This disclosure describes systems and methods for treating nuclear fuel with supercritical fluids, such as supercritical carbon dioxide. The addition of various ligands to the supercritical fluids is disclosed, where one or more ligands can be chosen to selectively remove one or more fission products from the nuclear fuel. The nuclear fuel may be treated either within the nuclear reactor or may be removed from the reactor before treatment. This disclosure also presents methods and systems for liquid nuclear fuel treatment with supercritical fluids in, for example, a molten salt fast reactor, a traveling wave reactor, and a containerized molten salt reactor.
Reactor 502

Extractant(s) storage 512

Supercritical fluid control 510

Supercritical fluid container 508

Fluid storage 506

Contact vessel 514

Transfer unit 518

Separation unit 516

Reprocessing /waste 520

Nuclear material

Nuclear fuel

CO2

Chelate &/or waste

FIG. 5
Generate supercritical fluid

Add extractant into supercritical fluid

Contact a volume of supercritical fluid with a fuel salt

Separate supercritical fluid from fuel salt

Direct contacted fuel salt to reactor core

Separate fission products from supercritical fluid
Supercritical B fluid control
Extractant(s) storage
Supercritical fluid container
Fluid storage
CO2
Separation unit
Chelate &/or waste
Reprocessing /waste

Assembly
Absorber

FIG. 9
Charge reactor core

Maintain chain reaction

Contact a volume of reacted fuel with a supercritical fluid

Initiate chain reaction in regenerated fuel

Separate fission products from supercritical fluid

FIG. 10
NUCLEAR MATERIALS PROCESSING

RELATED APPLICATIONS


INTRODUCTION

[0003] Several alternative designs for nuclear reactors have become the subject of investigation. Two of these are molten salt reactors and traveling wave reactors.

[0004] In a molten salt reactor, a radioactive fuel such as uranium or thorium is dissolved into fluoride or chloride salts to form a solution referred to as a “fuel salt.” The fuel salt under normal conditions is an immobile solid material, but when heated above approximately 500°C, it becomes a liquid. In a molten salt reactor, the liquid fuel salt acts as both the heat source and a heat transfer fluid that assists in removing heat from the reactor. Tubes of fuel salt are deployed in a reactor core and, if the concentration of the fissile material is high enough, a sustained fission reaction may be created causing the fuel salt’s temperature to increase. In one design, the heated fuel salt may then be pumped through a heat exchanger to transfer the heat to a different heat transfer fluid (e.g., water or another molten salt). In an alternative design, the second heat transfer fluid may be flowed around stationary tubes of heated fuel salt. In either design, the second heat transfer fluid is then used, directly or indirectly, to generate power for beneficial use. Molten salt reactors are considered safer than some other designs because, in the event of an accident, the fuel salt will return to a solid, safe state. The plant can operate near atmospheric pressure with a coolant that returns to a solid form at ambient temperatures. This feature simplifies the plant and enables safety systems that do not require external electric power to safely shutdown, thereby assuring greater safety for the public.

[0005] In a traveling wave reactor (TWR), sometimes also known as a nuclear fission deflagration wave reactor or nuclear-burning-wave reactor, the main reactor components are a reactor vessel filled with a liquid sodium coolant and a reactor core. The reactor core is submerged in the sodium pool in the reactor vessel. In the center of the core are a few rods of enriched uranium (U-235), surrounded by rods of depleted uranium (U-238). The U-235 serves as an igniter, kick starting the traveling wave reaction—a slow-moving chain reaction of parallel waves of fission traveling through the uranium rods. These parallel waves initiate in the center of the core, slowly consuming the fuel and generating heat in the core. The sodium coolant is used to remove the heat from the core. A containment vessel surrounds the reactor vessel to prevent loss of sodium coolant in case of an unlikely leak from the reactor vessel. The pumps circulate primary sodium coolant between the reactor core and intermediate heat exchangers located in the pool. These heat exchangers have non-radioactive intermediate sodium coolant on the other side of the heat exchanger. Heated intermediate sodium coolant is circulated to steam generators that generate steam to drive turbines of electrical generators.

[0006] In theory, TWRs require no fuel reprocessing, use depleted or natural uranium as their primary fuel, require only a small amount of enriched uranium at startup, and never need refueling. This core longevity depends on the size of the initial charge of the uranium and on the fuel burn-up achieved during reactor operation.

Nuclear Materials Processing

[0007] This disclosure describes systems and methods for treating nuclear fuel with supercritical fluids, such as supercritical carbon dioxide. The addition of various ligands to the supercritical fluids is disclosed, where one or more ligands can be chosen to selectively remove one or more fission products from the nuclear fuel. The nuclear fuel may be treated either within the nuclear reactor or may be removed from the reactor before treatment. This disclosure also presents methods and systems for liquid nuclear fuel treatment with supercritical fluids in, for example, a molten salt fast reactor, a traveling wave reactor, and a containerized molten salt reactor.

[0008] An aspect of the present disclosure is a nuclear fission reactor that includes a reactor core containing a quantity of fuel salt including at least some fissionable material. The reactor is adapted to create a chain reaction in the fuel salt, thereby generating heat and fission products in the fuel salt. At least one heat exchanger is provided to transfer heat from the fuel salt to a coolant and a power generation unit is also provided that converts heat in the coolant into power. The reactor is further provided with a supercritical fluid separation system that is adapted to remove at least some amount of fission products from the fuel salt. In an ex situ treatment embodiment, the supercritical fluid separation system includes: a supercritical fluid contact vessel which contacts fuel salt with a supercritical fluid; a fuel salt transfer unit that circulates fuel salt between the reactor core and the supercritical fluid contact vessel; a supercritical fluid source fluidly connected to the supercritical fluid contact vessel; and a controller controlling the transfer of fuel salt between the reactor core and supercritical fluid contact vessel and controlling the transfer of supercritical fluid from the supercritical fluid source through the supercritical fluid contact vessel. The supercritical fluid may include at least one ligand that dissolves at least one fission product into the supercritical fluid when the ligand contacts the at least one fission product in the fuel salt. The supercritical fluid separation system may further include a separation vessel fluidly connected to the contact vessel to receive the supercritical fluid and fission product mixture flowing out of the contact vessel and separate at least some of the amount of fission products from the supercritical fluid. In an embodiment, the supercritical fluid is supercritical carbon dioxide containing a ligand that dissolves at least one fission product.

[0009] The fuel salt transfer unit may or may not maintain the fuel salt in a molten state when transferring the fuel salt from the reactor core to the contact vessel or from the contact vessel to the reactor core depending on the embodiment. Likewise, the fuel salt may or may not remain in a molten state while in the contact vessel during contact with the supercritical fluid. When the fuel salt transfer unit maintains the fuel salt in a molten state when transferring the fuel salt from the reactor core to the contact vessel, the contact vessel may include a molten fuel salt injector and a contact vessel envi-
Another aspect of this disclosure is a nuclear fission reactor having a supercritical fluid extraction system adapted to pass a supercritical fluid through a fuel salt container. The nuclear fission reactor includes: one or more fuel salt containers, including a first fuel salt container containing a quantity of fuel salt including at least some fissionable material; a reactor core adapted to hold at least the first fuel salt container and adapted to create a chain reaction in the fuel salt, thereby generating heat in the fuel salt; at least one heat exchanger adapted to transfer heat from the fuel salt to a coolant; and the supercritical fluid extraction system. The supercritical fluid extraction system further includes: a supercritical fluid source fluidly connectable to the first fuel salt container, a separation vessel fluidly connectable to the first fuel salt container that receives the supercritical fluid from the first fuel salt container, and a controller controlling the transfer of supercritical fluid from the source through the first fuel salt container into the separation vessel. The nuclear fission reactor may further include a container transfer unit that transfers the fuel salt container between the reactor core and the separation system. The separation system may further be adapted to pass the supercritical fluid through the fuel salt container while the fuel salt container is in the reactor core.

In an embodiment, the supercritical fluid is supercritical carbon dioxide containing a ligand that dissolves at least one fission product. The ligand may be selected from cupferron, chloroanillic acid, β-diketone, N-benzoyl-N-phe-nylhydroxylamine, α-dioxime dianinobenzidine, a porphyrine compound such as porphine, 8-hydroxyquinoline, nitroneonaphths, nitroneonaphthols, ethylenediaminetetraacetic acid, diphenylcarbazide, diphenylcarboxzone, Azoazoxy BN, sodium diethyldithiocarbarnate, dithizone, bisnaphthol 11, thiophenyltrifluoroacetone, thioine, thiophosphoric acid, phosphine sulfide, phosphoroxy acid, and tributylphosphate.

Yet another aspect of this disclosure is a traveling wave reactor that includes a supercritical fluid separation system. The traveling wave reactor includes: a reactor core containing a first fuel assembly having at least one fuel pin containing a fuel material including at least some fissionable material and fission products; a reactor vessel containing a primary sodium coolant, wherein the reactor core is within the reactor vessel and in contact with the primary sodium coolant; an assembly shuffling system adapted to move the first fuel assembly from a first location within the reactor core to a second location within the reactor core; and a supercritical fluid separation system that removes fission products from the fuel material in the first fuel assembly. The supercritical fluid separation system may further include: a supercritical fluid source fluidly connectable to the first fuel assembly that delivers the supercritical fluid into the first fuel assembly; a separation vessel fluidly connectable to the first fuel assembly that receives the supercritical fluid from the first fuel assembly; and a controller controlling the transfer of supercritical fluid from the source through the first fuel assembly into the separation vessel. The supercritical fluid source may be fluidly connectable to at least one pin in the first fuel assembly and may deliver the supercritical fluid into at least one pin. The supercritical fluid separation system may further include a separation vessel fluidly connectable to at least one pin in the first fuel assembly that receives the supercritical fluid from the at least one pin in the first fuel assembly and a controller that controls the transfer of supercritical fluid from the source through the at least one pin of the first fuel assembly into the separation vessel. The assembly shuffling system may be adapted to remove the first fuel assembly from the reactor core and the supercritical fluid separation system may remove fission products from the first assembly when the first assembly is outside of the reactor core. In an alternative embodiment, the assembly shuffling system may be further adapted to move the first assembly from the first location to an intermediate location within the reactor core before moving the first assembly to the second location and the supercritical fluid separation system may remove fission products from the first assembly when the first assembly is in the intermediate location within the reactor core. In yet another embodiment, the supercritical fluid separation system may remove fission products from the first assembly when the first assembly is in the first location within the reactor core.

The traveling wave reactor may include a fission product handling system that receives fission products from the supercritical fluid separation system. The traveling wave reactor may also include a swelling monitoring device that monitors expansion of the fuel material during operation of the traveling wave reactor and the controller may control the transfer of supercritical fluid based on the expansion of the fuel material. The traveling wave reactor may further include a coolant monitoring device that monitors a concentration of fission products in the coolant and the controller may control the transfer of supercritical fluid based on the concentration of fission products in the coolant. The supercritical fluid separation system may be further adapted to remove fission products from the primary sodium coolant. The traveling wave reactor may further include a transfer system adapted to hold the first fuel assembly in argon and the supercritical fluid separation system may be further adapted to remove fission products from argon that has been used to cool the fuel assembly. The traveling wave reactor may further include a coolant cleaning system including an absorber that removes fission products from the primary sodium coolant and the supercritical fluid separation system may be adapted to remove fission products from the absorber.
In an embodiment, the supercritical fluid is supercritical carbon dioxide containing a ligand that dissolves at least one fissionable product. The ligand may be selected from cupferron, chloroanillic acid, β-diketone, N-benzoyl-N-phenylhydroxylamine, α-dioximes dianisobenzodiazine, a porphyrine compound such as porphine, 8-hydroxyquinoline, nitrosanaphthols, nitrosoanils, ethylenediaminetetraacetic acid, diphylletharbazide, diphenylcarbaborane, Azaaazoxy BN, sodium diethylidithiocarbamate, dithizone, bismuthol III, thioenolylalkoxacetone, thiocyan, thiophosphoric acid, phosphine sulfide, phosphorothioic acid, and tributylphosphate.

Yet another aspect of this disclosure is a method of operating a nuclear fission reactor. In this aspect, the method includes: charging a reactor core with an initial fuel containing fissionable material; maintaining a first chain reaction in the reactor core for a period of time, thereby generating a partially-reacted fuel containing less fissionable material than in the initial fuel and at least some amount of fission products greater than in the initial fuel; contacting at least some of the partially-reacted fuel with a supercritical fluid containing at least one ligand that forms a metal complex with at least one fission product, thereby creating a supercritical fluid and fission product mixture and a regenerated fuel containing a lower amount of fission products than in the partially-reacted fuel; separating at least some of the supercritical fluid and fission product mixture from the regenerated fuel; initiating a second chain reaction in the regenerated fuel; and extracting at least some of the fission product from the supercritical fluid and fission product mixture. In the method the contacting may be performed on the at least some of the partially-reacted fuel without removing it from the reactor core. In the method the contacting operation may be performed without interrupting the first chain reaction in the reactor core. The method may further include: removing the at least some of the partially-reacted fuel from the reactor core without interrupting the first chain reaction; performing the contacting operation after removing the at least some of the partially-reacted molten salt fuel from the reactor core; and returning the regenerated molten salt fuel to the reactor core. The method may further include returning the regenerated molten salt fuel to the reactor without interrupting the first chain reaction.

These and various other features as well as advantages which characterize the systems and methods described herein will be apparent from a reading of the following detailed description and a review of the associated drawings. Additional features are set forth in the description which follows, and in part will be apparent from the description, or may be learned by practice of the technology. The benefits and features of the technology will be realized and attained by the structure particularly pointed out in the written description and claims hereof as well as the appended drawings.

It is to be understood that both the foregoing general description and the following detailed description are explanatory and are intended to provide further explanation of the invention as claimed.

BRIEF DESCRIPTION OF THE DRAWINGS

The following drawing figures, which form a part of this application, are illustrative of described technology and are not meant to limit the scope of the invention as claimed in any manner, which scope shall be based on the claims appended hereto.

FIG. 1 illustrates a simplified schematic view of a molten salt fast spectrum nuclear reactor, in accordance with one or more embodiments of the present disclosure.

FIGS. 2A and 2B illustrate a simplified schematic view of a molten salt fast spectrum nuclear reactor with a protective layer disposed on one or more internal surfaces of the reactor, in accordance with one or more embodiments of the present disclosure.

FIG. 3 illustrates an embodiment of a nuclear power plant for generating power from a nuclear reaction using a molten chloride fast reactor.

FIG. 4 illustrates another embodiment of a simplified schematic view of a molten salt nuclear reactor.

FIG. 5 is a block flow diagram of an embodiment of an example supercritical fluid treatment system.

FIG. 6 is an embodiment of a method for treating a fuel salt reactor with a supercritical fluid treatment system.

FIG. 7 is a block flow diagram of the example fuel salt reactor used with the example supercritical fluid treatment components.

FIG. 8 is a block flow diagram of the example containerized fuel salt reactor used with the example supercritical fluid treatment components.

FIG. 9 is a block flow diagram of the example traveling wave reactor used with the example supercritical fluid treatment components of FIG. 5.

FIG. 10 is an embodiment of a method for operating a reactor with supercritical fluid separation.

DETAILED DESCRIPTION

This disclosure describes systems and methods for treating nuclear fuel with supercritical fluids, such as supercritical carbon dioxide. The addition of various ligands to the supercritical fluids is disclosed, where one or more ligands can be chosen to selectively remove one or more fission...
products from the nuclear fuel. The nuclear fuel may be treated either within the nuclear reactor or may be removed from the reactor before treatment. This disclosure also presents methods and systems for liquid nuclear fuel treatment with supercritical fluids in, for example, a molten salt fast reactor, a traveling wave reactor, and a containerized molten salt reactor.

[0032] As used herein, fissionable material includes any nuclide capable of undergoing fission when exposed to low-energy thermal neutrons or high-energy neutrons. Furthermore, for the purposes of this disclosure, fissionable material includes any fissile material, any fertile material or combination of fissile and fertile materials. As used herein, a direct fission product is the atom that remains after fission of a fissile atom. As used herein, an indirect fission product is a decay daughter, grand-daughter, etc., that results from the radioactive decay of a direct fission product. However, at any given point in time, some quantity of a particular species fission product compound, such as $^{99m}$Mo, will be a direct product and the remaining quantity will be indirect products, as there can be multiple decay chains at work. As used herein, a fuel salt can include target material (material that can undergo fission) as well as ancillary material (including post-irradiation fission products and non-fissile salts in the fuel).

[0033] The disclosed treatment systems and methods can be used with molten salt reactor designs and related systems, where the molten salt includes molten fluoride fuel salt, molten chloride fuel salt, fuel salts of UCLF$_2$, variety, as well as bromide fuel salts. Binary, ternary and quaternary chloride fuel salts of uranium, as well as other fissionable elements, are contemplated. This disclosure also presents methods and systems for manufacturing such fuel salts, for creating salts that reduce corrosion of the reactor components and for creating fuel salts that are not suitable for weapon applications.

[0034] Metalloids are elements with both metallic and non-metallic properties, and include arsenic, selenium and tellurium. A metal is an element that forms positive ions in solution, and produces oxides that form hydroxides, rather than acids, with water. Metals include alkali metals, alkali-earth metals, transition metals, noble metals (including the precious metals gold, platinum and silver), rare metals, rare-earth metals (lanthanides), actinides (including the transuranic metals), light metals, heavy metals, synthetic metals and radioactive metals. Specific examples are given herein of extraction methods for extracting lanthanides and actinides (collectively referred to as the f-group elements from the filling of their 4f and 5f orbitals). The f-group elements are commonly produced by nuclear fission reactions, and the actinides are radioactive. Transition metals are commonly used or produced in many industrial processes and products, such as mineral production or fly ash.

[0035] Suitable fluids and/or supercritical fluids for use in the disclosed embodiments include carbon dioxide, nitrogen, nitrous oxide, methane, ethylene, propylene and propylene. (See Table I) Carbon dioxide is a particularly suitable fluid for both subcritical and supercritical fluid extractions because of its moderate chemical constants ($T_c$=31°C, $P_c$=73 atm) and its inertness (i.e. it is non-explosive and thoroughly safe for extractions, even extractions performed at supercritical conditions). Carbon dioxide also is abundantly available and relatively inexpensive. Virtually any condition above the critical temperature and pressure for carbon dioxide is acceptable for producing a supercritical carbon dioxide fluid solvent useful for practicing the extraction process as described herein.

[0036] The fluids listed in Table I may be used either individually or in combination, as mixed fluids or supercritical fluid solvents.

<table>
<thead>
<tr>
<th>TABLE I</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>PHYSICAL PARAMETERS OF SELECTED SUPERCRITICAL FLUIDS</strong></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Fluid</th>
<th>Molecular Formula</th>
<th>$T_c$ (°C)</th>
<th>$P_c$ (atm)</th>
<th>$\rho_0$ (g/ml)</th>
<th>$\rho_{400}$ atm**</th>
</tr>
</thead>
<tbody>
<tr>
<td>Carbon dioxide</td>
<td>CO$_2$</td>
<td>31.1</td>
<td>72.9</td>
<td>0.47</td>
<td>0.06</td>
</tr>
<tr>
<td>Nitrous oxide</td>
<td>N$_2$O</td>
<td>36.5</td>
<td>71.7</td>
<td>0.45</td>
<td>0.04</td>
</tr>
<tr>
<td>Ammonia</td>
<td>NH$_3$</td>
<td>132.5</td>
<td>112.5</td>
<td>0.24</td>
<td>0.40</td>
</tr>
<tr>
<td>n-Pentane</td>
<td>C$<em>5$H$</em>{12}$</td>
<td>196.6</td>
<td>33.3</td>
<td>0.23</td>
<td>0.51</td>
</tr>
<tr>
<td>n-Butane</td>
<td>C$<em>4$H$</em>{10}$</td>
<td>91.2</td>
<td>37.5</td>
<td>0.23</td>
<td>0.50</td>
</tr>
<tr>
<td>n-Propane</td>
<td>C$_3$H$_6$</td>
<td>96.8</td>
<td>42.0</td>
<td>0.22</td>
<td>0.61</td>
</tr>
<tr>
<td>Sulfur hexafluoride</td>
<td>SF$_6$</td>
<td>45.5</td>
<td>37.1</td>
<td>0.74</td>
<td>1.61</td>
</tr>
<tr>
<td>Xenon</td>
<td>Xe</td>
<td>16.6</td>
<td>58.4</td>
<td>1.10</td>
<td>2.30</td>
</tr>
<tr>
<td>Dichlorodifluoromethane</td>
<td>CCl$_2$F$_2$</td>
<td>111.8</td>
<td>40.7</td>
<td>0.56</td>
<td>1.12</td>
</tr>
<tr>
<td>Trifluoromethane</td>
<td>CHF$_3$</td>
<td>25.9</td>
<td>46.9</td>
<td>0.52</td>
<td>—</td>
</tr>
<tr>
<td>Methanol</td>
<td>CH$_3$OH</td>
<td>240.5</td>
<td>78.9</td>
<td>0.27</td>
<td>—</td>
</tr>
<tr>
<td>Ethanol</td>
<td>C$_2$H$_5$OH</td>
<td>243.4</td>
<td>63.0</td>
<td>0.28</td>
<td>—</td>
</tr>
<tr>
<td>Isopropanol</td>
<td>C$_3$H$_7$OH</td>
<td>235.3</td>
<td>47.0</td>
<td>0.27</td>
<td>—</td>
</tr>
<tr>
<td>Diethyl ether</td>
<td>(C$_2$H$_5$)$_2$O</td>
<td>193.6</td>
<td>36.3</td>
<td>0.27</td>
<td>—</td>
</tr>
<tr>
<td>Water</td>
<td>H$_2$O</td>
<td>374.1</td>
<td>218.3</td>
<td>—</td>
<td>—</td>
</tr>
</tbody>
</table>


**$T_c$ = 1.03

[0037] In addition, a modifying solvent (also referred to as an extractant) may be added to the fluid, including supercritical fluids, to improve the solvent characteristics thereof. Currently, useful modifying solvents include water, organic solvents, such as low to medium boiling point alcohols and esters, particularly the lower alkyl alcohols and esters, such as methanol, ethanol, ethyl acetate and the like; and phosphate esters, particularly lower alkyl phosphate esters, such as tributyl phosphate. With more specificity, but without limitation, the modifiers are usually added to the fluids at proportions of between about 0.1% and 20.0% by weight. The extractants contemplated for use herein may not be supercritical fluids at the disclosed operating conditions. Such extractants may be simply dissolved in the fluid solvents, including the supercritical fluid solvents, to improve the solvent properties.

[0038] In one embodiment, the chosen extractant is combined with a supercritical fluid at the described proportions prior to feeding the supercritical fluid to the extraction vessel. Alternatively, the supercritical fluid is fed to the extraction vessel without the extractant. The extractant is then introduced into the extraction vessel and thereby combined with the supercritical fluid.

[0039] Extractants also include chelating agents. Chelating agents useful for solubilizing metals and metalloids in supercritical fluids are listed in Table II. The list of chelating agents is not exhaustive and for illustration only. Many other chelating agents, now known or hereafter discovered that are useful for forming metal and metalloid chelates, also may be used.
TABLE II

COMMONLY USED METAL CHELATING AGENTS

<table>
<thead>
<tr>
<th>Oxygen Donating Chelating Agents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cupferron</td>
</tr>
<tr>
<td>Chloroauric acid and related reagents</td>
</tr>
<tr>
<td>β-diketones and related reagents</td>
</tr>
<tr>
<td>N-Benzoyl-N-phenylthiohydroxylamine and related reagents</td>
</tr>
<tr>
<td>Macrocyclic compounds</td>
</tr>
<tr>
<td>Nitrogen Donating Chelating Agents</td>
</tr>
<tr>
<td>ε-dioximes</td>
</tr>
<tr>
<td>Diaminobenzidine and related reagents</td>
</tr>
<tr>
<td>Porphyrines and related reagents</td>
</tr>
<tr>
<td>Oxygen and Nitrogen Donating Chelating Agents</td>
</tr>
<tr>
<td>8-Hydroxy quinoline</td>
</tr>
<tr>
<td>Nitrosoanilides and nitrosophenols</td>
</tr>
<tr>
<td>EDTA and other complexonates</td>
</tr>
<tr>
<td>Diphenylcarbazide and diphenylcarbazone</td>
</tr>
<tr>
<td>Azosoxyl BN</td>
</tr>
<tr>
<td>Sulfur or Phosphorus Donating Chelating Agents</td>
</tr>
<tr>
<td>Sodium diethylidithiocarbamate and related reagents</td>
</tr>
<tr>
<td>Dilauroxide and related reagents</td>
</tr>
<tr>
<td>Burmuthiti II</td>
</tr>
<tr>
<td>Thiohexyltrifluoroacetone</td>
</tr>
<tr>
<td>Thiexine</td>
</tr>
<tr>
<td>Thiophosphonic acids</td>
</tr>
<tr>
<td>Phosphine Sulfides</td>
</tr>
<tr>
<td>Phosphorothioic acids</td>
</tr>
<tr>
<td>Tributylphosphate and related reagents</td>
</tr>
</tbody>
</table>

[0040] Prior to discussing treating fuel salt with supercritical fluid in greater detail, a brief discussion of the general components of various nuclear reactors will be discussed. FIGS. 1-7 generally describe systems and methods of operating a molten salt nuclear reactor, a traveling wave reactor, and a containerized molten salt reactor. For instance, FIGS. 1-3 depict various embodiments of a molten salt nuclear reactor 100 for operating in a fast spectrum breed-and-burn mode. These are just examples to provide context for discussion of treating fuel salt with supercritical fluid and the reader should understand that potentially any molten fuel nuclear reactor could be adapted to use the fuel embodiments described below. While various fluoride salts may be utilized in molten salt nuclear reactors as described below, fluoride-based fuel salts typically display heavy metal concentrations significantly below that which may be achieved with chloride-based and fluoride-fluoride-based fuel salts described in the present disclosure.

[0041] FIG. 1 illustrates a simplified schematic view of a molten salt fast spectrum nuclear reactor 100, in accordance with one or more embodiments of the present disclosure. In one embodiment, the reactor 100 includes a reactor core section 102. The reactor core section 102 (which may also be referred to as the “reactor vessel”) includes a fuel input 104 and a fuel output 106. The fuel input 104 and the fuel output 106 are arranged such that during operation a flow of the molten fuel salt 108 is established through the reactor core section 102. For example, the fuel input 104 and/or the fuel output 106 may consist of conical sections acting as converging and diverging nozzles respectively. In this regard, the molten fuel 108 is fluidically transported through the volume of the reactor core section 102 from the input 104 to the output 106 of the reactor core section 102. Although FIG. 1 shows fluid fuel flow with arrows, it is to be appreciated that the direction of flow may be modified as appropriate for different reactor and plant configurations. Specifically, FIG. 1 shows fluid fuel 108 flow from the ‘bottom’ to the ‘top’ in the central core region, and alternative apparatus or maintain a fluid fuel 108 flow from the top to the central region.

[0042] The reactor core section 108 may take on any shape suitable for establishing criticality within the molten fuel salt 108 within the reactor core section 102. By way of non-limiting example, the reactor 100 may include, but is not limited to, an elongated core section, as depicted in FIG. 1. In addition, the reactor core section 108 may take on any cross-sectional shape. By way of non-limiting example, the reactor core section 108 may have, but is not required to have, a circular cross-section, an ellipsoidal cross-section or a polygonal cross-section.

[0043] The dimensions of the reactor core section 102 are selected such that criticality is achieved within the molten fuel salt 108 when flowing through the reactor core section 102. Criticality refers to a state of operation in which the nuclear fuel sustains a fission chain reaction, i.e., the rate of production of neutrons in the fuel is at least equal to the rate at which neutrons are consumed (or lost). For example, in the case of an elongated core section, the length and cross-sectional area of the elongated core section may be selected in order to establish criticality within the reactor core section 102. It is noted that the specific dimensions necessary to establish criticality are at least a function of the type of fissile material, fertile material and/or carrier salt contained within the reactor 100. Principles of molten fuel nuclear reactors are described in U.S. patent application Ser. No. 12/118,118 to Leblanc, filed on May 9, 2008, which is incorporated herein in its entirety.

[0044] The reactor core section 102 is formed from any material suitable for use in molten salt nuclear reactors. For example, the bulk portion of the reactor core section 102 may be formed, but is not required to be formed, from one or more molybdenum alloy, one or more zirconium alloys (e.g., ZIRCALOY™), one or more niobium alloys, one or more nickel alloys (e.g., HASTELLOY™ N) or high temperature ferritic, martensitic, or stainless steel and the like. It is further noted that the internal surface may be coated, plated or lined with one or more additional materials in order to provide resistance to corrosion and/or radiation damage, as discussed in additional detail further herein.

[0045] In the embodiment shown, the reactor 100 includes a primary coolant system 110 that takes heat from the reactor core 102 and transfers that heat to the secondary coolant 126 in the secondary coolant system 120 via the heat exchanger 119. In the embodiment illustrated in FIG. 1, the molten fuel salt 108 is used as the primary coolant 118. Cooling is achieved by flowing molten fuel salt 108 heated by the ongoing chain reaction from the reactor core 102, and flowing cooler molten fuel salt 108 into the reactor core 102, at the rate necessary to maintain the temperature of the reactor core 102 within its operational range. In this embodiment, the primary coolant system 110 is adapted and sized to maintain the molten fuel salt 108 in a subcritical condition when outside of the reactor core 102.

[0046] The primary coolant system 110 may include one or more primary coolant loops 112 formed from piping 114. The primary coolant system 110 may include any primary coolant system arrangement known in the art suitable for implementation in a molten fuel salt context. The primary coolant system 110 may circulate fuel 108 through one or more pipes.
114 and/or fluid transfer assemblies of the one or more primary coolant loops 112 in order to transfer heat generated by the reactor core section 102 to downstream thermally driven electrical generation devices and systems. For purposes of simplicity, a single primary coolant loop 112 is depicted in FIG. 1. It is recognized herein, however, that the primary coolant system 110 may include multiple parallel primary coolant loops (e.g., 2-5 parallel loops), each carrying a selected portion of the molten fuel salt inventory through the primary coolant circuit.

In an alternative embodiment (an example of which is shown in FIG. 2A), the primary coolant system 110 may be configured such that a primary coolant 118 (different than the molten fuel salt 108) enters the reactor core section (e.g., main vessel). In this embodiment, the fuel salt 108 does not leave the reactor core section, or main vessel, but rather the primary coolant 118 is flowed into the reactor core 102 to maintain the temperature of the core within the desired range. It is noted that in this embodiment the reactor 100 may include an additional heat exchanger (not shown) in the reactor core section 102, or main vessel. In this embodiment, the secondary coolant system 120 may be optional; the usable power can be derived directly from the primary coolant system 110. In this embodiment, the primary coolant may be a chloride salt with a suitable melting point. For example, the salt may be a mixture of sodium chloride and magnesium chloride.

In the embodiment shown in FIG. 1, the primary coolant system 110 includes one or more pumps 116. For example, one or more pumps 116 may be fluidly coupled to the primary coolant system 110 such that the one or more pumps 116 drive the primary coolant 118, in this case the molten fuel 108, through the primary coolant/reactor core section circuit. The one or more pumps 116 may include any coolant/fuel pump known in the art. For example, the one or more fluid pumps 116 may include, but are not limited to, one or more mechanical pumps fluidly coupled to the primary coolant loop 112. By way of another example, the one or more fluid pumps 116 may include, but are not limited to, one or more electromagnetic (EM) pumps fluidly coupled to the primary coolant loop 112.

FIG. 1 further illustrates that the reactor 100 includes a secondary coolant system 120 thermally coupled to the primary coolant system 110 via one or more heat exchangers 119. The secondary coolant system 120 may include one or more secondary coolant loops 122 formed from piping 124. The secondary coolant system 120 may include any secondary coolant system arrangement known in the art suitable for implementation in a molten fuel salt context. The secondary coolant system 120 may circulate a secondary coolant 126 through one or more pipes 124 and/or fluid transfer assemblies of the one or more secondary coolant loops 122 in order to transfer heat generated by the reactor core section 102 and received via the primary heat exchanger 119 to downstream thermally driven electrical generation devices and systems. For purposes of simplicity, a single secondary coolant loop 124 is depicted in FIG. 1. It is recognized herein, however, that the secondary coolant system 120 may include multiple parallel secondary coolant loops (e.g., 2-5 parallel loops), each carrying a selected portion of the secondary coolant through the secondary coolant circuit. It is noted that the secondary coolant may include any second coolant known in the art. By way of example, the secondary coolant may include, but is not limited to, liquid sodium.

It is further noted that, while not depicted in FIG. 1, the reactor 100 may include any number of additional or intermediate heating/cooling systems and/or heat transfer circuits. Such additional heating/cooling systems may be provided for various purposes in addition to maintaining the reactor core 102 within its operational temperature range. For example, a tertiary heating system may be provided for the reactor core 102 and primary coolant system 110 to allow a cold reactor containing solidified fuel salt to be heated to an operational temperature in which the salt is molten and flowable.

Other ancillary components 127 may also be utilized, as illustrated, in the primary coolant loop 112. Such ancillary components 127 may be include one or more filters or drop out boxes for removing particulates that precipitate from the primary coolant 118 during operation. To remove unwanted liquids from the primary coolant 118, the ancillary components 127 may include any suitable liquid-liquid extraction system such as one or more co-current or counter-current mixer/settler stages, an ion exchange technology, or a gas absorption system. For gas removal, the ancillary components 127 may include any suitable gas-liquid extraction technology such as a flash vaporization chamber, distillation system, or a gas stripper. Some additional embodiments of ancillary components 127 are discussed in greater detail below.

It is noted herein that the utilization of various metal salts, such as metal chloride salts, in reactor 100 may cause corrosion and/or radiation degradation over time. A variety of measures may be taken in order to mitigate the impact of corrosion and/or radiation degradation on the integrity of the various salt-facing components (e.g., reactor core section 102, primary coolant piping 114, heat exchanger 119 and the like) of the reactor 100 that come into direct or indirect contact with the fuel salt or its radiation.

In one embodiment, the velocity of fuel flow through one or more components of the reactor 100 is limited to a selected fuel salt velocity. For example, the one or more pumps 116 may drive the molten fuel 108 through the primary coolant loop 112 of the reactor 100 at a selected fuel salt velocity. It is noted that in some instances a flow velocity below a certain level may have a detrimental impact on reactor performance, including the breeding process and reactor control. By way of non-limiting example, the total fuel salt inventory in the primary loop 112 (and other portions of the primary coolant system 110) may exceed desirable levels in the case of lower velocity limits since the cross-sectional area of the corresponding piping of the primary loop 112 scales upward as flow velocity is reduced in order to maintain adequate volumetric flow through the primary loop 112. As such, very low velocity limits (e.g., 1 m/s) result in large out-of-core volumes of fuel salt and can negatively impact the breeding process of the reactor 100 and reactor control. In addition, a flow velocity above a certain level may detrimentally impact reactor performance and longevity due to erosion and/or corrosion of the internal surfaces of the primary loop 112 and/or reactor core section 102. As such, suitable operational fuel salt velocities may provide a balance between velocity limits required to minimize erosion/corrosion and velocity limits required to manage out-of-core fuel salt inventory. For example, in the case of a molten chloride fuel salt, the fuel salt velocity may be controlled from 2-20 m/s, such as, but not limited to, 7 m/s.
FIGS. 2A and 2B illustrate a simplified schematic view of a molten salt fast spectrum nuclear reactor 100 with a protective layer 128 disposed on one or more internal surfaces of the reactor 100, in accordance with one or more embodiments of the present disclosure.

In one embodiment, the protective layer 128 is disposed on one or more surfaces of the reactor 100 facing the fuel salt 108 of the reactor 100. The protective layer 128 may provide resistance to corrosion and/or radiation degradation of one or more reactor salt-facing surfaces of the reactor 100. For the purposes of the present disclosure, a material resistant to corrosion and/or radiation degradation is interpreted as any material displaying resistance to corrosion and/or radiation degradation superior to the underlying bare surface of the reactor 100.

The protective layer 128 may include any material known in the art suitable for providing an internal surface of a reactor with corrosion and/or radiation resistance to a corresponding fuel salt. Thus, the material of the protective layer 128 may vary depending on the salt 108 used. In one embodiment, the protective layer 128 includes one or more refractory metals. For example, the protective layer 128 may include, but is not limited to, one or more of niobium, molybdenum, tantalum, tungsten or rhenium. In another embodiment, the protective layer 128 includes one or more refractory alloys. For example, the protective layer 128 may include, but is not limited to, one or more of a molybdenum alloy (e.g., titanium-zirconium-molybdenum (TZM) alloy), a tungsten alloy, tantalum, niobium or rhenium. In another embodiment, the protective layer 128 includes one or more nickel alloys. In another embodiment, the protective layer 128 includes a carbide, such as silicon carbide.

In an embodiment, the protective layer 128 is formed by placing the internal surface of the one or more portions (e.g., piping 114 or primary loop 112) of the reactor 100 with the selected protective material. In another embodiment, the protective layer 128 includes one or more coatings of the selected protective material disposed on the internal surface of one or more portions of the reactor 100. In yet another embodiment, the bulk material of the various components may be formed with one or more of the protective materials described above. For instance, the piping 114 of the primary coolant loop 112 may include, but is not limited to, TZM piping. In one embodiment, as shown in FIG. 2A, the internal salt-facing surface of the reactor core section 102 includes a protective layer 128. For example, the vessel of the reactor core section 102 may be formed from steel or a zirconium alloy, with refractory alloy or nickel alloy platting disposed on the internal salt-facing surface of the reactor core section 102 to form the protective layer 128. For instance, the reactor core section 102 may include, but is not limited to, a molybdenum-based protective layer 128 having a thickness from approximately 5-7 mm, with the vessel of the reactor core section 102 having a wall thickness of approximately 9-11 cm thick.

Similarly, as shown in FIG. 2B, the salt-facing surface of the piping 114 of the primary coolant loop 112 (which may be the internal and/or external surface of piping or other components) includes a protective layer 128. For example, refractory alloy or nickel alloy platting may be disposed on the salt-facing surface of the piping 114 of the primary coolant loop 112 to form the protective layer 128.

FIG. 3 illustrates an embodiment of a nuclear power plant for generating power from a nuclear reaction using a molten fuel salt, in this case a molten chloride fast reactor (MCFR). For a power plant application, the heat generated by the MCFR will be converted into electrical power by power conversion hardware. In the embodiment shown, Rankine cycle power conversion hardware was used with water (steam) as the working fluid. The conversion efficiency of a Rankine cycle plant is in large part determined by the temperature (and pressure) of the steam entering the turbines, where higher temperatures correlate to higher efficiency. Performance is coupled to steam pressure as well as temperature and the highest efficiency Rankine cycle plants use supercritical and ultra-supercritical steam. Although a Rankine cycle steam turbine was used for illustration purposes, heat engines based on other cycles are also feasible such as closed-cycle gas turbines (e.g., air, helium, or CO₂) based on the Brayton cycle.

The power conversion system encompasses all systems that come into contact with the power conversion system working fluid. In the case of a steam Rankine cycle plant as illustrated, this includes a steam generator 152, a turbine system 154, water circulation loop 162 including one or more water circulation pumps 156 and a cooling tower 158, electrical generation equipment 160 and a control system 162. In addition, a fuel storage system 166 for storing new fuel salt and a reaction product storage system 168 to receive and safely contain used fuel salt are illustrated. As illustrated in FIG. 3, the power conversion system starts with a primary coolant transferring heat to the power cycle working fluid through a heat exchanger (e.g., steam generator 152).

The power conversion system receives thermal power from the reactor 100 and converts that heat into mechanical and then electrical power. The analysis specifically focused on using conventional steam Rankine cycle hardware for power conversion. The analyzed configuration has three turbines, with a high pressure turbine (HPT), intermediate pressure turbine (IPT), and low pressure turbine (LPT), illustrated simply as the turbine system 154.

The model in FIG. 3 is simplified in that it shows only the major components of the power plant. In the model used, the HPT receives steam from the “main steam” generation system that is heated by the primary cooling fluid carrying thermal energy from the reactor. Exhaust from HPT is sent to the reheating steam generation system, where the primary cooling fluid transfers heat to the exhaust from the HPT, and that heated steam is fed to the IPT. The exhaust from the IPT is fed to the LPT to extract additional enthalpy. There are often multiple turbines in parallel, particularly for the LPT. In the model used, there are twin LPTs that are used for the final expansion step. In the model, all turbines are on a common shaft and direct coupled to an electrical generator 160. The outlet of the LPT flows to a condenser that cools the steam to near ambient temperature. For this analysis, the LPT is assumed to be a once-through condenser that receives heat from a large body of water, like a large lake or river. After the condenser, the water is pumped and sent through several feedwater heaters. The feedwater heaters preheat the feedwater by mixing with steam extracted from various points on the turbines. The preheated fluid from the feedwater heaters is then fed to the steam generator, where it is heated to temperature for the main turbine.

FIG. 4 illustrates another embodiment of a simplified schematic view of a molten salt nuclear reactor 200. The reactor 200 is a pool-type reactor in which in some examples the fuel salt 108, may be flowing through the pool, such as
through piping. In other examples, the fuel salt 108 is circulating within the pool but is contained within and is never removed from the core.

[0065] In the example shown in FIG. 4, the fuel salt is contained in tubes 204 that are located at the center of a pool 210 of coolant 202 in a closed reactor vessel 206. One or more tubes 204 that contain nuclear fuel 108 may be mechanically treated as unit and referred to as a “fuel assembly”. The top portion of the reactor vessel 206 may be filled with some inert gas 218 such as argon. The fuel tubes 204 are arranged in an array similar to conventional solid fuel arrays in a light water reactor. The coolant 202 transfers heat from the center of the pool 210 to heat exchangers 208 located on the periphery of the pool 210. In the embodiment shown, the circulation of the coolant 202 (illustrated by the dashed arrows 212) within the pool 210, which may be natural or induced by an impeller or other mechanism (not shown), convects heat away from the fuel tubes 204 to be removed by the heat exchangers 208.

[0066] The heat exchangers 208 transfer heat from the pool 210 to a secondary coolant system 214. In an embodiment, the secondary coolant is water that is boiled in the heat exchangers and the resulting steam 216 is used to drive turbines (not shown) for the generation of power.

[0067] An optional set of reflector modules 232 may be provided around the array of fuel tubes either within the reactor vessel as shown in FIG. 4, and/or external to the reactor vessel, to increase the efficiency of the reactor. Optional shutdown rods may be provided to maintain the reactor subcritical when needed.

[0068] Following its initial start-up with enriched (~12% 235U) fuel, an MCFR may not require the ongoing feed of enriched fissile material. Instead, an MCFR can be fed depleted or natural uranium, among other fertile materials. During normal operations, modelling shows that the reactor slowly breeds up in reactivity. To counter this increase in reactivity, a small quantity of fully mixed fuel salt may be removed and replaced with fertile fuel salt. The addition of fertile material is, in effect, a control rod that reduces reactivity.

[0069] Rather than going to disposability, used MCFR fuel can be collected until a sufficient amount is available to start a new reactor. Such a daughter reactor contains a chemically identical fuel salt, and thus, is able to be started without any new enrichment. By transferring used fuel, in total, to a daughter plant for use as the initial fuel for that plant, creation of a fleet of MCFRs significantly reduces the use of actinides and defers the vast majority of radioactive waste until the end of the fleet buildup. For ultimate disposal of actinide-bearing fuel salt, prior work found that the salt itself could be packaged, without chemical conversion, into a suitable waste form.

[0070] In an embodiment, the reactor 200 in FIG. 4 is operated as a traveling wave reactor (TWR). Such a traveling wave reactor is disclosed in more detail in U.S. patent application Ser. No. 11/605,943 filed Nov. 28, 2006 in the names of Roderick A. Hyde, et al. and titled “Automated Nuclear Power Reactor For Long-Term Operation”, which application is assigned to the assignee of the present application, the entire disclosure of which is hereby incorporated by reference.

[0071] In operation, the fuel in the TWR may remain solid, such solid fuel often referred to as a fuel pin 204 because of its cylindrical shape. In this embodiment, pins 204 may be in the form of a solid cylinder of material, which may or may not be porous and may or may not be enclosed in a tube. In an alternative embodiment, the fuel is in the form of particulate material loosely packed and contained by the tubes 204, thus the fuel pin 204 in this embodiment includes the particulate fuel and tube 204.

[0072] In an embodiment of a TWR, fuel assemblies of one or more pins may be moved during operation in order to maintain the burnfront in a static location within the array of pins. For example, as the fuel in a particular assembly in a central location is bred up and then fissioned to the point that it is no longer contributing to the overall criticality of the reactor, that assembly may be moved to a location at the periphery of the tubes and replaced with a fresh assembly. In this manner, the traveling burnfront may be maintained at a static location within the reactor, with spent assemblies being replaced by fresh assemblies from within the reactor as needed to maintain criticality.

[0073] TWRs were generally described above in the introduction. In an embodiment, the TWR 200 has a cylindrical reactor core submerged in a large sodium pool 202 in the reactor vessel 206. In an embodiment, the reactor vessel and fuel assemblies are completely surrounded on the sides and below by a contiguous containment vessel (not shown) that does not have any penetrating components so that in the event that sodium coolant leaks from the reactor vessel, there can be no release of coolant to the environment unless both the reactor vessel and the containment vessel are breached.

[0074] In operation, pumps circulate the primary sodium coolant 202 within the pool so that it passes up, through the reactor core toward the surface of the coolant pool. In the embodiment shown, intermediate heat exchangers are submerged in the coolant pool above the reactor core. Through the intermediate heat exchangers, heat is transferred from the primary sodium coolant being circulated in the pool to the intermediate sodium cooling system. Heated intermediate sodium coolant is circulated to the steam generators (not shown) that generate steam to drive turbine and electrical generators. In an embodiment in which the primary sodium coolant may be exposed to fission products, this design prevents radioactive materials from being removed from the reactor vessel as part of the cooling and electricity generation process. Reactor containment is completed using an upper steel dome (not shown) that engages with the containment vessel to completely enclose the reactor core and reactor vessel.

[0075] During periods of reactor shut down, the plant electrical loads are provided by the grid and decay heat removal is provided by pony motors on the coolant pumps delivering reduced flow through the heat transport systems. In the event that grid power is not available, decay heat is removed using two dedicated safety class decay heat removal systems: the Reactor Vessel Air Cooling System (RVACS) and the Auxiliary Cooling System (ACS), which operate entirely by natural circulation with no need for electrical power.

[0076] In an embodiment, the TWR 200 may have a cylindrical geometry composed of hexagonally shaped fuel pin bundles, or assemblies. The assemblies contain a combination of enriched and depleted uranium metal alloy fuel pins. In an embodiment, the fuel pins are clad in ferritic-martensitic steel tubes.

[0077] In the center of the core are a few pins, or assemblies of pins, containing enriched uranium (235U), surrounded by pins or assemblies of depleted uranium (238U). The 235U serves as an igniter, kick starting the traveling wave reaction—a slow-moving chain reaction of parallel waves of fis-
sion traveling through the uranium rods. These parallel waves initiate in the center of the core, slowly consuming the fuel and generating heat in the core. In operation, the chain reaction creates a breed-burn zone in the core that does not move through fixed core material. Instead, a “standing” wave of breeding and burning is established by periodically moving core material in and out of the breed-burn region around the center of the core. This movement of fuel assemblies is referred to as “fuel shuffling.”

[0079] In the embodiment, metal fuel is used because it takes advantage of the high heavy metal loadings and excellent neutron economy, which allows an effective breed and burn process in TWRs. The uranium metal may be alloyed with 5 to 8% zirconium to dimensionally stabilize the alloy during irradiation and to inhibit low-temperature eutectic and corrosion damage of the cladding. A sodium thermal bond may be used to fill the gap that exists between the uranium alloy fuel and the inner wall of the clad tube to allow for fuel swelling and to provide efficient heat transfer which keeps the fuel temperatures low. Individual fuel pins may have a thin wire from 0.8 to about 1.6 mm diameter helically wrapped around the circumference of the clad tube to provide coolant space and mechanical separation of individual pins within the hexagonal fuel assembly housing that also serves as the coolant duct. The cladding, wire wrap and housing may be fabricated from ferritic-martensitic steel to take advantage of its superior irradiation performance.

Treatment with Supercritical Fluids

[0080] The embodiments shown in FIGS. 5-10 will be discussed with supercritical carbon dioxide (sCO₂) as the supercritical fluid. However, various other supercritical fluids, such as those discussed above, can also be used, and the application of the following embodiments is in no way limited to the application of sCO₂.

[0081] For ILs, the sCO₂ may be useful as a means of introducing uranium into the IL. In other cases, it may be appropriate to have direct dissolution of oxides into an IL.

[0082] Metals of interest to nuclear waste processing, such as actinides, lanthanides, and transition metals, have been characterized chemically using highly soluble fluorinated (3-diketones in sCO₂). Extraction can be accomplished by using appropriate chelating agents as extractants. For example, La and Eu extraction with greater than 90% effectiveness has been demonstrated using fluorinated diketones combined with tri-4-pyridylphosphate (TBP). In this process, a room temperature ionic liquid, an imidazolium-based 1-buty1-3-methylimidazolium (BMIIM) with bis(trifluoromethyl)sulfonyl)amide (also known as TI2N—, which is properly described as (CF3SO2)2N—) was used as a complexing agent because of the complexing agent’s ability to solubilize CO₂. In this manner, a full water/RTIL/sCO₂ system is developed. A similar process with other ionic liquids and metal chelating agents (extraction agents) was conducted and is summarized in Table IV. Note that Eu and La are both extracted with all systems except when using thenonyl tri-fluoracetone (TTA) without TBP. The latter only extracted La while not separating (extracting) Eu.

[0083] Using sCO₂ separation on target material and/or during the post-processing of nuclear fuel, β-diketones can be used to selectively bind with oxides or metal in the presence of uranium species. The extractions performed in Table IV were carried out with the extractant/sCO₂ mixture at 150 atm for one hour at 50°C. Based on this information, it is anticipated that β-diketones can be used to selectively bind with radioisotope oxides or metals while not substantially dissolving fissionable material.

<table>
<thead>
<tr>
<th>TABLE III-continued</th>
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<tbody>
<tr>
<td>Occurrence of selected elements in TWR spent fuel and IL solubility.</td>
</tr>
<tr>
<td>Element</td>
</tr>
<tr>
<td>_______</td>
</tr>
<tr>
<td>Sn</td>
</tr>
<tr>
<td>Mo</td>
</tr>
<tr>
<td>Cs</td>
</tr>
<tr>
<td>Tc</td>
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<td>Rh</td>
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<tr>
<td>Nd</td>
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<tr>
<td>Xe</td>
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<tr>
<td>Eu</td>
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</table>

<table>
<thead>
<tr>
<th>TABLE IV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Degree of extraction (%) of EuIII and LaIII from BMIIM12N with different beta-diketones (with or without TBP).</td>
</tr>
<tr>
<td>____________________</td>
</tr>
<tr>
<td>HFA w/o TBP</td>
</tr>
<tr>
<td>HFA w/TBP</td>
</tr>
<tr>
<td>TTA w/o TBP</td>
</tr>
<tr>
<td>TTA w/TBP</td>
</tr>
</tbody>
</table>

HFA = hexafluoroacetone,
TTA = 4,4,4-trifluoro-1(2-thienyl)-1,1,1-butanone

[0084] In general, an obstacle to CO₂ solvation is low solvent power of CO₂ (non-polar). Metals and metal chelates have low solubility in sCO₂ with CO₂ solubility parameters in the range of 4-5 cal/cm³. This can be overcome by adding CO₂-phlic functional groups such as fluoroethers, fluoro-
acrylates, fluoroalkyls, silicones, and certain phosphazenes. Fluorinated beta-diketones (with and without tributyl phosphosphate) have been demonstrated in current techniques to extract a variety of metals. Bis(trifluoroethyl) dithiocarbamate exhibits higher solubility than non-fluorinated counterparts; 10-4 mol/L for fluorinated vs. 10-6 to 10-7 mol/L for non-fluorinated. As another example, Diethylidithiocarbamate (DDC) can be 3-800 times less soluble in sCO2 at 100 atm than bis(trifluoroethyl)dithiocarbamate (FDSC). Since sCO2 density change is nearly linear with pressure, the solubility also changes nearly linearly with solubility increasing with increasing pressure. Lanthanides, actinides, copper, arsenic, and antimony (and other products of irradiated targets) can have concentrations on the order of 100 mg/L. Water and soil extraction has been demonstrated in current techniques with 1000-10000 molar ratio of chelate to metal in solution.

[0085] A system for removing fission products from salt-based fuels may be chemically similar to the process developed for metallic fuels. This is because salts, by themselves, are insoluble in sCO2. Extraction agents, such as diketones, may be used to draw select metals into the sCO2 phase as described above. Physically, the clean-up system may be made to avoid pressurization of the reactor vessel during a leak in the sCO2 clean-up system. Additionally, the salts in their liquid states may be at temperatures high enough to dissociate the diketones. To avoid both of these obstacles, a system may be designed such that the molten-salt is pumped external to the reactor vessel and injected into a vessel containing the sCO2. The sCO2 system may be maintained at a temperature low enough to solidify the molten-salt, resulting in a high surface area solid. Provided the sCO2 can be maintained at a sufficiently low temperature, the beta-diketones or other appropriate separation agent may be co-mixed with the sCO2 during salt injection, avoiding dissociation.

[0086] Alternatively, the separation agent may be injected into the system in a batch-wise fashion following salt injection. In either case, the result is a solution of (selected) metal-complexes solvated in the sCO2 diketone solution. The solution may then be pumped to a secondary system where temperature or pressure is adjusted to remove the metal complexes (product) from the solution without substantial effect on the target molten salt fuel. Again, it is likely that the metal complex is removable from the target solution without dropping the CO2 to a gaseous state (below the critical point) via heating, cooling, or both. Heat may be used to volatilize the metal complexes so that a separate gas phase occurs within the sCO2 solution. The sCO2 may alternatively be cooled or heated near and above the critical point where its solubility typically changes significantly with changes in temperature and pressure, resulting in a separate, liquid metal complex phase which was forced out of solution due changes in thermodynamic condition. This phase can then be transferred, such as by way of pumping, from the extraction system to system designed for interim or long term storage. Whether further heating or cooling is used to separate the metal complex or other product, ultimately further heating can be used to thermally decompose the diketones, leaving behind the metal fission product(s).

[0087] FIG. 5 is a block flow diagram of an embodiment of an example supercritical fluid treatment system 500. The example system 500 includes a reactor 502 and supercritical fluid treatment components 504, including fluid storage 506, supercritical fluid container 508, supercritical fluid control 510, extractant(s) storage 512, contact vessel 514, separation unit 516, transfer unit 518, and reprocessing/waste 520. Ancillary components, such as pumps, valves, sensors, etc., are not shown. Other embodiments can include more or fewer components.

[0088] The reactor 502 is a nuclear fuel reactor. For example, reactor 502 is a fuel salt reactor, a containerized fuel salt reactor, or a traveling wave reactor, as those reactors, their contents, and methods of operation, are described above. Reactor 502 may be a different type of reactor in other examples.

[0089] The supercritical fluid separation components 504 bring a supercritical fluid into contact with irradiated nuclear fuel and/or the reactor 502. Generally, supercritical fluids have properties that can permit selective separation of produced radionuclides from a target material, such as uranium, and/or remove residues containing fission products from equipment in or related to the reactor 502. Supercritical fluids retain both gas and liquid properties. They can have viscosities that resemble gas and diffusion properties between a gas and liquid. Supercritical fluids can effectively penetrate solid materials. Various supercritical fluids can be used for material processing and chemical reactions owing to their unique physico-chemical properties. Examples of supercritical fluids and some of their properties are discussed above.

[0090] Fluid CO2 is stored in fluid storage 506. There, the fluid CO2 is stored at a temperature and pressure such that the CO2 is in non-supercritical form. Fluid storage 506 can be a gas reservoir that is in fluid communication with the sCO2 container 508. However, as noted below with reference to separation unit 516, economics and other factors might dictate that sCO2 is not transitioned to the gas phase, in which case separation components 504 does not include fluid storage 506.

[0091] The sCO2 container 508 is pressurized and has a temperature such that the CO2 is in supercritical form. For example, sCO2 container 508 operates at a temperature greater than 32°C. and at a pressure greater than 73 atm.

[0092] The supercritical fluids used in system 500 can be combined with specific extractants to selectively remove or separate radionuclides. These extractants are stored in extractant storage 512 and mixed with the sCO2. Example extractants are discussed above, such as, for example, cupferron, chloroanillic acid, β-diketone, N-benzoyl-N-phenylhydroxylamine, α-dioximes dianisobenzidine, a porphyrine such as porphine, 8-hydroxyquinoline, nitroanilines, nitrophenols, ethylenediaminotetraacetic acid, diphenylcarbazide, diphenylcarbazone, Azoxyazo BN, sodium diethyldithiocarbamate, dithizone, bismuthioil, II-thiohenoxytrifluoroacetone, thioxone, thiophosphoric acid, phosphine sulfide, phosphorothioic acid, and tributylphosphate. In some embodiments, sCO2 is used without being mixed with any extractants.

[0093] As shown in FIG. 5, the extractants are mixed with the sCO2 within the supercritical fluid container 508, at a supercritical fluid control 510, or at some point before the sCO2 contacts the nuclear fuel. For example, the sCO2 is passed through a column containing the extractants to dissolve the extractants into the sCO2 stream. If the selected extractant is not particularly soluble in sCO2, this operation may also include modifying the extractant to make it more soluble, such as by adding CO2-philic functional groups such as fluoroethers, fluoroacrylates, fluoroalkyls, silicones, and certain phosphazenes to a selected ligand. In an embodiment, the extractant may be a fluorinated β-diketone and a trialkyl
phosphate, or a fluorinated β-diketone and a trialkylphosphine oxide. In another embodiment, the ligand may be selected from dithiocarbamates, thio-carbazones, β-diketones and crown ethers.

[0094] The scCO₂ and irradiated nuclear fuel from reactor 502 are introduced to the contact vessel 514. In embodiments, the scCO₂ is introduced directly into the reactor 502. The nuclear fuel includes a plurality of radioisotopes and radio-nucleides, such as, for example, ⁹⁰ᵐ⁹⁰⁸Mo, ²³⁴U, ¹³¹I, ¹³⁳Cr, ²²⁶Ra, and ²³⁵Ac.

[0095] In embodiments, irradiated nuclear fuel is injected into the contact vessel 514 via a fuel salt injector. The fuel salt injector includes one or more nozzles that disperse the molten fuel salt into a spray, mist, or fog of droplets. The contact vessel 514 can be operated as a batch or continuous process, or a combination of both.

[0096] Also, the contact vessel 514 can include an environmental control system. The environmental control system is capable of monitoring and regulating the temperature and pressure within the contact vessel 514, as well as the flow rates of the scCO₂ and nuclear fuel. In embodiments, the environmental control system includes a pressure sensor, a temperature sensor, a heater, such as a heating jacket, and a heat exchanger. The environmental control system can also include the fuel salt injector, thereby controlling the flow rate of nuclear fuel into the contact vessel and a supercritical fluid injection valve that controls the flow rate of supercritical fluid into the contact vessel 514. In embodiments, the environmental control system can maintain the contact vessel 514 at a temperature and pressure that causes the dispersed molten fuel salt to solidify into fuel salt particles.

[0097] The environmental control system can also regulate the flow of supercritical fluid and nuclear fuel out of the contact vessel 514. For example, one or more extraction valves, controlled by the environmental control system, can regulate the flow rate of supercritical fluid out of the contact vessel 514. In embodiments, the fluid flow out of the contact vessel is a combination of scCO₂ and nuclear fuel. In other embodiments, the nuclear fuel exits contact vessel 514 in one stream (and is routed to transfer unit 518), and the scCO₂ is routed to separation unit 516.

[0098] After contacting the nuclear fuel and/or reactor 502 components, the scCO₂ will have one or more fission products dissolved therein. Separation unit 516 separates the scCO₂ from the chelate and/or waste, and in embodiments, from the nuclear material. The scCO₂ exits the separation unit 516 and is routed back to fluid storage 506 or supercritical fluid container 508. The nuclear material exits the separation unit 516 and is routed to the transfer unit 518, and the chelate and/or waste exits the separation unit 516 and is routed to reprocessing/waste 520.

[0099] It may be impractical to transition scCO₂ to the gas phase in separation unit 516 economically because that transition would require either re-compression of the CO₂ to the supercritical state or a steady supply of high pressure CO₂. Additionally, there is a potential safety risk inherent to confining a high pressure solution of a highly compressible fluid. Furthermore, the off-gas CO₂ would need to be collected in a container capable of further decontamination or disposal, because some residual radioactive materials or decay products might remain in the carbon dioxide gas.

[0100] In embodiments, the separation unit 516 includes a “back extraction” process which does not require gasification of the scCO₂ as part of the separation of the radioisotopes from the scCO₂. In this type of process, metal or metalloid species are removed from solid or liquid solutions by using supercritical fluids to form a metal or metalloid chelate. The supercritical fluid will typically contain a solvent extractant, such as a few percent H₂O or MeOH. The metals or metalloids are then back-extracted from the scCO₂ solution by using an acidic solution, one of which may be halogenated. By back-extracting to another (aqueous) solution, decompression of the scCO₂ is avoided. What is left is the other solution bearing the selected radioisotopes, which is routed to reprocessing/waste 520, and the scCO₂ that can be readily reused by routing it to the supercritical fluid container 508.

[0101] Using a back extraction process can be advantageous in an automated system and in a continuous treatment, although even in a semi-automated, batch treatment system the ability to recycle scCO₂ without the added step of repressurization would be cost-advantageous. Back extraction may, or may not, remove the extractant with the radioisotope. In an embodiment, fresh extractant may need to be added to the scCO₂ before it can be reused as an extraction compound. It should be noted that ILs could also be used for the back extraction process.

[0102] Alternatively, in separation unit 516 the solution removed from the contact vessel 514, containing the scCO₂, elements and isotopes removed from the used fuel, can be brought to below the critical point and converted to the gaseous phase. This conversion leaves behind the extractant ligand and the separated elements or isotopes. The extractant can then be brought to above its volatilization temperature and converted to a vapor phase, leaving behind the selected element or isotopes. Variations of this scheme may be used as appropriate. For example, lowering the solution to below the liquidus point of the carbon dioxide may be preferred if the chosen extractant and liquid CO₂ are insoluble.

[0103] Another alternative is to, in separation unit 516, raise the temperature of the supercritical solution to above the volatilization point of the extractant (e.g. greater than 100° C. to 200° C.) or to above the decomposition temperature (e.g. greater than 200° C. to 300° C.). In either case, the metal may substantially or partially precipitate from the scCO₂ once the extractant is lost. Removal of the extractant vapor or decomposition product can be accomplished by a gas phase separation or, as above, by converting the CO₂ to a liquid phase. Furthermore, the solution may change temperature or pressure from a supercritical condition to a second supercritical condition, the second condition having a solubility of the extractant lower than the solubility of the first condition. By this process, all or a portion of the extractant may be recovered without leaving the supercritical state.

[0104] Current techniques use scCO₂-ionic liquid processes to remove both lanthanides and actinides from aqueous solutions, and some current modified versions have been proposed. One such process is called super-DIREX, which is short for ‘supercritical direct extraction’. The super-DIREX process is expected to minimize the cost of reprocessing because the heavy metals (U, Pu, Np, Am and Cm) are directly extracted from a spent fuel powder in a column covering the dissolution. One experiment claimed up to 30% reduction in waste stream volumes if scCO₂ methods were utilized. However, other work has cited other critical issues, such as counteracting the acidity of TBP-HNO₃ solutions and the build-up of ¹⁴C.

[0105] Metallic fuel, including those metal fuels appropriate for vented pin configurations and/or a traveling wave
reactor, typically includes metal fuel capable of high burn-up contained within vented, ferritic martensitic stainless steel cladding. At the end of life, the fuel generally has a highly porous matrix of metallic form fuel and solid fission products which precipitated from the fuel during the burn cycle.

[0106] The transfer unit 518 is an optional component that circulates the liquid nuclear fuel between the reactor 502 and the contact vessel 514. The transfer unit 518 can include one or more pumps, valves, sensors, and flow meters. Additionally, the transfer unit 518 can include a holding vessel for holding the nuclear material and raising the temperature and/or pressure of the nuclear material.

[0107] As shown, the transfer unit 518 directs nuclear material back to the reactor 502. Alternatively, some or all of the nuclear material can be directed to waste processing. Another alternative is to re-use the nuclear material, once the fissions products are removed in contact vessel 514 and/or separation unit 516, in another facility similar to or different from the reactor 502. For example, the reactor 502 is a breeder and burn type reactor such as a TWR. It may be practical to remove the fission products and then perform a thermo-mechanical treatment within the contact vessel 514 and/or separation unit 516 used for solvation in order to modify the structural material for continued in-reactor use. Once the fission products are removed, the nuclear material may be brought to significantly higher temperatures (which could be made to exceed the fuel melting point) and pressures (10’s of MPa’s).

[0108] Reprocessing/waste system 520 includes one or more processes and/or storage facilities. As shown, the extractants, such as chelates and ligands, as well as fission products, are routed to reprocessing/waste system 520. Through one or more processes, the ligands can be separated from the fission products. In some instances, the separated ligands can be reused in the system 500.

[0109] FIG. 6 is an embodiment of a method 600 for treating a fuel salt reactor with a supercritical fluid treatment system. The example method 600 includes generating a supercritical fluid (operation 602), introducing an extractant into the supercritical fluid (operation 604), contacting a volume of the supercritical fluid with a molten fuel salt (operation 606), separating the supercritical fluid from the fuel salt (operation 608), directing contacted fuel salt to a reactor core (operation 610), and separating fission products from the supercritical fluid (operation 612). The supercritical fluid treatment system 504 shown and described with reference to FIG. 5, and any one of the reactors described above with reference to FIGS. 1-5, can be used to implement example method 600. Other embodiments can include more or fewer operations.

[0110] The example method 600 begins by generating a supercritical fluid in a fluid providing operation 602. As discussed above, the supercritical fluid can be supercritical carbon dioxide (sCO₂) which can be generated at using known techniques. Once the supercritical fluid is generated, an extractant is introduced into the supercritical fluid such that the ligand is dissolved into the supercritical fluid in an extractant adding operation. As discussed above the extractant adding operation may include modifying or preparing the selected extractant for effective use with the chosen supercritical fluid.

[0111] Next, a volume of irradiated, molten fuel salt is contacted with a volume of the supercritical fluid in a contacting operation 606. This contacting can occur within the reactor core or in a separate contact vessel, as described above with reference to FIG. 5.

[0112] During contact, the supercritical fluid removes one or more types of fission products from the irradiated fuel salt. The extractant in the supercritical fluid forms a complex with one or more fission products, resulting in a sCO₂-radioisotope complex solution.

[0113] The supercritical fluid is then separated from the fuel salt in a separation operation 608. In embodiments, the contacted fuel salt is directed back to the reactor core in a fuel return operation 610. As discussed above with reference to FIG. 5, the temperature and pressure of the fuel salt may need to be increased before re-introduction into the reactor core.

[0114] The separated supercritical fluid is also subjected to one or more separation processes to extract the supercritical fluid from the fission products in a separation operation 612. Techniques and conditions for this separation are discussed above with reference to FIG. 5.

[0115] FIG. 7 is a block flow diagram of the example fuel salt reactor 100 used with the example supercritical fluid treatment components 504. The example fuel salt reactor 100 is shown and described above with reference to at least FIGS. 1-3. The example supercritical fluid treatment components are also shown and described above with reference to FIG. 5. Other embodiments can include more or fewer components.

[0116] In the embodiment shown, molten fuel salt is fed into the contact vessel 514 from reactor 100. The fuel salt returns to the reactor 100 via transfer unit 518 and the supercritical fluid, including the chelates in solution, are sent to the separation unit 516 for additional processing.

[0117] Alternatively, the supercritical fluid is fed directly into the reactor 100 during reactor operation. In another embodiment, the supercritical fluid is fed into the reactor 100 during a reactor shut-down period where little to no fuel salt is present.

[0118] FIG. 8 is a block flow diagram of the example containerized fuel salt reactor 400 used with the example supercritical fluid treatment components 504. The example fuel salt reactor 400 is shown and described above with reference to at least FIGS. 1-4. The example supercritical fluid treatment components are also shown and described above with reference to FIG. 5. Other embodiments can include more or fewer components.

[0119] The reactor 400 includes one or more fuel salt containers 490, each of which containing fuel salt that includes at least some fissionable material. The containers 490 may be storage containers, or, alternatively may be containers for use within the reactor such as fuel tubes or assemblies.

[0120] In the embodiment shown, supercritical fluid is directed into one or more fuel containers 490. The containers 490 may be located in the reactor core or may be removed for treatment. After treatment, the containers 490 may be returned to the reactor core or stored. The containers 490 may hold a fuel salt which may be molten or solid, or may hold another nuclear fuel such as loose particulate fuel or porous matrix fuel as described with reference to the TWR above. The flow rate of supercritical fluid into the fuel container 490 is regulated by the supercritical fluid control 510. Inside the fuel container 490, the supercritical fluid contacts the fuel. As discussed above, one or more fission products are extracted by the supercritical fluid during this contact by dissolution. Then, the supercritical fluid exits the fuel container 490 and is directed to the separation unit 516. In embodiments, the flow
rate of supercritical fluid out of the fuel container 490 is regulated by a controller in communication with a sensor and valve. [0121] In embodiments, the supercritical fluid is used to prepare the fuel container 490 for shut-down. Removing fission products from the fuel container 490 may greatly enhance the disposability of the fuel container 490, as >90% of targeted fission products may be removed using scO₂ treatment, and even more can be removed with multiple scO₂ solution treatments. In some cases, it may be advantageous to apply multiple cycles such as repeated treatments or multiple different treatments to increase the removal of fission products. For example, in some cases, two treatments could give 99% removal of accessible fission products. Three would give 99.9% and so forth. [0122] Any appropriate factors may be used to determine the number and/or type of processing treatments and may be based on fission products dissolved or stuck inside the solid fuel matrix where scO₂ solution cannot penetrate. It should be noted, however, that it may be possible to operate at temperature and timescales which would allow for diffusion of solution soluble metals out of the bulk fuel matrix and into solution. This may lower the short term heat load of the spent fuel assembly, decrease the dangers of handling and transporting the assembly, and make it more suitable for long-term disposal. [0123] FIG. 9 is a block flow diagram of the example TWR 900 used with the example supercritical fluid treatment components of FIG. 5. The example TWR 900 is shown and described above with reference to at least FIG. 4. The example supercritical fluid treatment components are also shown and described above with reference to FIG. 5. Other embodiments can include more or fewer components. [0124] The TWR 900 includes a reactor core and a reactor vessel containing a primary sodium coolant. The reactor core is submerged with the primary sodium coolant. The TWR 900 also includes at least one assembly 904 in the reactor core that includes one or more solid fuel pins that contain fissionable material and fission products. Also, the TWR 900 includes an assembly shuffling system that is configured to move the assemblies between various positions within the reactor core. [0125] The supercritical fluid can contact nuclear fuel from the TWR 900 in a contact vessel 514, where the nuclear fuel is directed away from the reactor core. Alternatively, the supercritical fluid can be contacted with an assembly when the shuffling system has removed, temporarily, the assembly from the reactor core. Still another alternative is that the supercritical fluid can contact nuclear fuel directly within the reactor core. [0126] The supercritical fluid control 510 can control the transfer of supercritical fluid to the TWR 900 into the fuel assembly 904 and/or contact vessel 514. The control can be based on the expansion of the nuclear fuel and/or based on the concentration of fission products in a coolant used in the TWR 900. [0127] In embodiments, the TWR 900 includes a transfer vessel that is configured to hold a fuel assembly 904 in argon. The supercritical fluid control 510 can direct supercritical fluid to contact a fuel assembly 904 that is held in argon, not shown in FIG. 9. There, the supercritical fluid can remove fission products from the argon that is exposed to the fuel assembly 904. The supercritical fluid, now a mixture with fission products, is returned to the separation unit 516 as discussed above. [0128] In embodiments, the TWR 900 includes a coolant cleaning system that includes an absorber 902. The absorber, such as a packed bed or adsorption membrane, removes fission products from the primary sodium coolant used in the TWR 900. The supercritical fluid control 510 can direct supercritical fluid to the absorber 902 to dissolve and remove fission products from the absorber 902. The supercritical fluid, now a mixture with fission products, is returned to the separation unit 516 as discussed above. [0129] A system using supercritical fluid treatments may remove fission products prior to the end of life by incorporating the separation process such as scO₂ process into the fuel management or ‘shuffling’ cycle to remove fission products periodically during irradiation (operation of the reactor). For example, a TWR re-fueling system may incorporate a sealed enclosure for raising the assembly out of the coolant pool. The TWR containment enclosure may be provided with sufficient cooling capability to manage assembly decay heat during the treatment process while the assembly 904 is out of the coolant pool. In an alternative embodiment, the system may be designed that one or more assembly positions around the periphery of the core are treatment positions where the assemblies may be connected to the treatment system 504. The system may be made more robust such that fission products may be removed, in containment, with minimal system modifications. Such a system would not require large vessels and piping, due to the high density of scO₂. Concentrations of greater than 10-4 kg metal/kg solution are possible. At the end of life, each assembly contains the maximum amount of fission products, on the order of 50 kg. The solution density is on the order of 1000 kg/m³. Therefore only 5 m³ of scO₂ solution would be needed in some cases to contain all the fission products in a single assembly. Treating the assembly at more frequent intervals would obviously reduce this maximum volume. Furthermore, since the CO₂ may be separated from the fission products and re-entered into the system, the inventory can be additionally reduced. [0130] FIG. 10 is an embodiment of a method 700 for operating a reactor with supercritical fluid separation. The example method 700 includes charging the reactor core (operation 702), maintaining a chain reaction (operation 704), contacting a volume of reacted fuel with a supercritical fluid (operation 706), initiating a chain reaction in regenerated fuel (operation 708), and separating fission products from the supercritical fluid (operation 710). The supercritical fluid treatment system 504 shown and described with reference to FIG. 5, and the reactor 100 described above at least with reference to FIGS. 1-3, can be used to implement example method 700. Other embodiments can include more or fewer operations. [0131] The example method 700 begins by charging the reactor core with nuclear fuel in a charging operation 702. The charging operation 702 varies depending upon the type of nuclear reactor employed in the example method 700. Examples of charging a molten salt reactor, a traveling wave reactor, and a containerized molten salt reactor are described above. [0132] After charging the reactor core in charging operation 702, a chain reaction is maintained within the reactor core at or above criticality in a first fission operation 704. Maintaining the chain reaction varies depending upon the type of nuclear reactor employed in the example method 700. [0133] At some point after the chain reaction is started and maintained, a volume of supercritical fluid is contacted with a
volume of irradiated nuclear fuel. As discussed above, supercritical carbon dioxide (sCO₂) can be used as a supercritical fluid. Contacting the supercritical fluid and nuclear fuel varies depending on the specific type of reactor used, with some examples discussed above.

As an example, and as discussed above, the supercritical fluid can be contacted with at least some of the partially-reacted nuclear fuel without removing the nuclear fuel from the reactor core. This introduction of supercritical fluid can be performed without interrupting one or more chain reactions within the reactor core.

As another example, and as discussed above, some of the partially-reacted fuel can be removed from the reactor core and then contacted with the supercritical fluid, for example in a contact vessel. The removal of the partially-reacted fuel can be performed without interrupting one or more chain reactions within the reactor core. After contacting with the supercritical fluid, the partially-reacted fuel can be returned to the reactor core without interrupting the one or more chain reactions within the reactor core.

As discussed above with reference to the TWR, the nuclear fuel can be contained within one or more fuel pins or pin assemblies. The pins can be moved within the reactor core to different positions throughout operation. During or after movement of a pin, the supercritical fluid can be contacted with the fuel within the pin.

After the contacting the supercritical fluid and partially-reacted nuclear fuel in contacting operation 706, a chain reaction is initiated in the now-treated nuclear fuel in a second fission operation 708. As discussed above, in embodiments the partially-reacted nuclear fuel can be contacted with supercritical fluid within the reactor. In those embodiments, the reactor operation can continue as before. In other embodiments, the partially-reacted nuclear fuel is directed out of the reactor core and then is contacted with the supercritical fluid. In those embodiments, the partially-reacted nuclear fuel can be redirected back to the same reactor core or directed to a different, perhaps newly starting, reactor core.

The method 700 also includes separating the fission products from the supercritical fluid in a separation operation 710. Embodiments of systems and methods for separating fission products from the supercritical fluid are discussed at least with reference to FIG. 5.

Yet another possible application of supercritical fluids is reformulation of spent or previously-irradiated fuel. Ref ormation of fuel after irradiation generally may be designed to allow treatment of the entire fuel assembly for fission product, lanthanide, or actinide removal treatments without modification of the nuclear fuel assembly or fuel pins contained within. In one example of a sealed vessel and supercritical fluid treatment, a previously burned nuclear fuel assembly may be placed into a sealed pressure vessel. The vessel may then be filled with pressurized sCO₂ and one or more extraction agent (such as diketones, or any other appropriate agent) is added to create an extracting solution in the absence of an IL or aqueous component. Because of the presence of a vent in the existing fuel assembly for fission gas venting, and the nature of supercritical fluids, the sCO₂ extractant solution will work to fill the fuel pin and the matrix of porous fuel (i.e. supercritical fluids behave as low surface tension, low viscosity fluids which fill the volume they are contained within). The sCO₂ solution will begin to solvate targeted fission products (or other materials, if so desired and a proper ligand chosen), leaving the uranium metal matrix unaffected. The fission products will then begin to diffuse out of the fuel pin such that the concentration of the overall system tending toward equilibrium. The solution containing the dissolved metal can then be slowly released from the pressure vessel. New, clean solution may be re-added to the pressure vessel. Agitation, heat and/or continued pressurization and depressurization may be applied to the system to enhance the solvation rate. For example, the system may operate at greater than 7.5 MPa (approximate critical point at 51° C.) and be oscillated by ±0.1 MPa to enhance "pumping" of sCO₂ solution in and out of the porous fuel.

CONCLUSION

While particular aspects of the present subject matter described herein have been shown and described, it will be apparent to those skilled in the art that, based upon the teachings herein, changes and modifications may be made without departing from the subject matter described herein and its broader aspects and, therefore, the appended claims are to encompass within their scope all such changes and modifications as are within the true spirit and scope of the subject matter described herein. It will be understood by those within the art that, in general, terms used herein, and especially in the appended claims (e.g., bodies of the appended claims) are generally intended as “open” terms (e.g., the term “including” should be interpreted as “including but not limited to,” the term “having” should be interpreted as “having at least,” the term “includes” should be interpreted as “includes but is not limited to,” etc.). It will be further understood by those within the art that if a specific number of an introduced claim recitation is intended, such an intent will be explicitly recited in the claim, and in the absence of such recitation no such intent is present.

For example, as an aid to understanding, the following appended claims may contain usage of the introductory phrases “at least one” and “one or more” to introduce claim recitations. However, the use of such phrases should not be construed to imply that the introduction of a claim recitation by the indefinite articles “a” or “an” limits any particular claim containing such introduced claim recitation to claims containing only one such recitation, even when the same claim includes the introductory phrases “one or more” or “at least one” and indefinite articles such as “a” or “an” (e.g., “a” and/or “an” should typically be interpreted to mean “at least one” or “one or more”); the same holds true for the use of definite articles used to introduce claim recitations. In addition, even if a specific number of an introduced claim recitation is explicitly recited, those skilled in the art will recognize that such recitation should typically be interpreted to mean at least the recited number (e.g., the bare recitation of “two recitations,” without other modifiers, typically means at least two recitations, or two or more recitations).

Unless otherwise indicated, all numbers expressing quantities of ingredients, properties such as molecular weight, reaction conditions, and so forth used in the specification and claims are to be understood as being modified in all instances by the term “about.” The term “about” is not intended to either expand or limit the degree of equivalents which may otherwise be afforded a particular value. Further, unless otherwise stated, the term “about” shall expressly include “exactly,” consistent with the discussions regarding ranges and numerical data. The term “about” in the context of the present disclosure means a value within 15% (±15%) of the value recited immediately after the term “about,” includ-
ing any numeric value within this range, the value equal to the upper limit (i.e., +15%) and the value equal to the lower limit (i.e., −15%) of this range. For example, the value “100” encompasses any numeric value that is between 85 and 115, including 85 and 115 (with the exception of “100%”, which always has an upper limit of 100%).

[0143] Concentrations, amounts, and other numerical data may be expressed or presented herein in a range format. It is to be understood that such a range format is used merely for convenience and brevity and thus should be interpreted flexibly to include not only the numerical values explicitly recited as the limits of the range, but also to include all the individual numerical values or sub-ranges encompassed within that range as if each numerical value and sub-range is explicitly recited. As an illustration, a numerical range of “4% to 7%” should be interpreted to include not only the explicitly recited values of about 4 percent to about 7 percent, but also include individual values and sub-ranges within the indicated range. Thus, included in this numerical range are individual values such as 4.5, 5.25, and 6 and sub-ranges such as from 4.5–5, from 5–5.7, and from 5.5–6.5; etc. This same principle applies to ranges reciting only one numerical value. Furthermore, such an interpretation should apply regardless of the breadth of the range or the characteristics being described.

[0144] Notwithstanding that the numerical ranges and parameters setting forth the broad scope of the invention are approximations, the numerical values set forth in the specific examples are reported as precisely as possible. Any numerical value, however, inherently contain certain errors necessarily resulting from the standard deviation found in their respective testing measurements.

[0145] Furthermore, in those instances where a convention analogous to “at least one of A, B, and C, etc.” is used, in general such a construction is intended in the sense one having skill in the art would understand the convention (e.g., “a system having at least one of A, B, and C” would include but not be limited to systems that have A alone, B alone, C alone, A and B together, A and C together, B and C together, and/or A, B, and C together, etc.). In those instances where a convention analogous to “at least one of A, B, or C, etc.” is used, in general such a construction is intended in the sense one having skill in the art would understand the convention (e.g., “a system having at least one of A, B, or C” would include but not be limited to systems that have A alone, B alone, C alone, A and B together, A and C together, B and C together, and/or A, B, and C together, etc.). It will be further understood by those within the art that typically a disjunctive word and/or phrase presenting two or more alternative terms, whether in the description, claims, or drawings, should be understood to contemplate the possibilities of including one of the terms, either of the terms, or both terms unless context dictates otherwise. For example, the phrase “A or B” will be typically understood to include the possibilities of “A” or “B” or “A and B.”

[0146] In some instances, one or more components may be referred to herein as “configured to,” “configurable to,” “operable/operative to,” “adapted/adaptive,” “able to,” “conformable/conformed to,” etc. Those skilled in the art will recognize that such terms (e.g., “configured to”) can generally encompass active-state components and/or inactive-state components and/or standby-state components, unless context requires otherwise.

[0147] With respect to the appended claims, those skilled in the art will appreciate that recited operations therein may generally be performed in any order. Also, although various operational flows are presented in a sequence(s), it should be understood that the various operations may be performed in other orders than those which are illustrated, or may be performed concurrently. Examples of such alternate orderings may include overlapping, interleaved, interrupted, reordered, incremental, preparatory, supplemental, simultaneous, reverse, or other variant orderings, unless context dictates otherwise. Furthermore, terms like “responsive to,” “related to,” or other past-tense adjectives are generally not intended to exclude such variants, unless context dictates otherwise.

[0148] It will be clear that the systems and methods described herein are well adapted to attain the ends and advantages mentioned as well as those inherent therein. Those skilled in the art will recognize that the methods and systems within this specification may be implemented in many manners and as such is not to be limited by the foregoing exemplified embodiments and examples. In this regard, any number of the features of the different embodiments described herein may be combined into one single embodiment and alternate embodiments having fewer than or more than all of the features herein described are possible.

[0149] While various embodiments have been described for purposes of this disclosure, various changes and modifications may be made which are well within the scope of the technology described herein. For example, although not explicitly stated Raman spectroscopy may be but one of many techniques used to monitor fuel salt quality during operation of a molten salt reactor and, likewise, multiple Raman probes may be used in order to get an understanding of the variations in fuel salt quality at different locations within the reactor. Numerous other changes may be made which will readily suggest themselves to those skilled in the art and which are encompassed in the spirit of the disclosure and as defined in the appended claims.

What is claimed is:
1. A nuclear fission reactor comprising:
a reactor core containing a quantity of fuel salt including at least some fissionable material and adapted to create a chain reaction in the fuel salt, thereby generating heat and fission products in the fuel salt;

at least one heat exchanger adapted to transfer heat from the fuel salt to a coolant;
apower generation unit that converts heat in the coolant into power; and

a supercritical fluid separation system adapted to remove at least some amount of fission products from the fuel salt.

2. The nuclear fission reactor of claim 1 wherein the supercritical fluid separation system comprises:
a supercritical fluid contact vessel which contacts fuel salt with a supercritical fluid;
a fuel salt transfer unit that circulates fuel salt between the reactor core and the supercritical fluid contact vessel;
a supercritical fluid source fluidly connected to the supercritical fluid contact vessel, the supercritical fluid including at least one ligand that dissolves at least one fission product into the supercritical fluid when the ligand contacts the at least one fission product; and

a controller controlling the transfer of fuel salt between the reactor core and supercritical fluid contact vessel and controlling the transfer of supercritical fluid from the supercritical fuel source through the supercritical fluid contact vessel.
3. The nuclear fission reactor of claim 2 wherein the fuel salt transfer unit maintains the fuel salt in a molten state when transferring the fuel salt between the reactor core and the contact vessel.

4. The nuclear fission reactor of claim 3 wherein the fuel salt remains in a molten state while in the contact vessel during contact with the supercritical fluid.

5. The nuclear fission reactor of claim 3 wherein the fuel salt transfer unit maintains the fuel salt in a molten state when transferring the fuel salt from the reactor core to the contact vessel, and the contact vessel further comprises:
   a fuel salt injector;
   a contact vessel environmental control system; and
   wherein the controller operates the injector and environmental control system so that the molten fuel salt solidifies into fuel salt particles upon injection into the contact vessel.

6. The nuclear fission reactor of claim 5 wherein the fuel salt transfer unit removes the fuel salt particles from the contact vessel after the particles contact the supercritical fluid.

7. The nuclear fission reactor of claim 6 wherein the fuel salt transfer unit melts the fuel salt particles to a molten fuel before returning the molten fuel salt to the reactor core.

8. The nuclear fission reactor of claim 5 wherein the fuel salt injector comprises:
   one or more nozzles adapted to disperse the molten fuel salt into a spray of drops within the supercritical fluid contained in the contact vessel, wherein the contact vessel is maintained at a temperature and pressure by the environmental control system that causes the dispersed molten fuel salt to solidify into fuel salt particles.

9. The nuclear fission reactor of claim 5 wherein the contact vessel environmental control system comprises one or more of:
   a pressure sensor;
   a temperature sensor;
   a heater;
   a heat exchanger;
   a fuel salt injection valve controlling the rate of flow of the molten fuel salt into the contact vessel;
   a supercritical fluid injection valve controlling the rate of flow of supercritical fluid into the contact vessel; and
   a supercritical fluid extraction valve controlling the rate of flow of supercritical fluid out of the contact vessel.

10. The nuclear fission reactor of claim 2, wherein the supercritical fluid flowing out of the contact vessel includes an amount of fission products removed from the fuel salt and wherein the supercritical fluid separation system further comprises:
    a separation vessel fluidly connected to the contact vessel to receive the supercritical fluid flowing out of the contact vessel and separate at least some of the amount of fission products from the supercritical fluid.

11. A nuclear fission reactor comprising:
    one or more fuel salt containers, including a first fuel salt container containing a quantity of fuel salt including at least some fissionable material;
    a reactor core adapted to hold at least the first fuel salt container and adapted to create a chain reaction in the fuel salt, thereby generating heat in the fuel salt;
    at least one heat exchanger adapted to transfer heat from the fuel salt to a coolant; and
    a supercritical fluid extraction system adapted to pass a supercritical fluid through the first fuel salt container, the supercritical fluid extraction system including
    a supercritical fluid source fluidly connectable to the first fuel salt container that, when connected, delivers the supercritical fluid into the first fuel salt container;
    a separation vessel fluidly connectable to the first fuel salt container that receives the supercritical fluid from the first fuel salt container; and
    a controller controlling the transfer of supercritical fluid from the source through the first fuel salt container into the separation vessel.

12. A method of operating a nuclear fission reactor comprising:
    charging a reactor core with an initial fuel containing fissionable material;
    maintaining a first chain reaction in the reactor core for a period of time, thereby generating a partially-reacted fuel containing less fissionable material than in the initial fuel and at least some amount of fission products greater than in the initial fuel;
    contacting at least some of the partially-reacted fuel with a supercritical fluid containing at least one ligand that forms a metal complex with at least one fission product, thereby creating a supercritical fluid and fission product mixture and a regenerated fuel containing a lower amount of fission products than in the partially-reacted fuel;
    separating the supercritical fluid and fission product mixture from the regenerated fuel;
    initiating a second chain reaction in the regenerated fuel; and
    extracting at least some of the fission product from the supercritical fluid and fission product mixture.

13. The method of claim 12 wherein the contacting is performed on the at least some of the partially-reacted fuel without removing it from the reactor core.

14. The method of claim 13 wherein the contacting is performed without interrupting the first chain reaction in the reactor core.

15. The method of claim 12 further comprising:
    removing the at least some of the partially-reacted fuel from the reactor core without interrupting the first chain reaction;
    performing the contacting operation after removing the at least some of the partially-reacted fuel from the reactor core; and
    returning the regenerated fuel to the reactor core.

16. The method of claim 15, further comprising:
    returning the regenerated fuel to the reactor without interrupting the first chain reaction.

17. The method of claim 12 wherein the nuclear fission reactor is a traveling wave reactor and the method further comprises:
    charging the reactor core with a plurality of pins containing the initial fuel containing fissionable material;
    maintaining the first chain reaction in a breed-burn region of reactor core for a period of time, thereby generating at least one pin containing partially-reacted fuel;
    moving the at least one pin containing partially-reacted fuel to a different position within the reactor; and
    during or after the moving operation, contacting the partially-reacted fuel in the at least one pin by passing the
supercritical fluid containing at least one ligand through the at least one pin to obtain at least one pin containing regenerated fuel.

18. The method of claim 17, wherein the moving operation is part of a pin shuffling operation.

19. The method of claim 17 wherein initiating a second chain reaction in the regenerated fuel is achieved by placing the at least one pin containing the regenerated fuel at a location in the breed-burn region of the reactor core.

20. The method of claim 12 wherein the nuclear fission reactor is a molten salt reactor and wherein the contacting is performed on partially-reacted molten salt fuel without removing it from the reactor core.

21. The method of claim 20, further comprising:
   removing the at least some of the partially-reacted molten salt fuel from the reactor core without interrupting the first chain reaction;
   performing the contacting operation after removing the at least some of the partially-reacted molten salt fuel from the reactor core; and
   returning regenerated molten salt fuel to the reactor core.

22. The method of claim 21, further comprising:
   returning the regenerated molten salt fuel to the reactor without interrupting the first chain reaction.

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