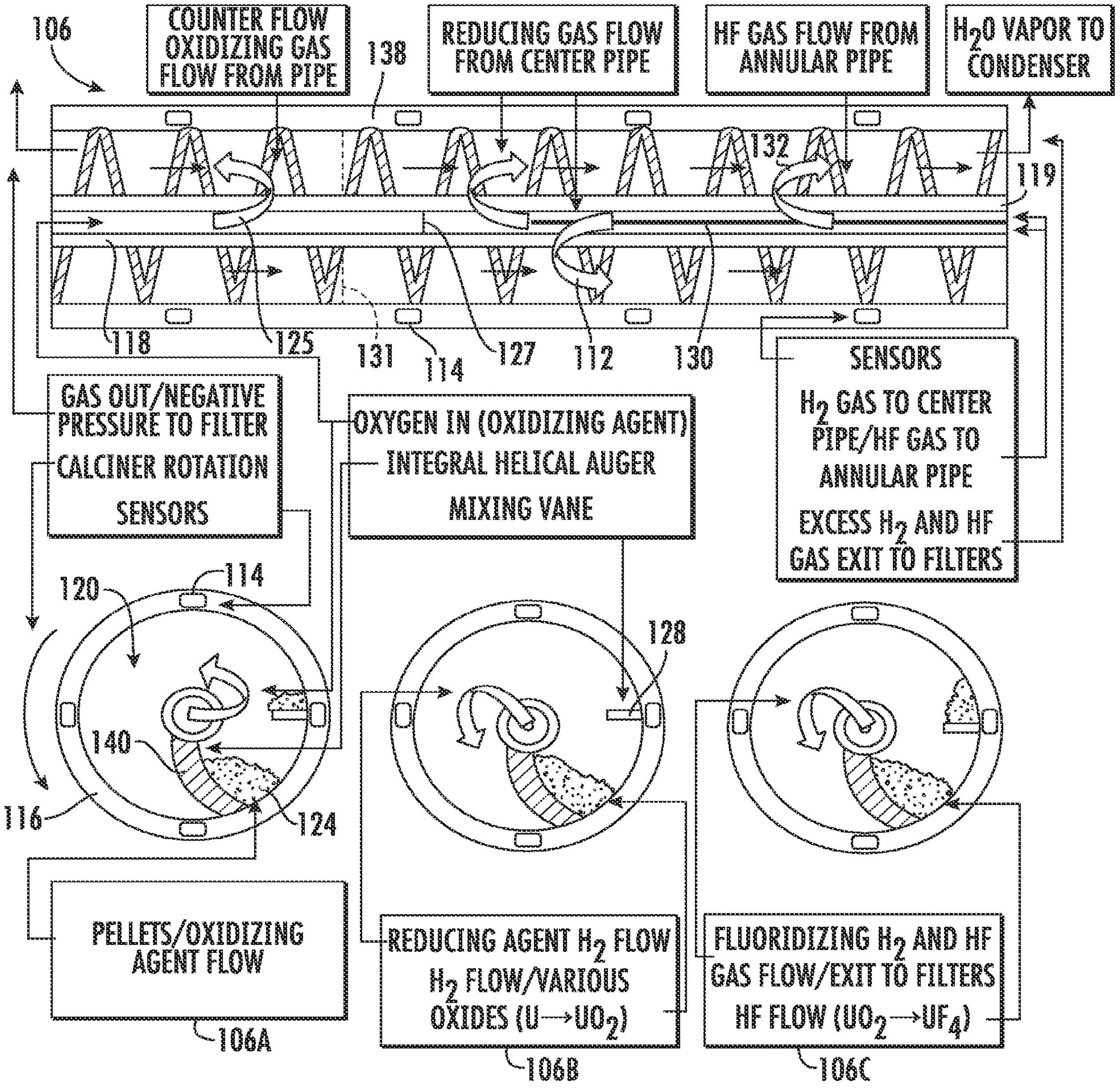
106
COUNTER FLOW OXIDIZING GAS FLOW FROM PIPE
138
REDUCING GAS FLOW FROM CENTER PIPE
HF GAS FLOW FROM ANNULAR PIPE
H_2O VAPOR TO CONDENSER
132
119
118
125
131
114
127
112
130
GAS OUT/NEGATIVE PRESSURE TO FILTER
CALCINER ROTATION
SENSORS
OXYGEN IN (OXIDIZING AGENT)
INTEGRAL HELICAL AUGER
MIXING VANE
SENSORS
H_2 GAS TO CENTER PIPE/HF GAS TO ANNULAR PIPE
EXCESS H_2 AND HF GAS EXIT TO FILTERS
120
114
128
140
116
124
PELLETS/OXIDIZING AGENT FLOW
106A
REDUCING AGENT H_2 FLOW
H_2 FLOW/VARIOUS OXIDES (U→UO_2)
106B
FLUORIDIZING H_2 AND HF GAS FLOW/EXIT TO FILTERS
HF FLOW (UO_2→UF_4)
106C

FIG. 1

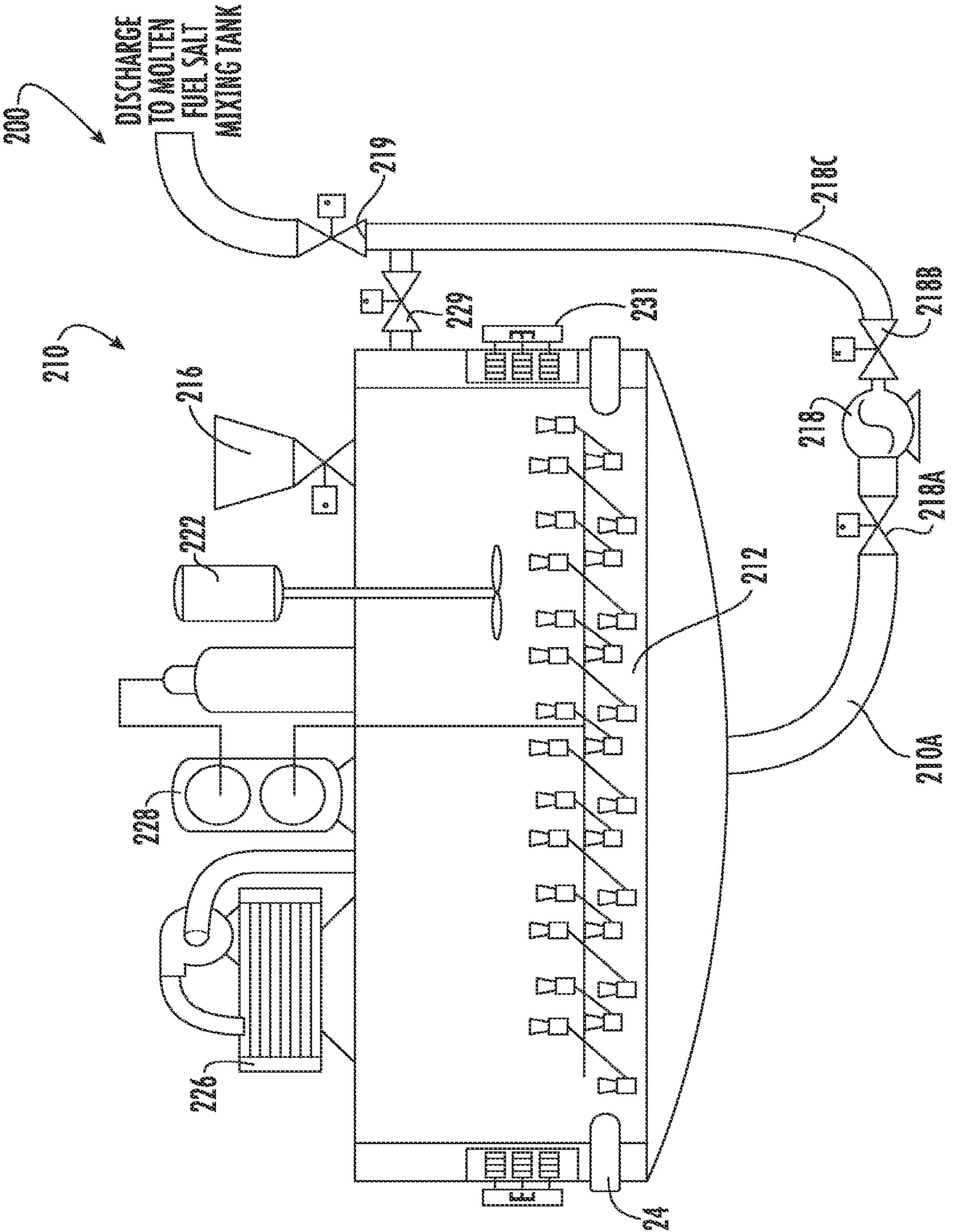
200
DISCHARGE TO MOLTEN FUEL SALT MIXING TANK
219
218C
229
231
218B
210
216
218
218A
222
212
210A
228
226
224

FIG. 2

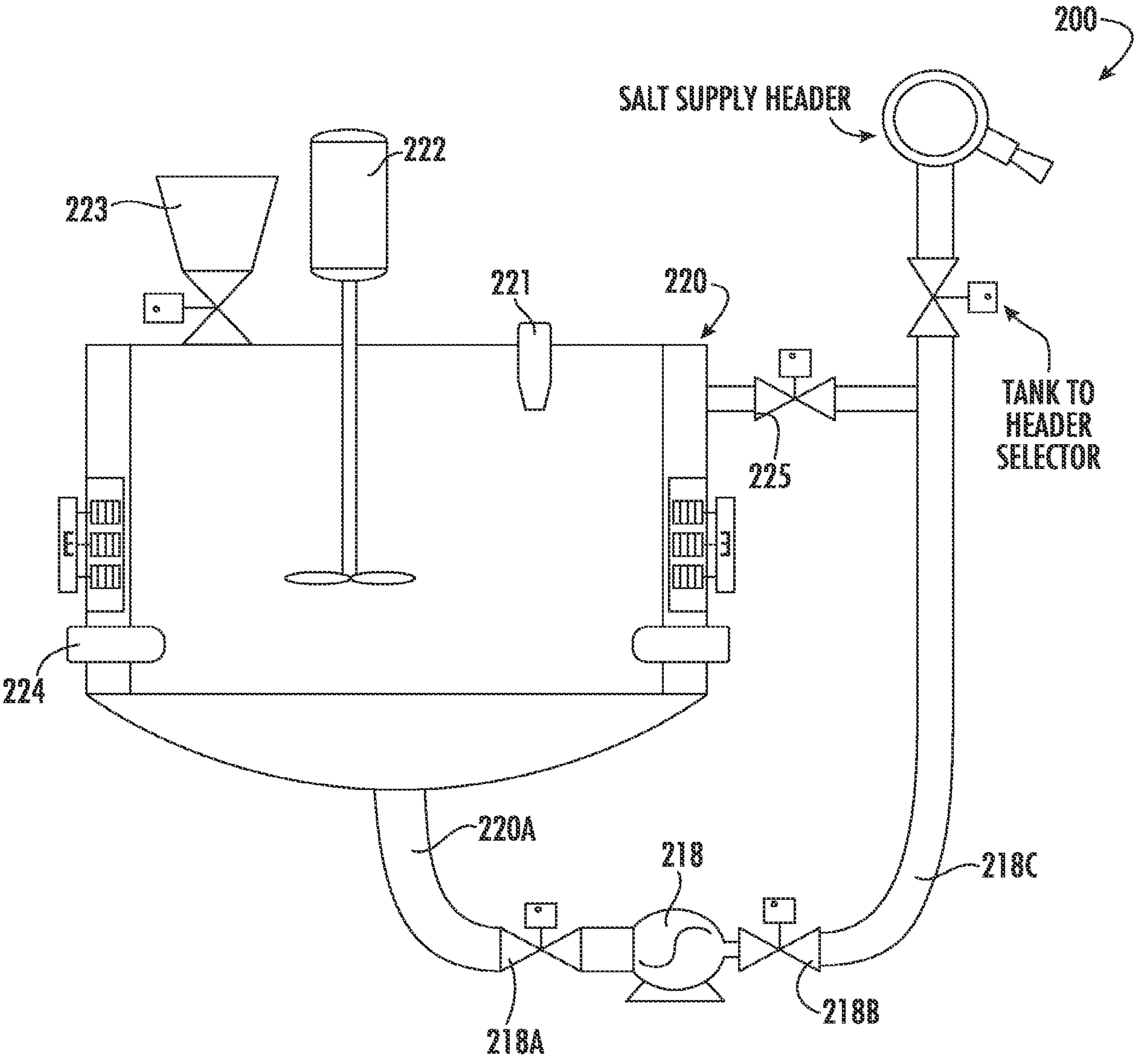
200
SALT SUPPLY HEADER
222
223
221
220
225
TANK TO
HEADER
SELECTOR
224
220A
218
218C
218A
218B

FIG. 3

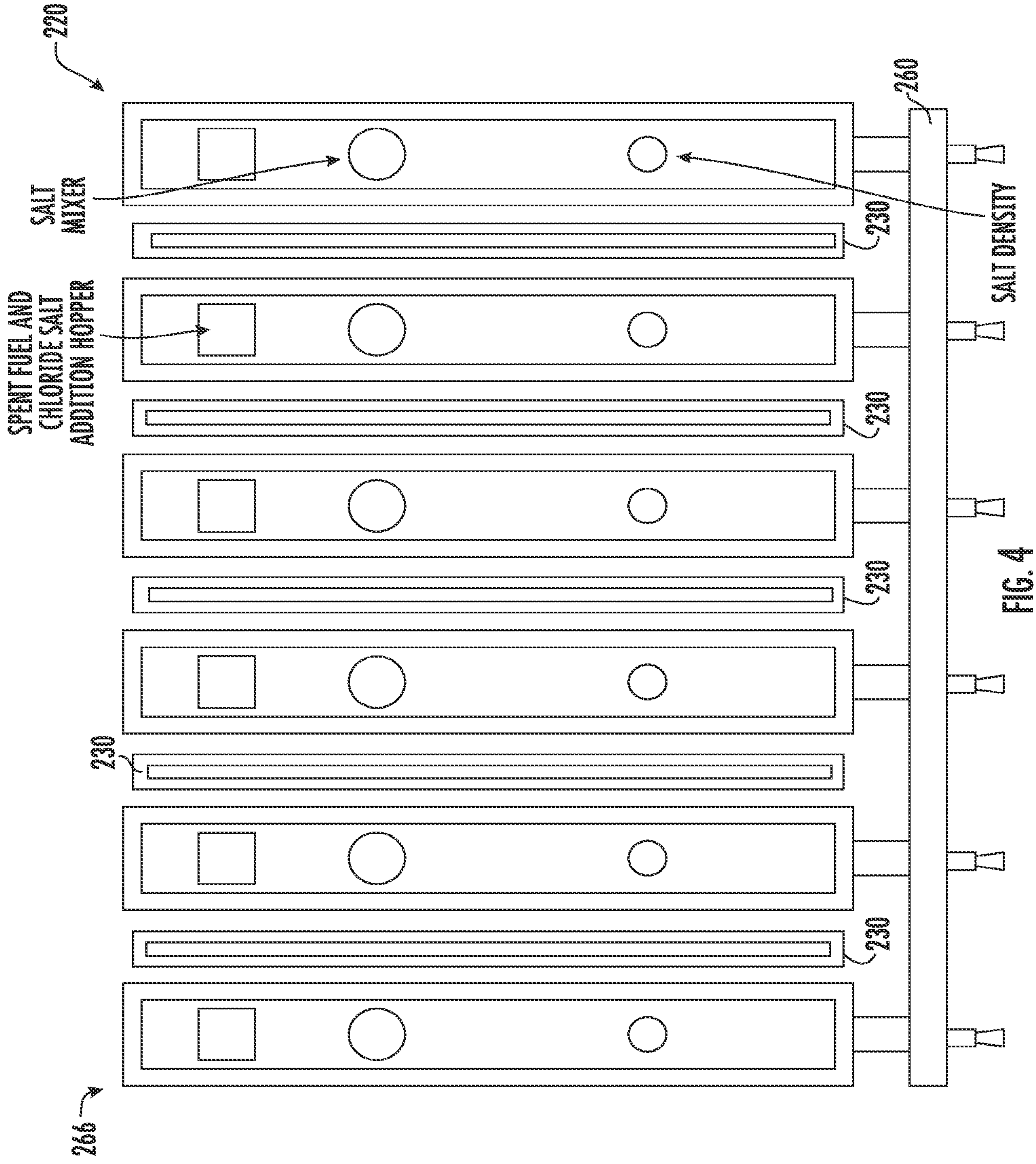
220
SALT MIXER
SPENT FUEL AND CHLORIDE SALT ADDITION HOPPER
SALT DENSITY
260
230
230
230
230
230
265

FIG. 4

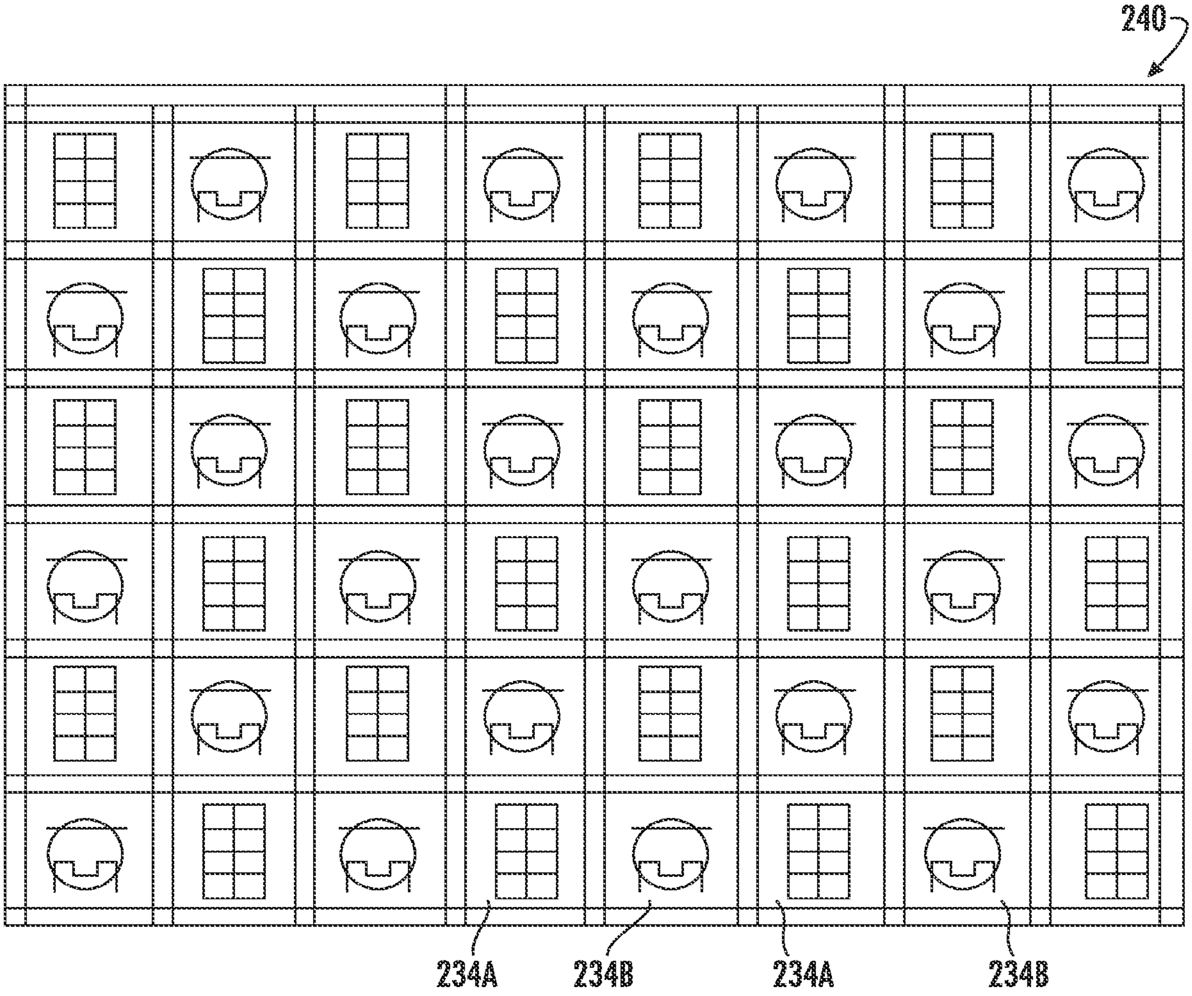

FIG. 5

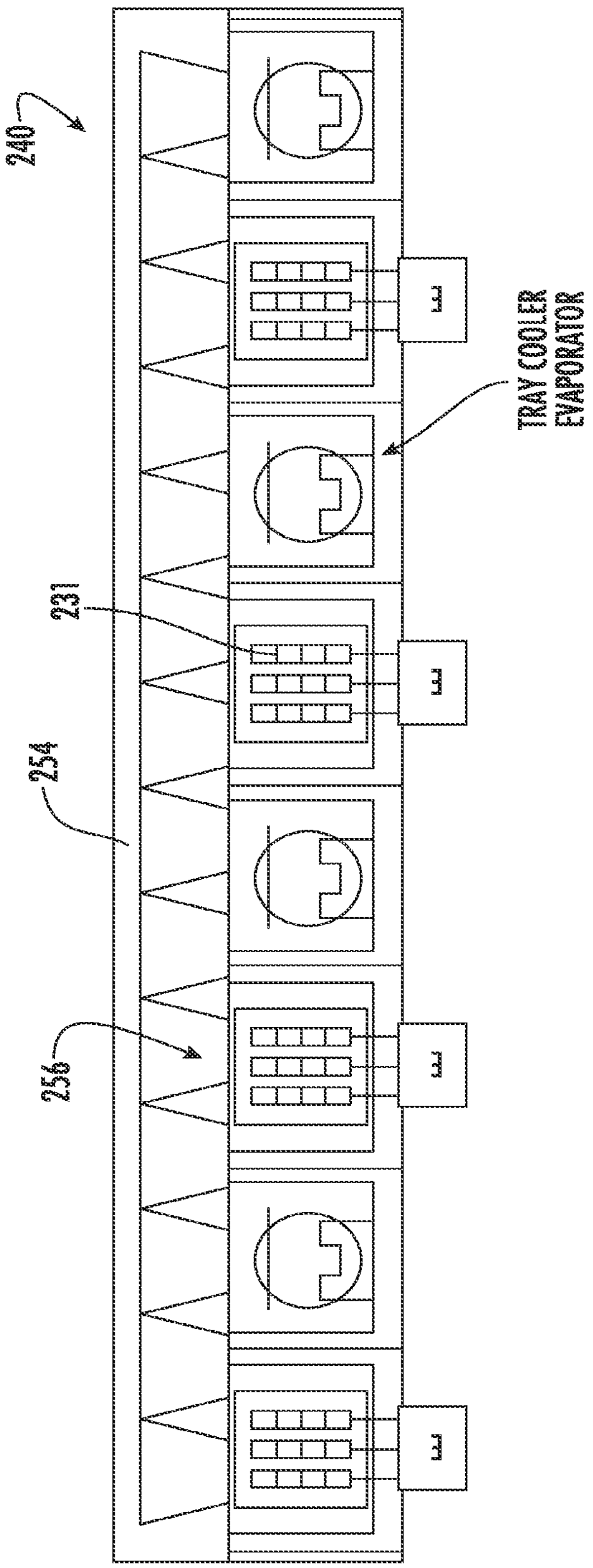

FIG. 6

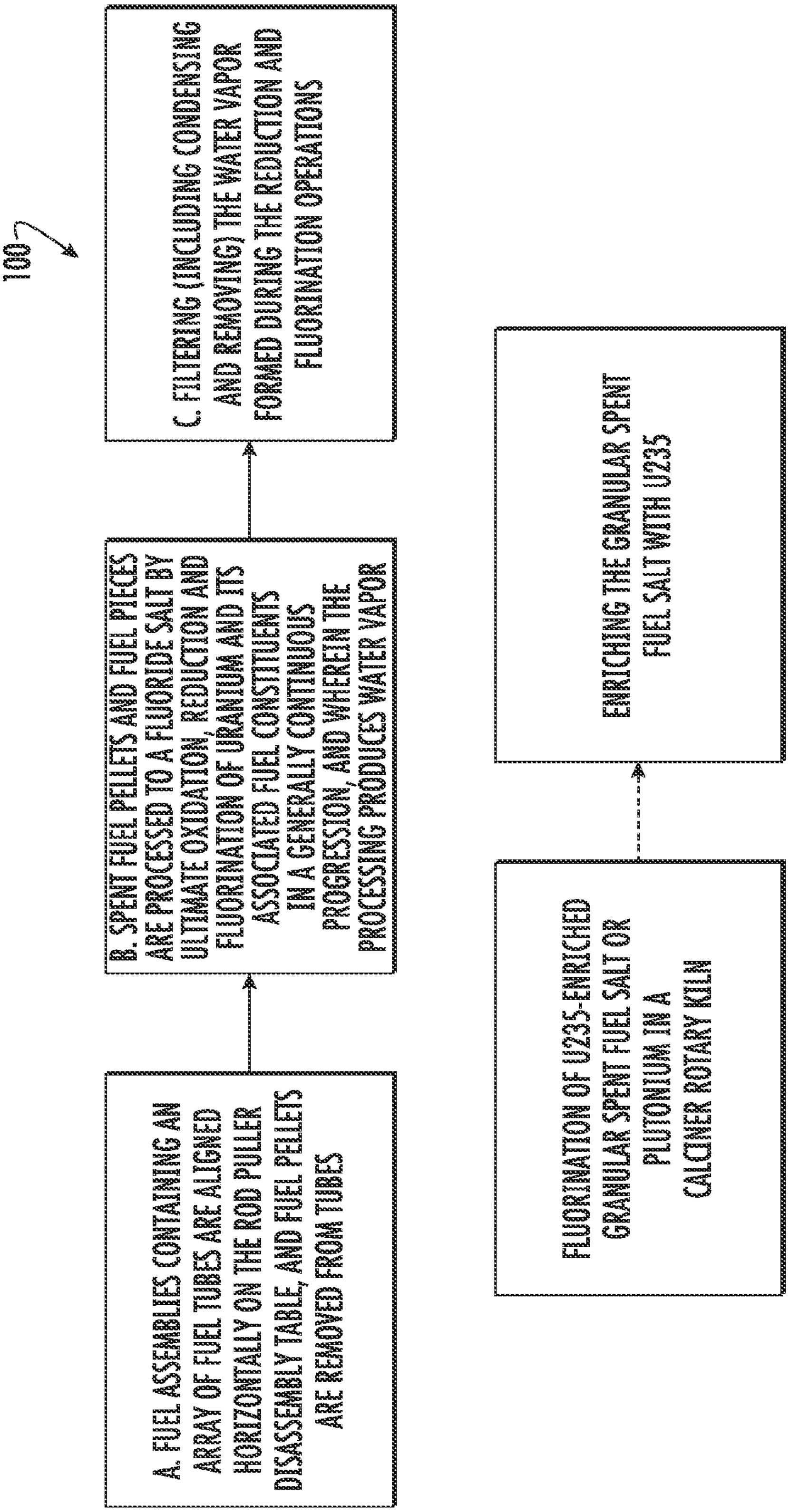
100
A. FUEL ASSEMBLIES CONTAINING AN ARRAY OF FUEL TUBES ARE ALIGNED HORIZONTALLY ON THE ROD PULLER DISASSEMBLY TABLE, AND FUEL PELLETS ARE REMOVED FROM TUBES
B. SPENT FUEL PELLETS AND FUEL PIECES ARE PROCESSED TO A FLUORIDE SALT BY ULTIMATE OXIDATION, REDUCTION AND FLUORINATION OF URANIUM AND ITS ASSOCIATED FUEL CONSTITUENTS IN A GENERALLY CONTINUOUS PROGRESSION, AND WHEREIN THE PROCESSING PRODUCES WATER VAPOR
C. FILTERING (INCLUDING CONDENSING AND REMOVING) THE WATER VAPOR FORMED DURING THE REDUCTION AND FLUORINATION OPERATIONS
FLUORINATION OF U235-ENRICHED GRANULAR SPENT FUEL SALT OR PLUTONIUM IN A CALCINER ROTARY KILN
ENRICHING THE GRANULAR SPENT FUEL SALT WITH U235

FIG. 7

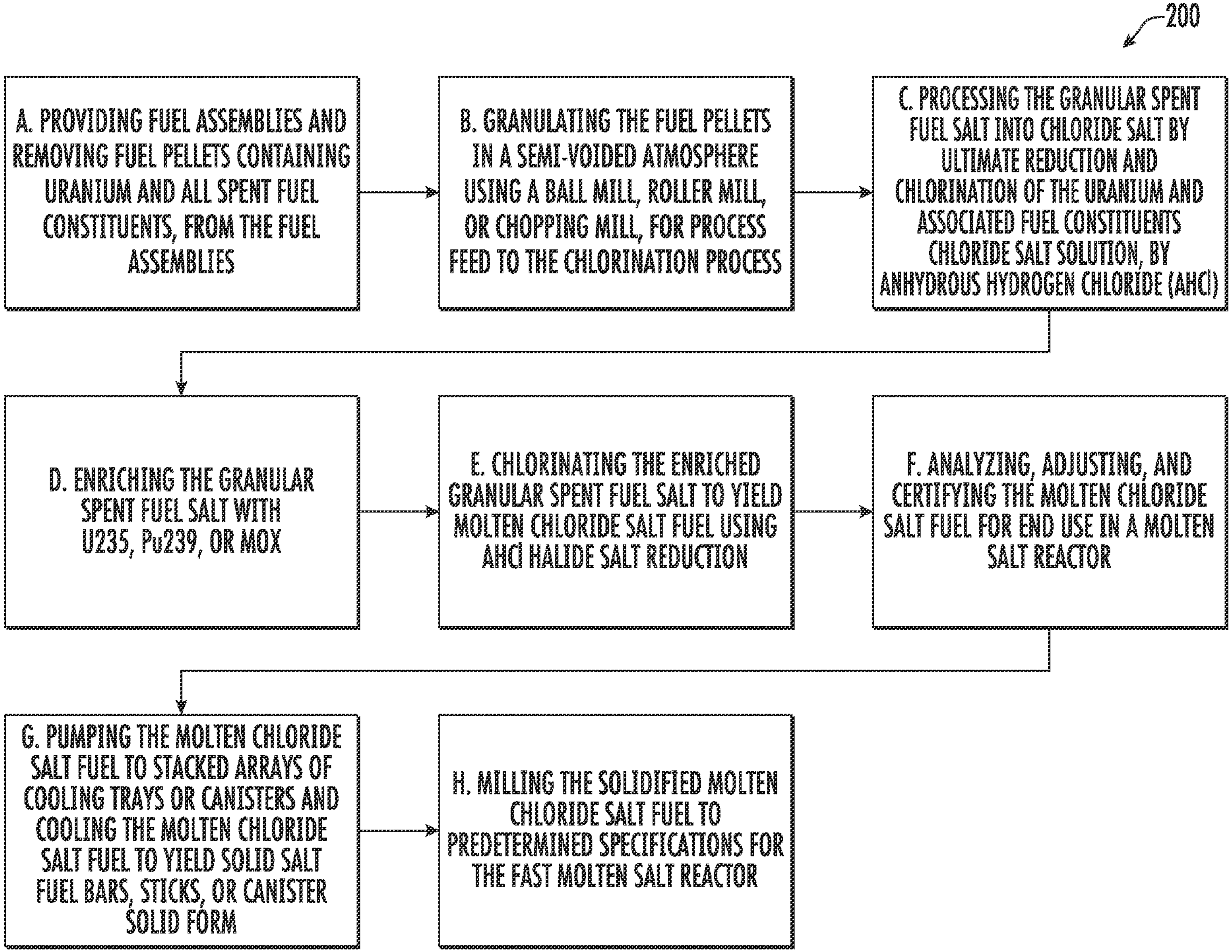
200
A. PROVIDING FUEL ASSEMBLIES AND REMOVING FUEL PELLETS CONTAINING URANIUM AND ALL SPENT FUEL CONSTITUENTS, FROM THE FUEL ASSEMBLIES
B. GRANULATING THE FUEL PELLETS IN A SEMI-VOIDED ATMOSPHERE USING A BALL MILL, ROLLER MILL, OR CHOPPING MILL, FOR PROCESS FEED TO THE CHLORINATION PROCESS
C. PROCESSING THE GRANULAR SPENT FUEL SALT INTO CHLORIDE SALT BY ULTIMATE REDUCTION AND CHLORINATION OF THE URANIUM AND ASSOCIATED FUEL CONSTITUENTS CHLORIDE SALT SOLUTION, BY ANHYDROUS HYDROGEN CHLORIDE (AHCl)
D. ENRICHING THE GRANULAR SPENT FUEL SALT WITH U235, Pu239, OR MOX
E. CHLORINATING THE ENRICHED GRANULAR SPENT FUEL SALT TO YIELD MOLTEN CHLORIDE SALT FUEL USING AHCl HALIDE SALT REDUCTION
F. ANALYZING, ADJUSTING, AND CERTIFYING THE MOLTEN CHLORIDE SALT FUEL FOR END USE IN A MOLTEN SALT REACTOR
G. PUMPING THE MOLTEN CHLORIDE SALT FUEL TO STACKED ARRAYS OF COOLING TRAYS OR CANISTERS AND COOLING THE MOLTEN CHLORIDE SALT FUEL TO YIELD SOLID SALT FUEL BARS, STICKS, OR CANISTER SOLID FORM
H. MILLING THE SOLIDIFIED MOLTEN CHLORIDE SALT FUEL TO PREDETERMINED SPECIFICATIONS FOR THE FAST MOLTEN SALT REACTOR

FIG. 8

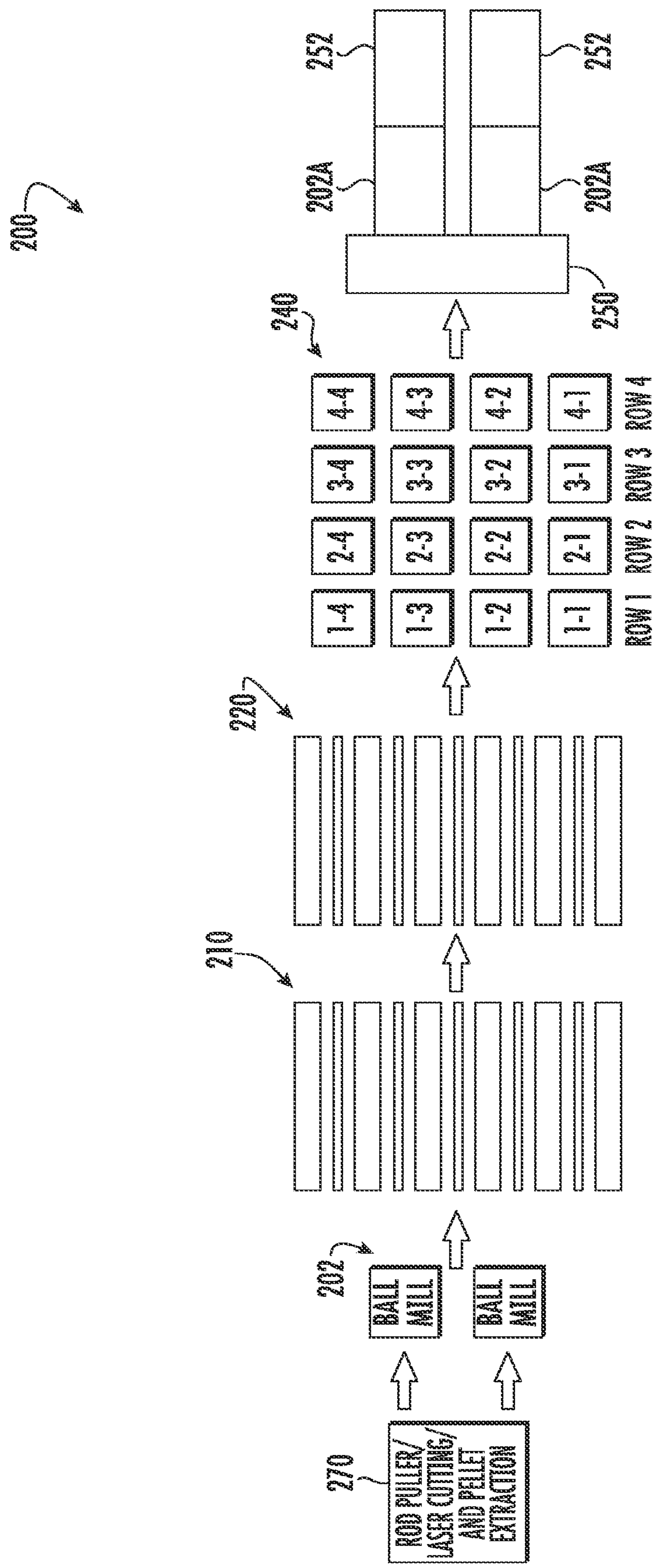
200
270
ROD PULLER/
LASER CUTTING/
AND PELLET
EXTRACTION
202
BALL
MILL
BALL
MILL
210
220
240
1-4
1-3
1-2
1-1
ROW 1
2-4
2-3
2-2
2-1
ROW 2
3-4
3-3
3-2
3-1
ROW 3
4-4
4-3
4-2
4-1
ROW 4
250
202A
202A
252
252

FIG. 9

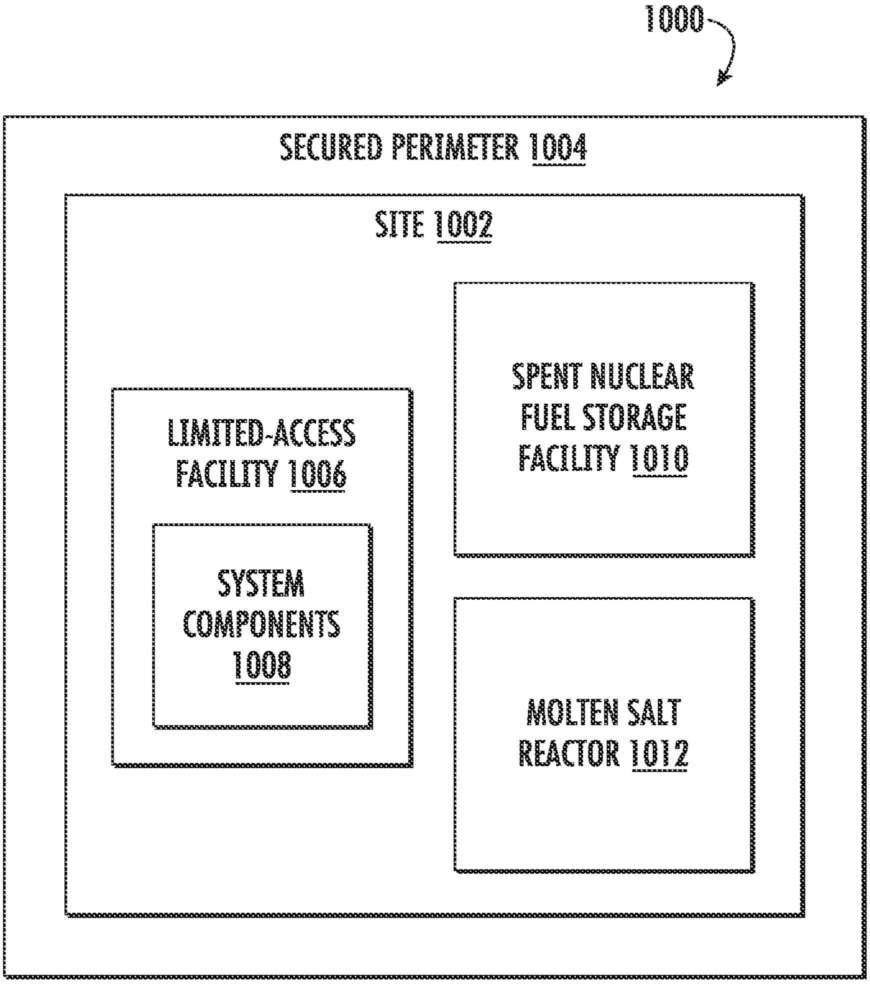
1000
SECURED PERIMETER 1004
SITE 1002
LIMITED-ACCESS FACILITY 1006
SYSTEM COMPONENTS 1008
SPENT NUCLEAR FUEL STORAGE FACILITY 1010
MOLTEN SALT REACTOR 1012

FIG. 10

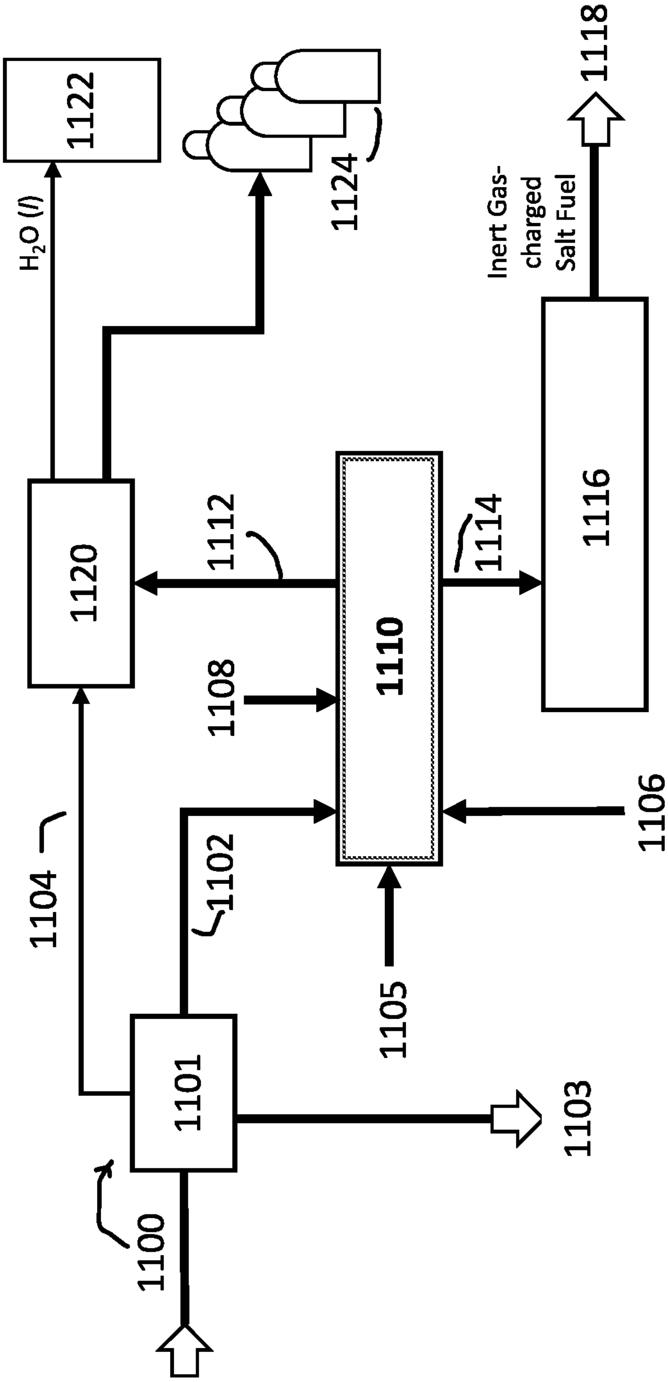
1100
1101
1102
1103
1104
1105
1106
1108
1110
1112
1114
1116
1118
1120
1122
1124
H_2O (*l*)
Inert Gas-charged Salt Fuel

Fig. 11

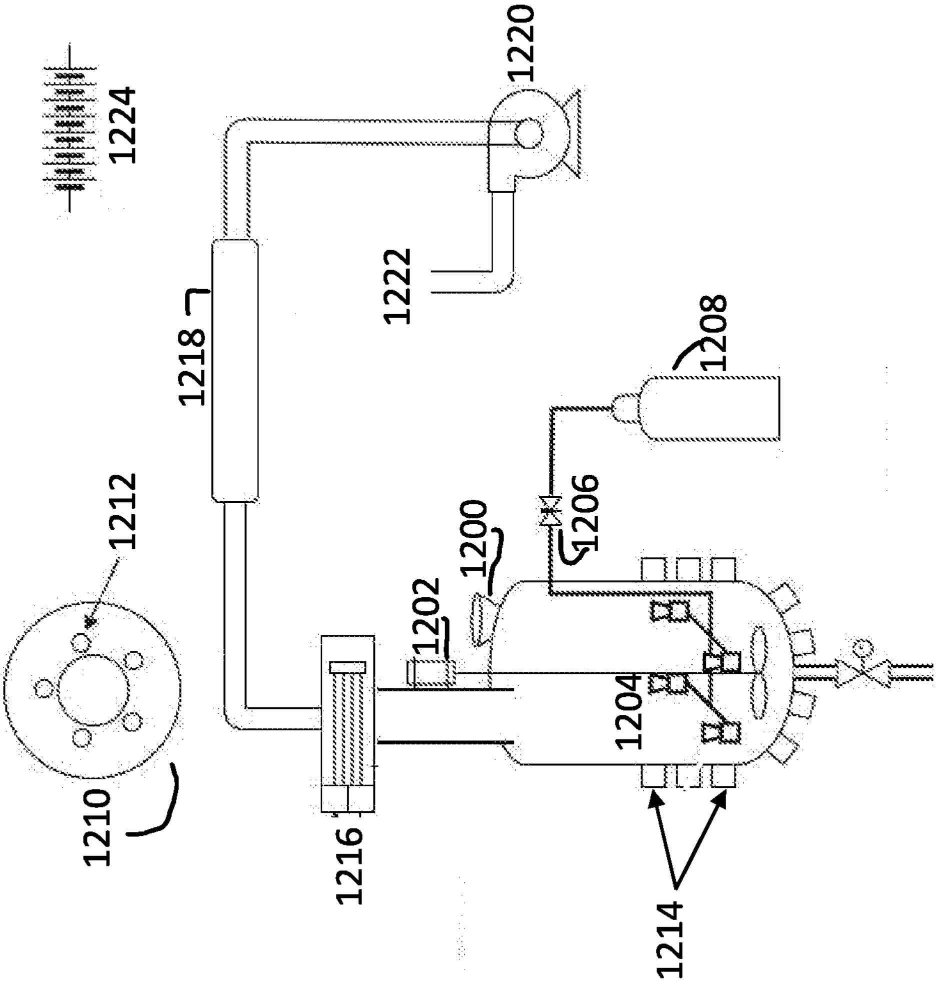
1224
1220
1222
1218
1212
1210
1216
1202
1200
1206
1208
1204
1214

Fig. 12

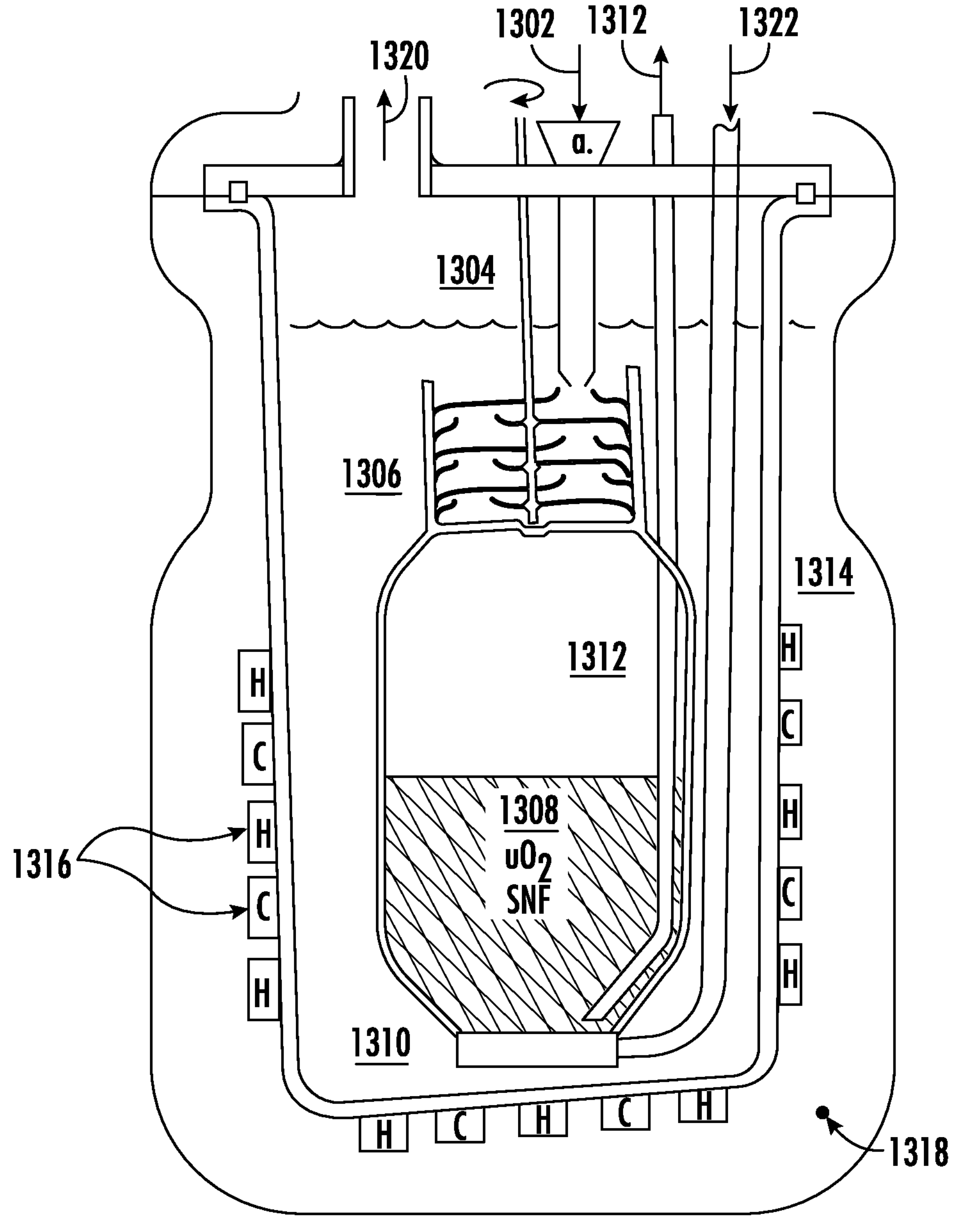
1302
1312
1322
1320
a.
1304
1306
1314
1312
H
C
H
C
H
1308
uO2
SNF
1316
H
C
H
C
H
1310
H
C
H
C
H
1318

FIG. 13

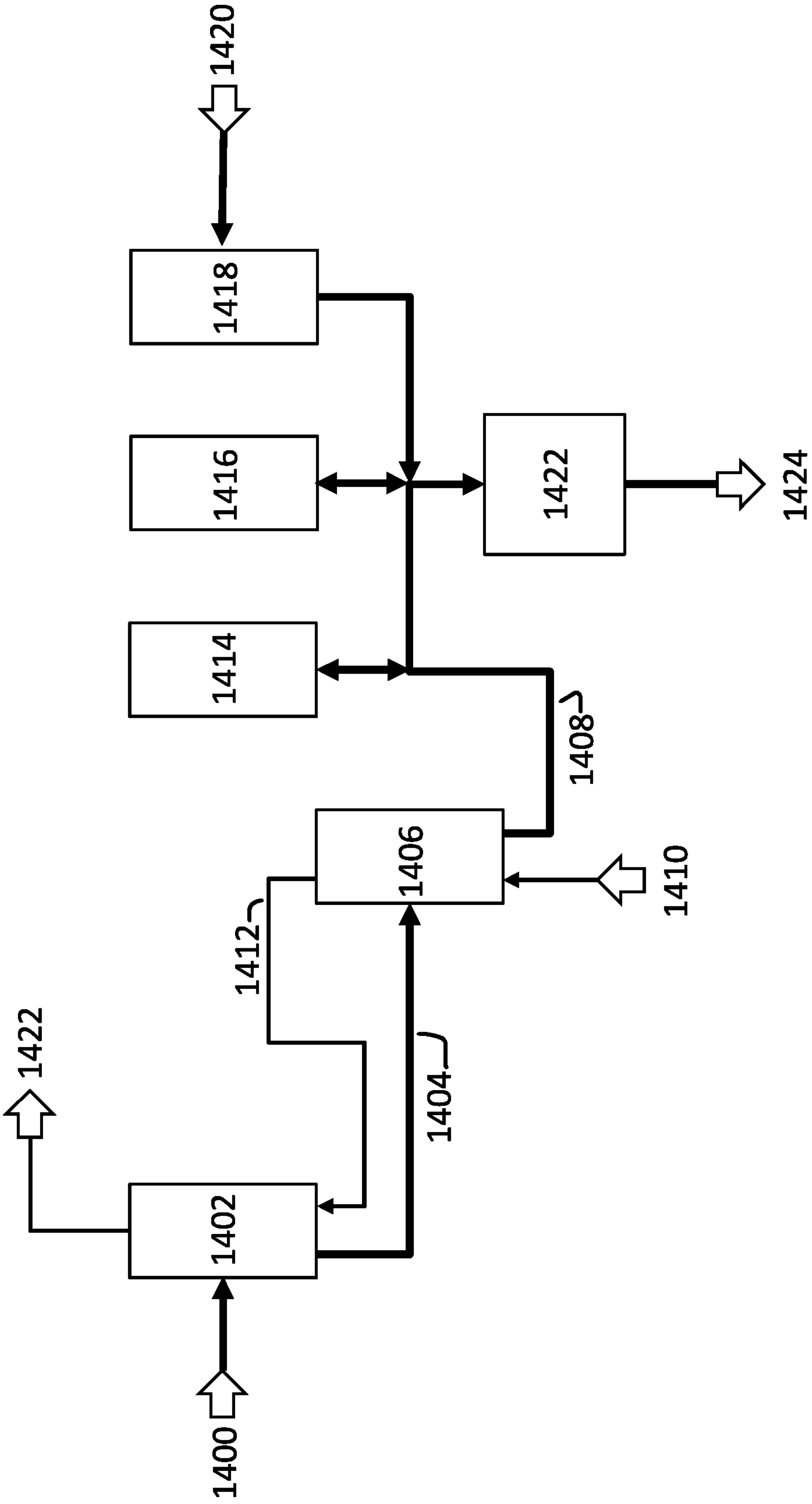
1400
1402
1422
1404
1412
1406
1410
1408
1414
1416
1418
1420
1422
1424

Fig. 14

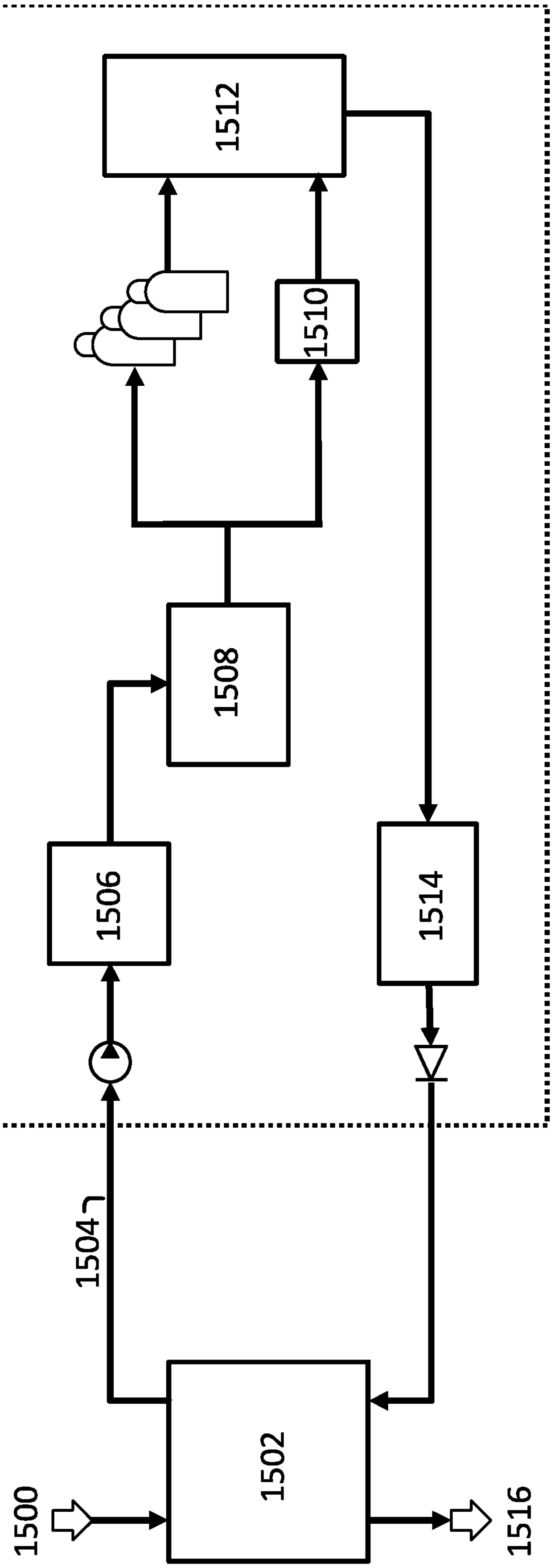

Fig. 15

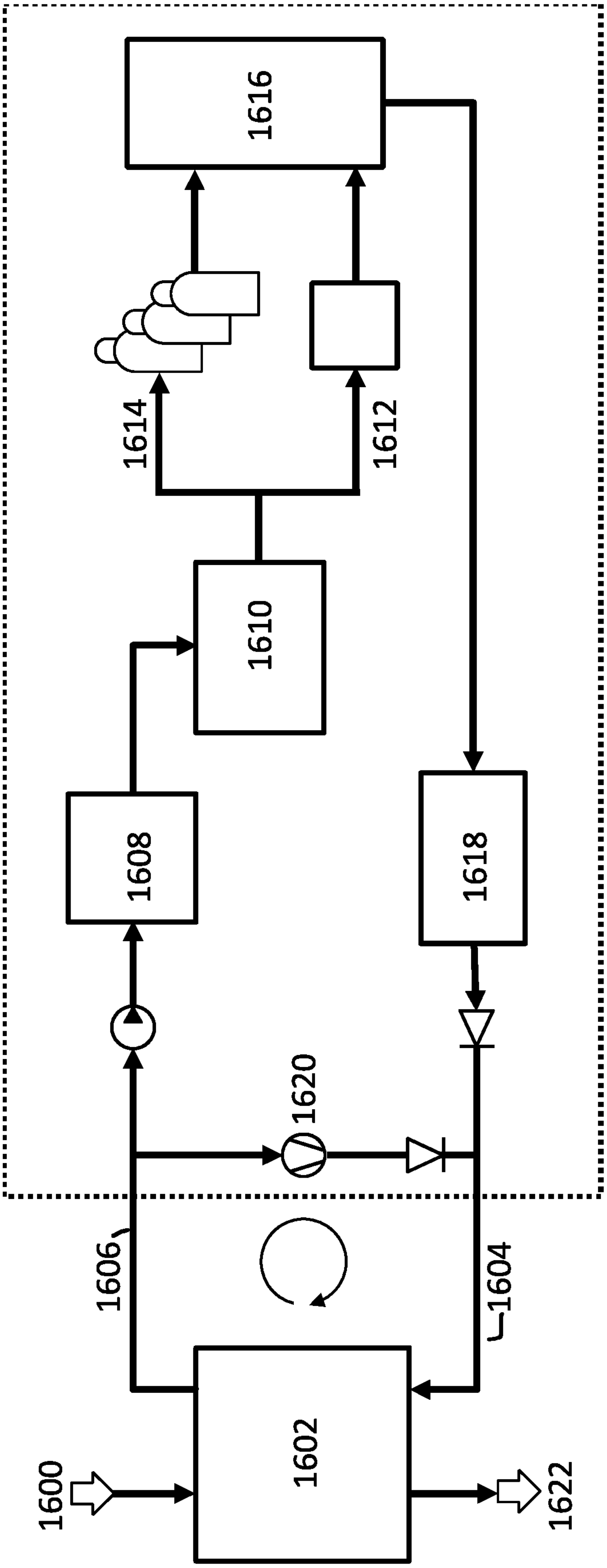

Fig. 16

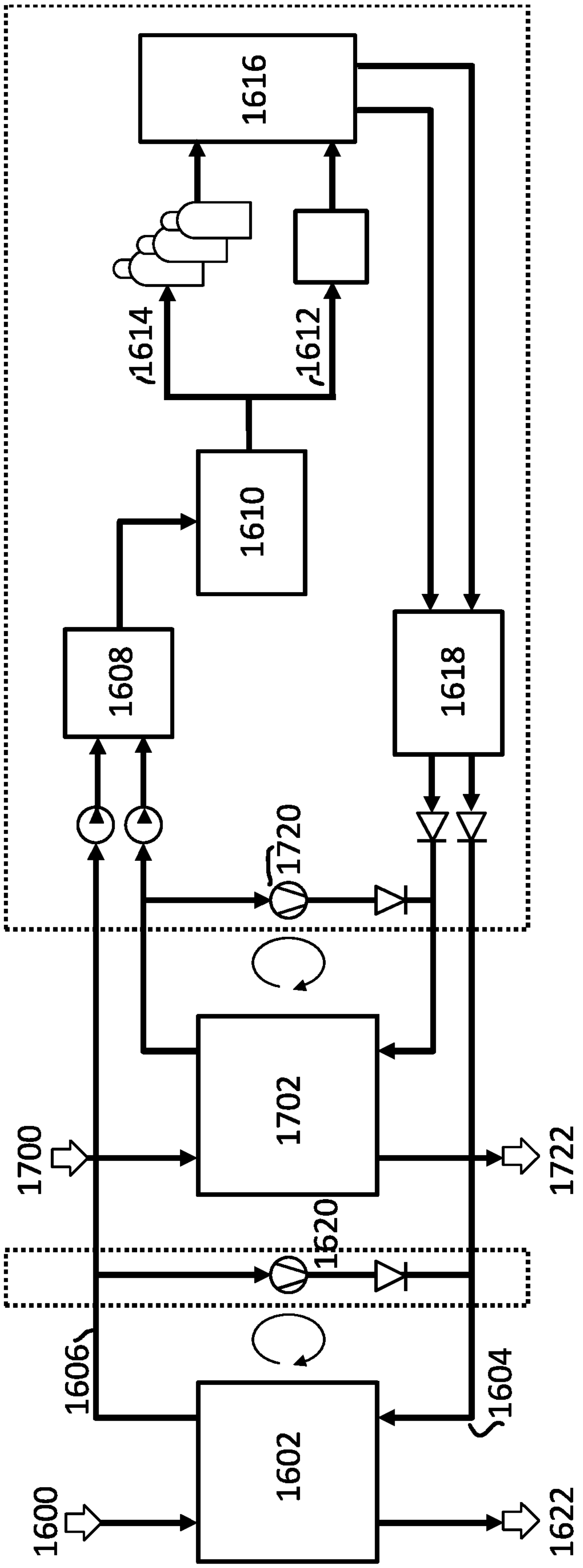
1616
1614
1612
1610
1608
1618
1720
1702
1700
1722
1620
1606
1604
1602
1600
1622

Fig. 17

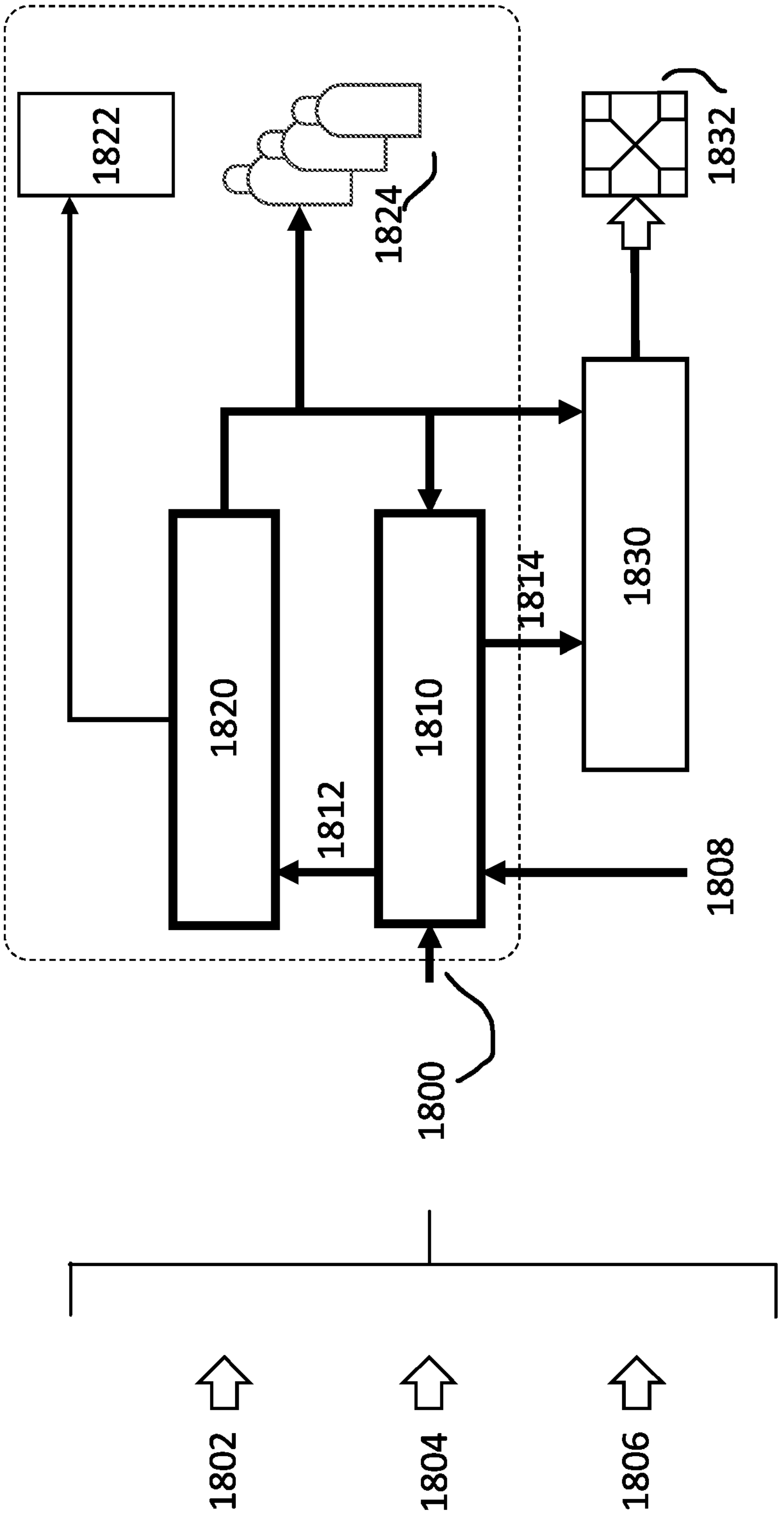
1822
1824
1832
1820
1810
1830
1814
1812
1808
1800
1802
1804
1806

Fig. 18

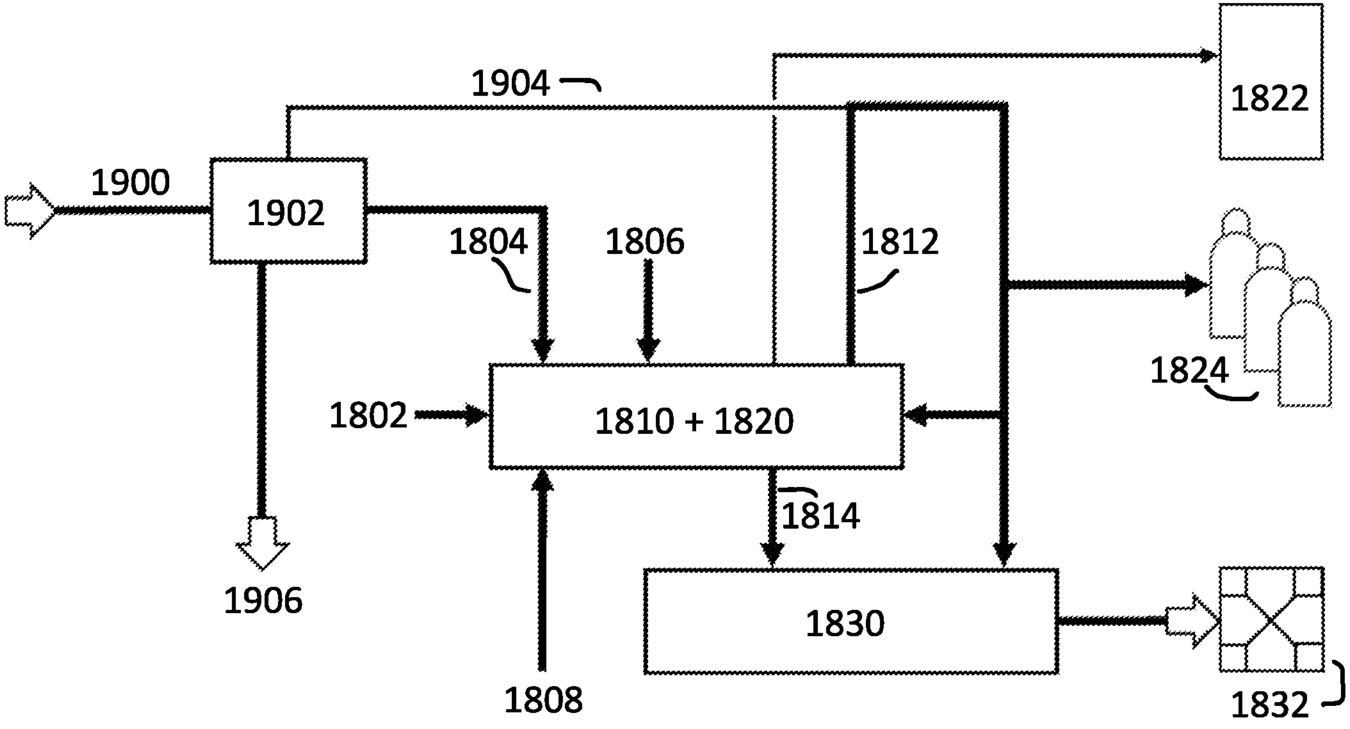
1904
1822
1900
1902
1804
1806
1812
1824
1802
1810 + 1820
1814
1906
1830
1808
1832

Fig. 19

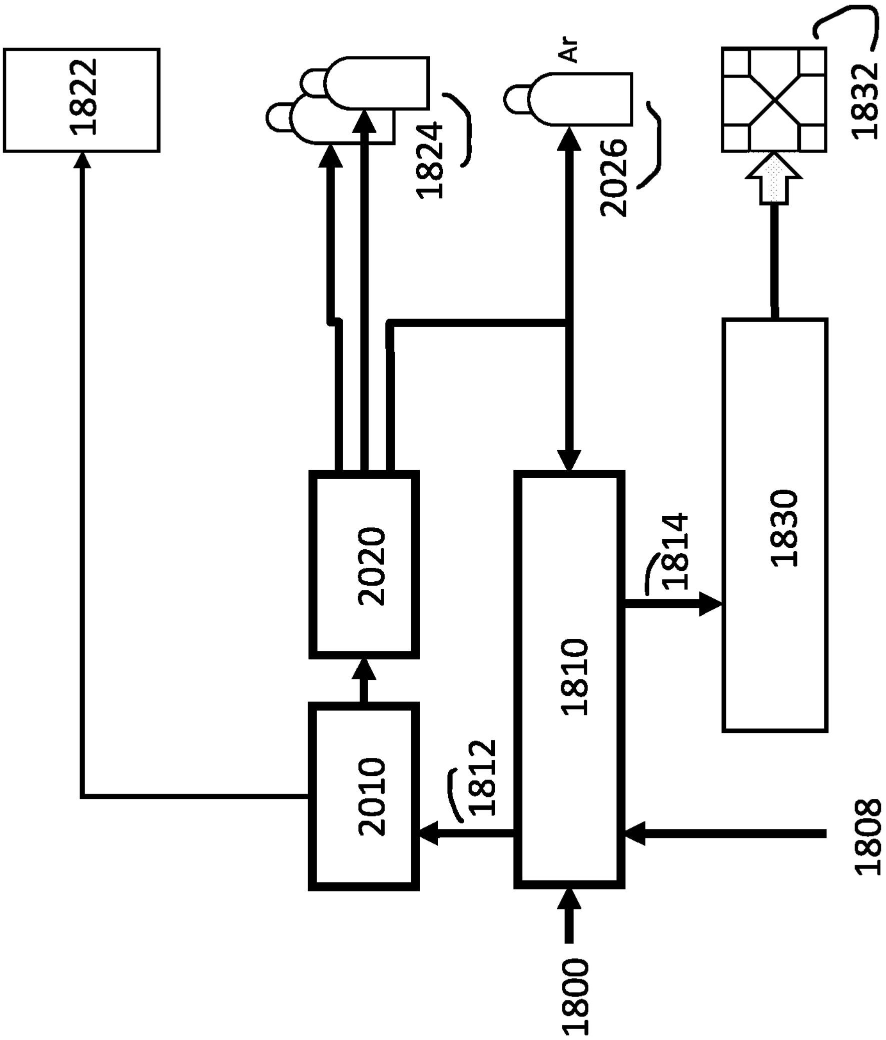
1822
1824
Ar
2026
1832
2020
2010
1812
1810
1814
1830
1800
1808

Fig. 20

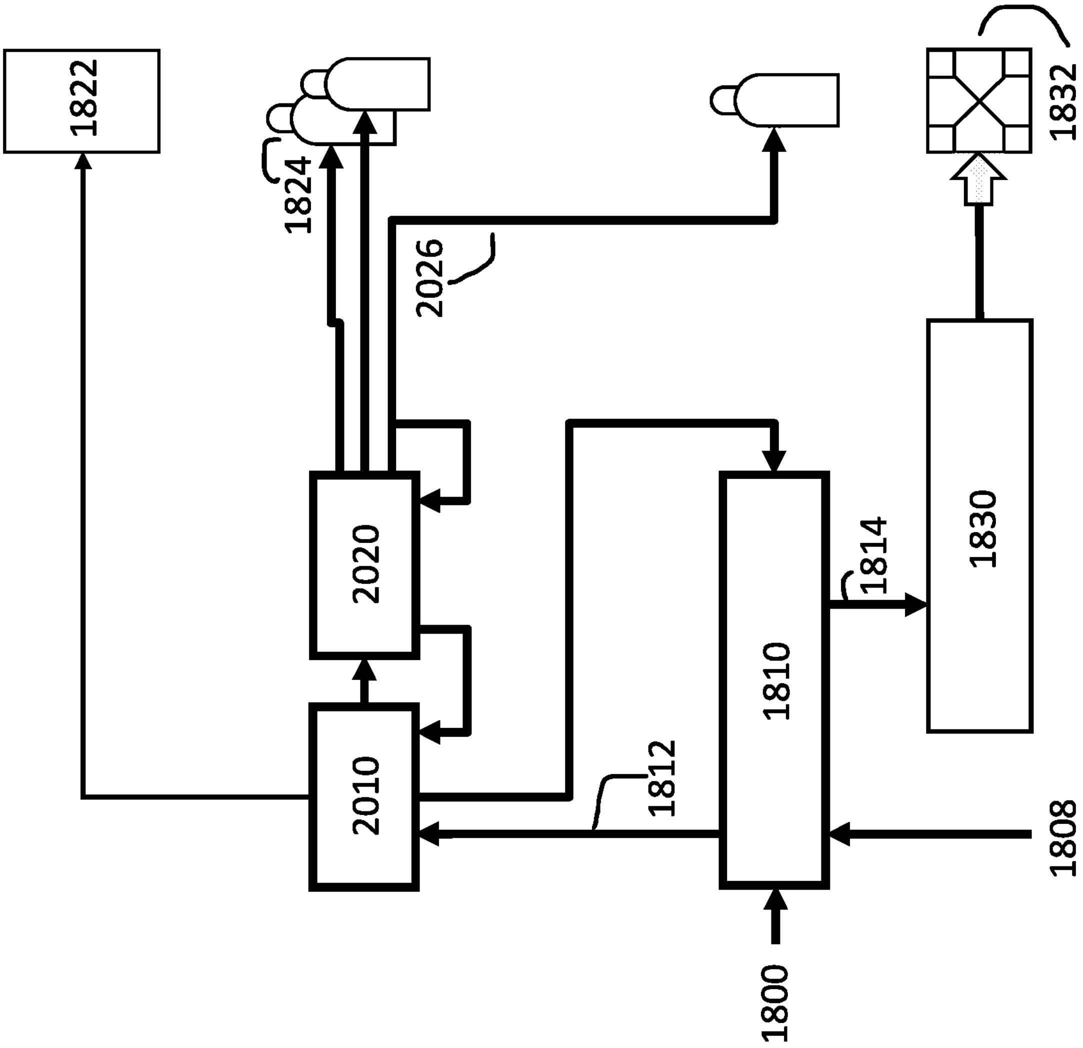

Fig. 21

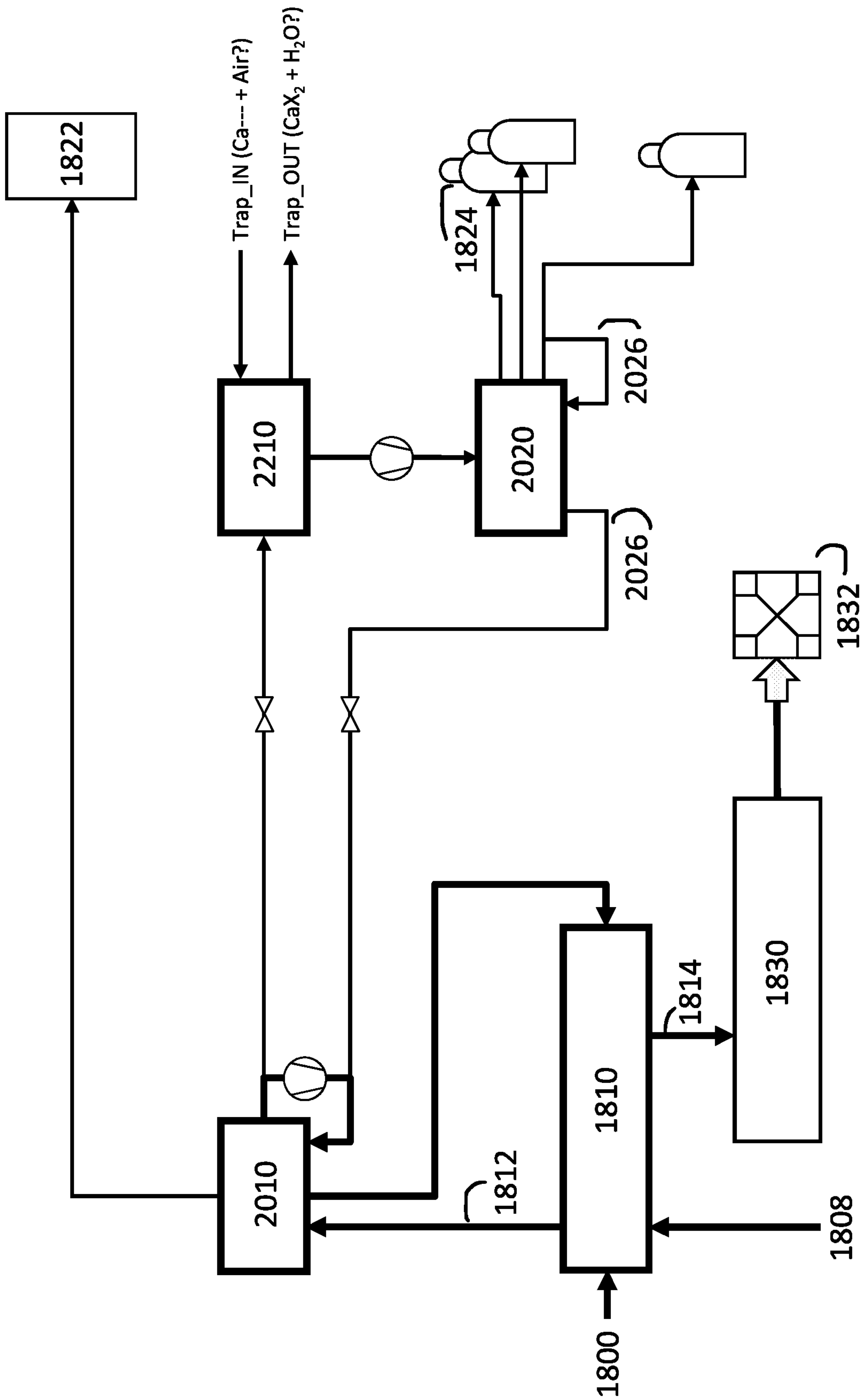
1822
Trap_IN (Ca--- + Air?)
Trap_OUT (CaX$_2$ + H$_2$O?)
1824
2210
2020
2026
2026
1832
1830
1814
1810
2010
1812
1808
1800

Fig. 22

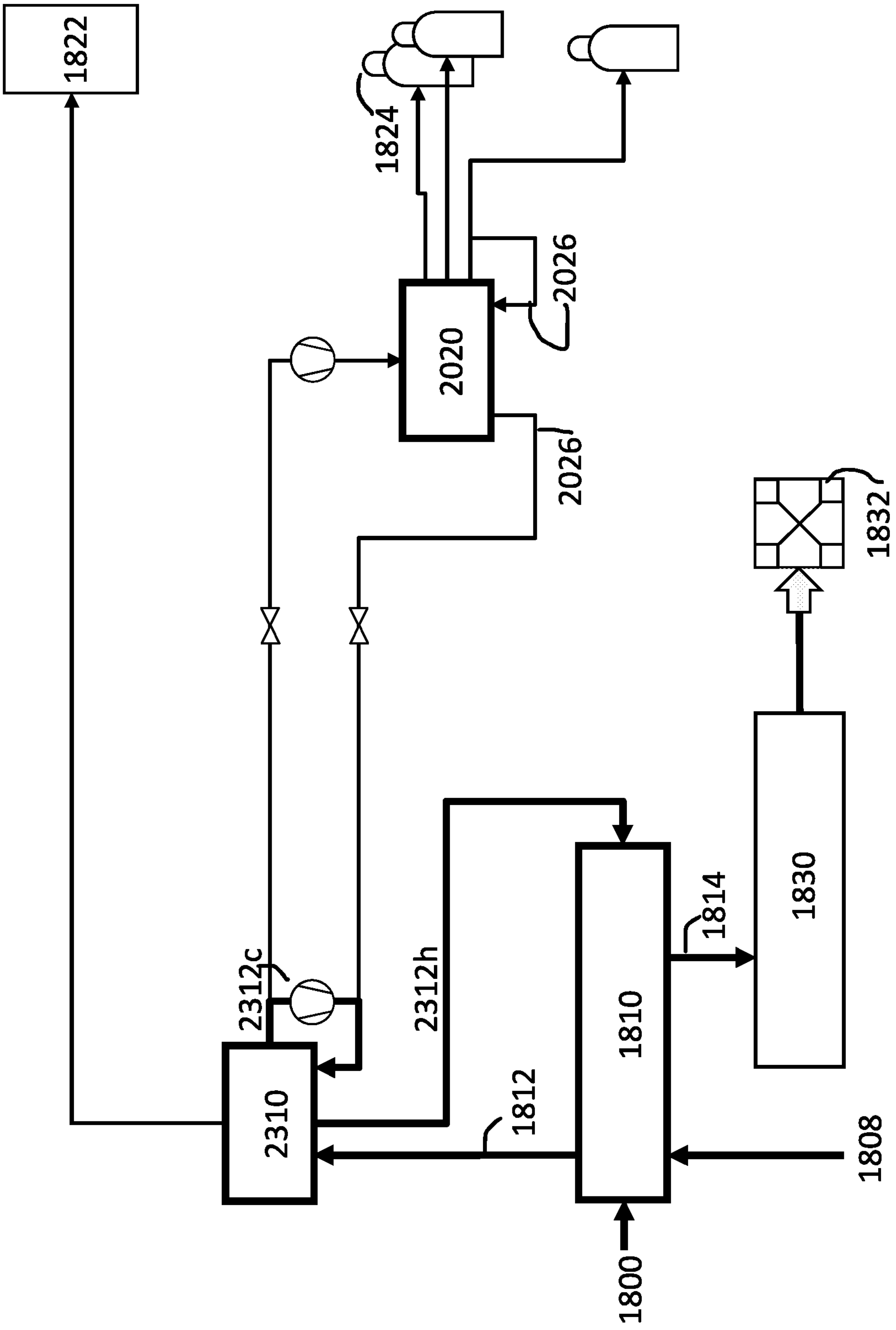
1822
1824
2020
2026
2026
1832
2312c
2310
2312h
1812
1810
1814
1830
1808
1800

Fig. 23

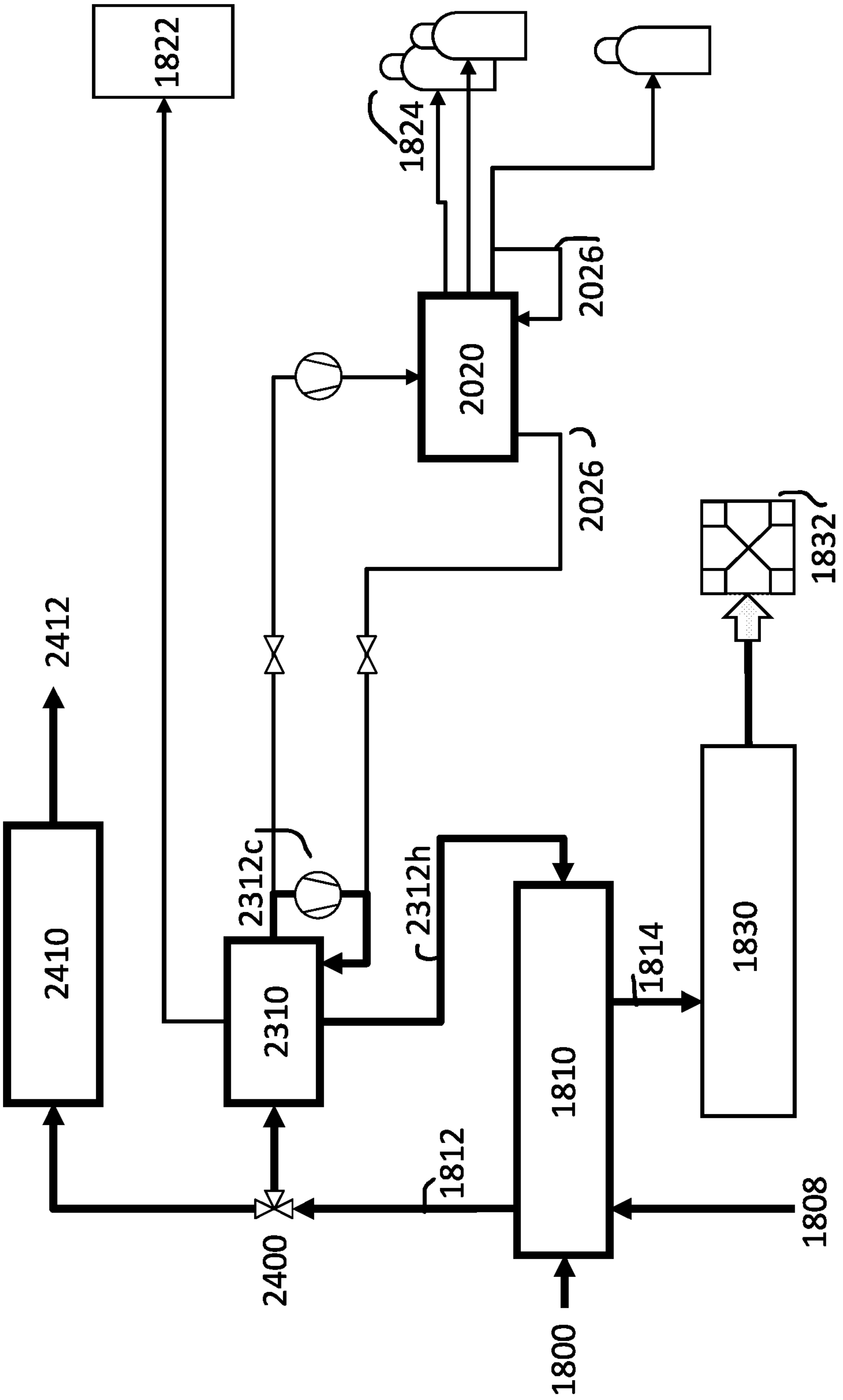
2412
2410
1822
1824
2026
2026
2020
2312c
2312h
2310
1810
1812
2400
1800
1808
1814
1830
1832

Fig. 24

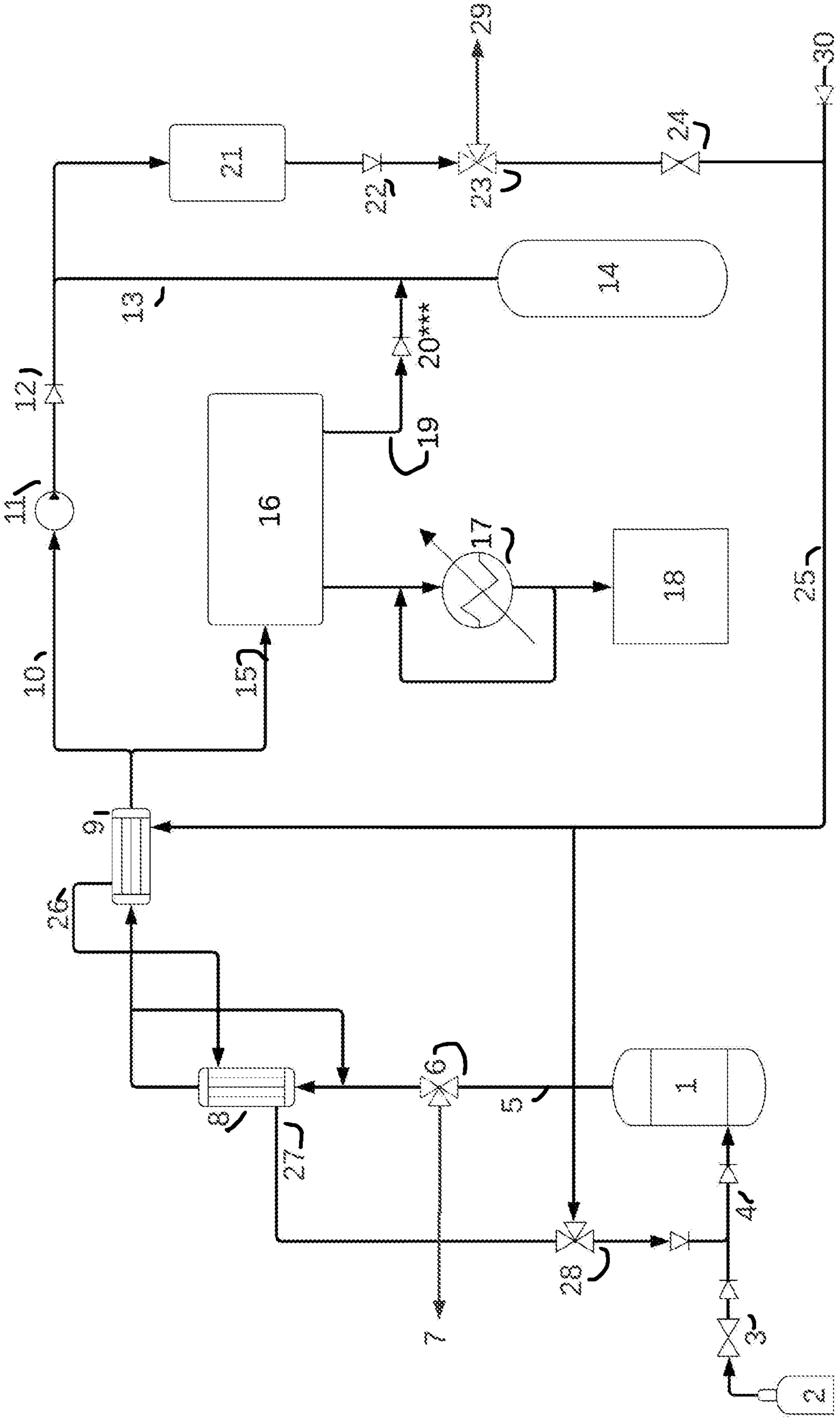
29
30
21
22
23
24
14
13
20***
12
11
16
19
17
18
25
10
15
9
26
8
6
5
1
27
4
28
7
3
2

Fig. 25

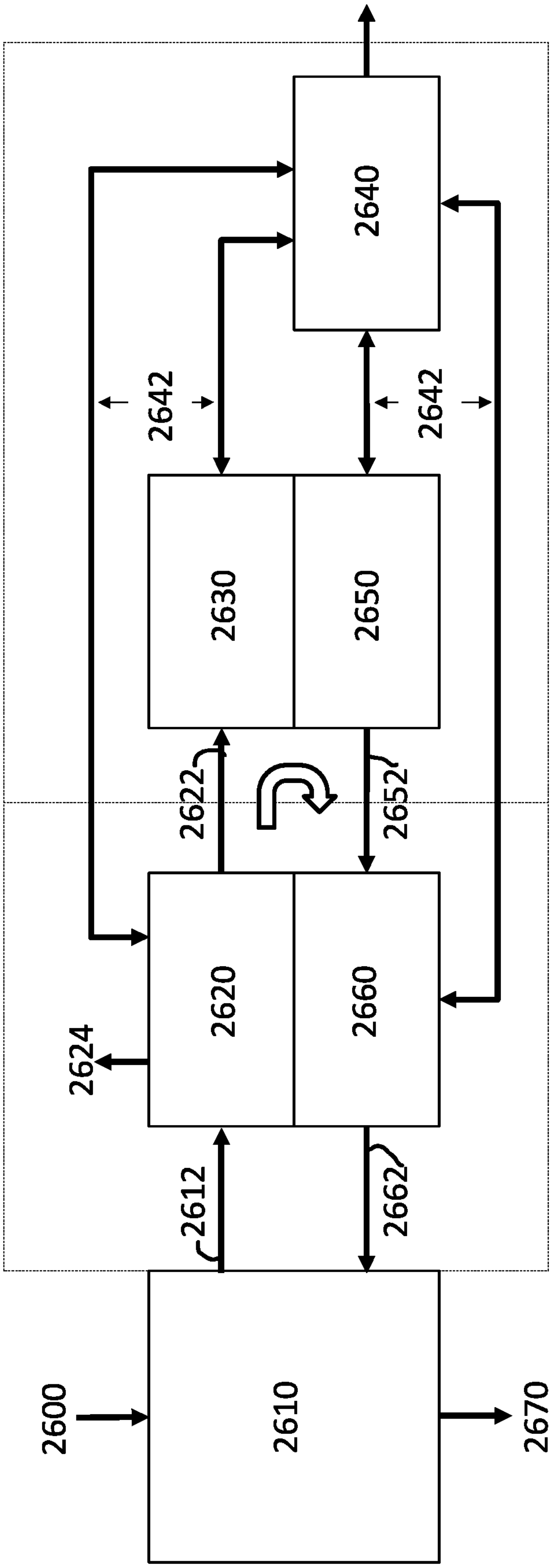
2600
2610
2670
2612
2662
2620
2660
2624
2622
2652
2630
2650
2642
2642
2640

Fig. 26

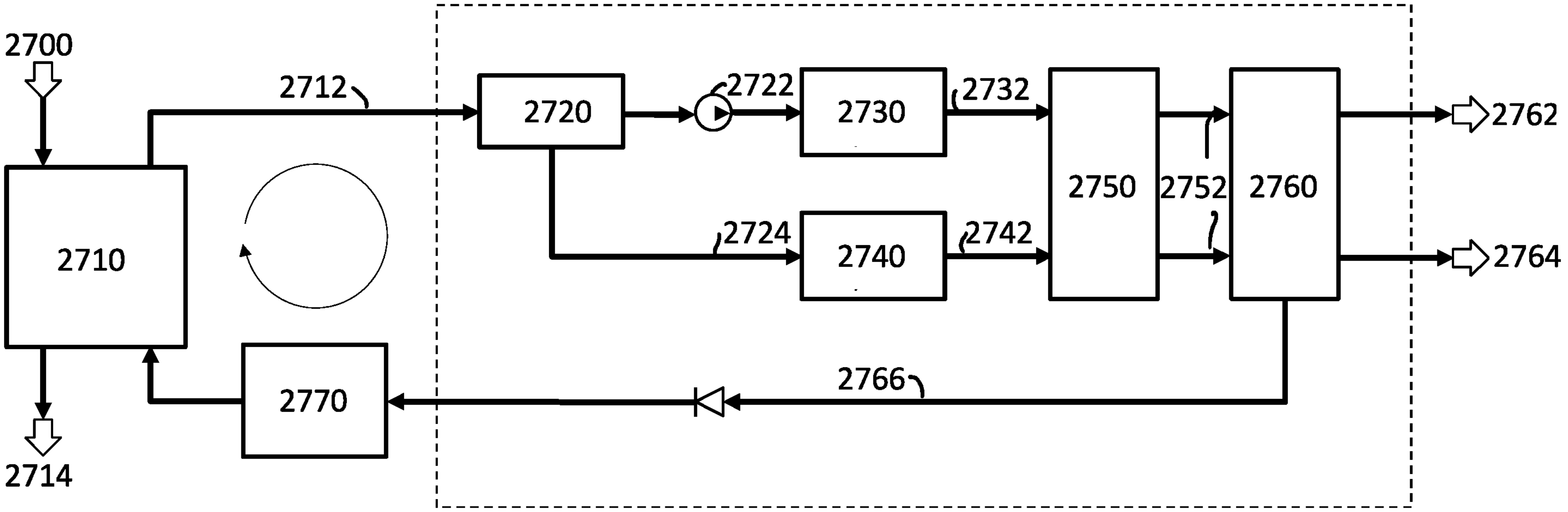
2700
2712
2720
2722
2730
2732
2750
2752
2760
2762
2710
2724
2740
2742
2764
2766
2770
2714

Fig. 27

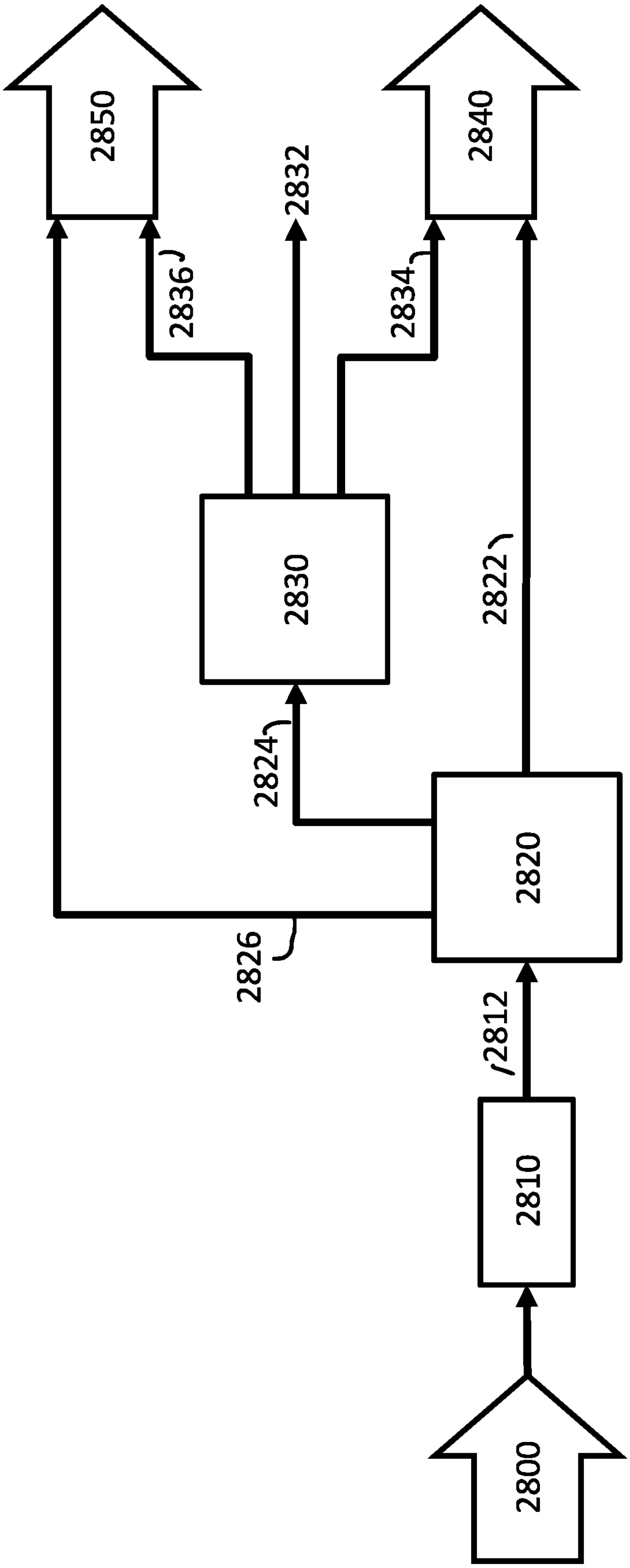
2800
2810
2812
2820
2826
2824
2830
2822
2832
2834
2836
2840
2850

Fig. 28