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(54) **Apparatus and method for reprocessing and separating spent nuclear fuels**

(57) A method and apparatus for separating and reprocessing spent nuclear fuels includes a separation vessel housing a molten metal solvent in a reaction region, a reflux region positioned above and adjacent to the reaction region, and a porous filter member defining the bottom of the separation vessel in a supporting relationship with the metal solvent. Spent fuels are added to the metal solvent. A non-oxidizing nitrogen-containing gas is introduced into the separation vessel, forming solid actinide nitrides in the metal solvent from actinide fuels, while leaving other fission products in solution. A pressure of about 1.1 to 1.2 atm is applied in the reflux region, forcing the molten metal solvent and soluble fission products out of the vessel, while leaving the solid actinide nitrides in the separation vessel.
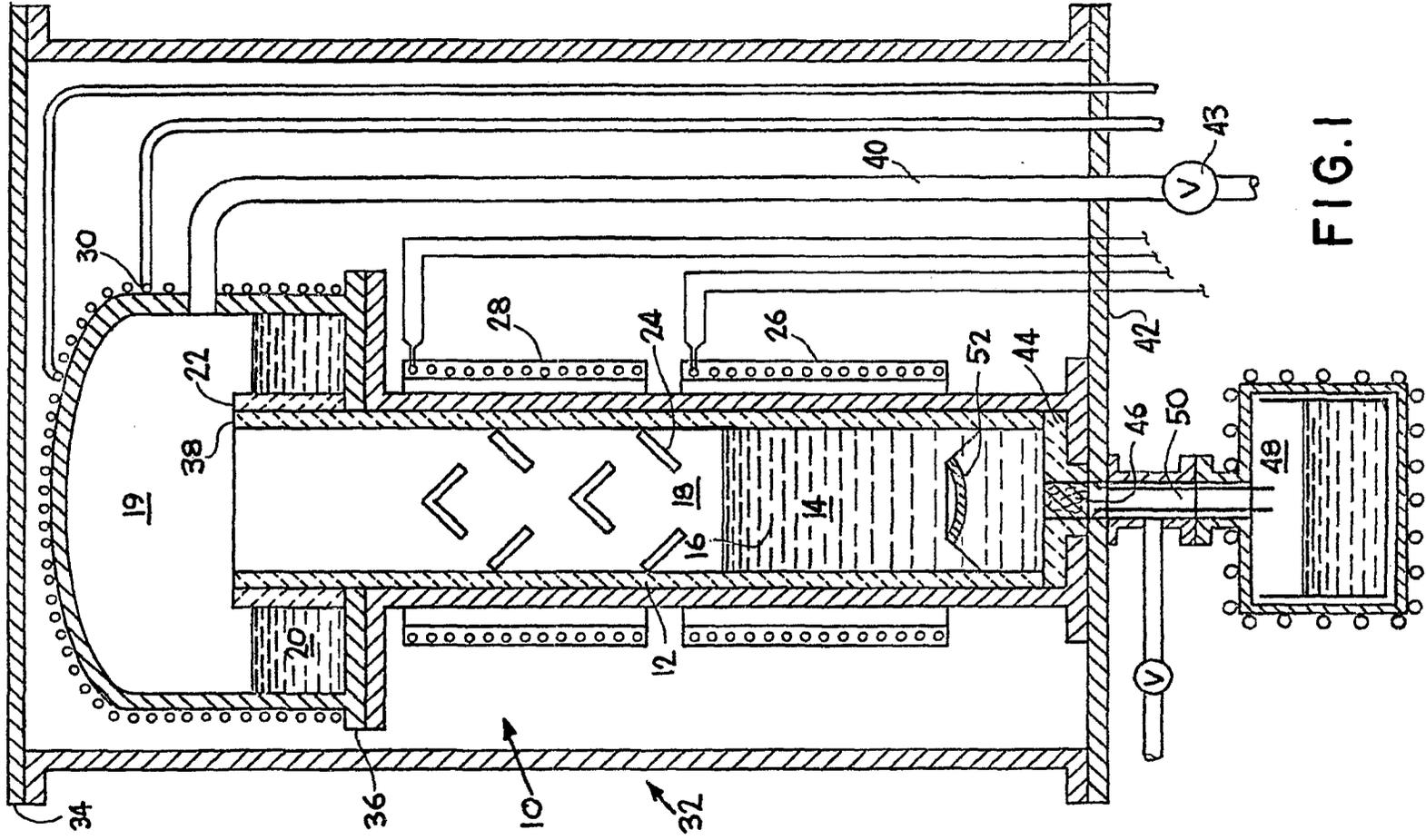
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FIG. I

SPECIFICATION
Apparatus and method for reprocessing and separating spent nuclear fuels

Background of the invention

The present invention relates generally to a method and apparatus for the reprocessing and
5 separating of spent nuclear fuels contaminated with fission products in a molten metal solvent. More   5
particularly, the present invention is directed to a method and apparatus for reprocessing and
separating spent fuel elements in a separation apparatus housing a molten metal solvent, and including
a porous filter member to remove the molten solvent and compounds soluble therein from the
separation apparatus.
10   A molten metal solvent, usually tin, has been used as the solvent and reaction medium in the   10
reprocessing and separation of spent nuclear fuel elements, including actinide fuels, e.g., the removal
of fission products and other impurities from spent uranium-plutonium and thorium-uranium
(plutonium) fuels in oxide, metal, or carbide form. Initially, the spent fuel is declad, if necessary, then
put into a solution of molten tin maintained at a temperature of about 1600°C. If the fuel is an oxide a
15 carbothermic reduction process is necessary. For this purpose, the separation vessel housing the   15
molten tin is comprised of a material which is a source of carbon, preferably graphite. Carbon dissolves
in the molten tin and reacts with the actinide and fission product oxides, converting them to a metallic
solution and generating CO gas. For a uranium-plutonium fuel the principal reaction is represented as:

$$(U,Pu)O_{2(s)}+2C_{(in\ Sn\ solution)}\rightarrow(U,Pu)_{(in\ Sn\ solution)}+2CO_{(g)} \qquad (1)$$

20   During dissolution of the fuel, volatile fission products are released and swept out of the   20
separation vessel by the CO, while the more refractory fission products remain behind in the molten tin
solvent. Although the volatile fission products have varying volatilities in the molten tin, they are all
removed in one step. Along with the volatile fission products a significant portion of the molten tin
solvent also evaporates and is removed from the vessel.
25   Thereafter, the actinides (in solution) are separated from the majority of the fission products   25
remaining in the molten tin in a nitriding reaction. A non-oxidizing nitrogen containing atmosphere is
introduced into the vessel, resulting in the formation of actinide nitrides in the molten tin. For a
uranium-plutonium fuel the reaction is represented by the following equation:

$$(U,Pu)_{(in\ Sn\ solution)}+1/2N_{(2)(g)}\rightarrow(U,Pu)N_{(s)} \qquad (2)$$

30 During the nitriding process, molten tin may also evaporate and escape from the separation vessel.   30
Once nitriding is complete, the solid actinide nitrides are separated from the molten tin and the fission
products remaining in the tin solvent.
U.S. Patents No. 3,843,765 dated October 22, 1974 to Anderson et al, and 3,843,766 dated
October 22, 1974 to Anderson et al. are illustrative of such molten tin separation methods and
35 apparatus.   35
The above-cited patents disclose that the actinide nitrides are separated from fission product
impurities in the separation vessel by including mechanical means to physically move one from the
other. However, the necessary hardware to achieve this kind of physical separation is cumbersome to
incorporate in with a separation system operating at liquid tin temperatures. Additionally, the hardware
40 is very costly.   40

Summary of the invention

Accordingly, an object of the invention is to provide an apparatus and method for reprocessing
and separating spent nuclear fuels including actinide fuels and fission products, wherein actinide fuels
are separated from fission products at elevated temperatures.
45   Another object of the invention is to provide an apparatus and method for separating and   45
reprocessing spent nuclear fuels, wherein actinide fuels are separated from fission products without
the use of mechanical means to separate the two.
Yet another object of the invention is to provide an apparatus and method for separating and
reprocessing spent nuclear fuels, wherein a transport system to physically move the actinides from the
50 fission products is unnecessary.   50
Additional objects, advantages and novel features of the invention will be set forth in part in the
description which follows, and in part will become apparent to those skilled in the art upon examination
of the following, or may be learned by practice of the invention. The objects and advantages of the
invention may be realized and attained by means of the instrumentalities and combinations particularly
55 pointed out in the appended claims.   55
To achieve the foregoing and other objects, and in accordance with the purpose of the present
invention as embodied and broadly described herein, the apparatus for reprocessing and separating spent
nuclear fuels employs a molten metal solvent. A separation vessel is provided which includes a reaction
region to house the metal solvent, and a reflux region positioned above and adjacent to the reaction

region. The two regions are temperature-independent, capable of operating at different temperatures. Disposed within a separation vessel wall member, in a position adjacent to the reflux region, is an aperture. A porous filter member is disposed within a separation vessel wall member, defining the bottom of the separation vessel in supporting relationship to the metal solvent. Means for varying the
5   temperature of the separation vessel are included. Additionally, means for evacuating the separation   5
vessel and means for introducing various atmcspheres into the vessel are provided.

In a further aspect of the present invention, in accordance with its objects and purposes, the method for reprocessing and separating spent nuclear fuels including actinide fuels, volatile and non-volatile fission products, utilizes a molten metal solvent. A separation vessel is provided which includes
10   a reaction region housing the metal solvent, and a reflux region positioned above and adjacent to the   10
reaction region. The separation vessel further includes an aperture disposed within a vessel wall member in a position adjacent to the reflux region, and a porous filter member disposed within a vessel wall member defining the bottom of the vessel in supporting relationship to the metal solvent. Spent nuclear fuels are introduced into the separation vessel, followed by the introduction of a non-oxidizing
15   nitrogen-containing atmosphere. Solid actinide nitrides are formed within the metal solvent, while the   15
non-volatile fission products remain in solution. The reflux region is subsequently pressurized sufficiently to force the molten metal solvent through the porous filter, leaving the actinide nitrides behind the reaction region of the separation vessel.

Inclusion of the porous filter member enables the separation of the solidified actinide nitrides
20   from the molten metal solvent and the non-volatile fission products remaining in solution without the   ·20
need to provide a transport system to collect the solid actinide nitrides, and then physically transport them away from the molten solution. Additionally, substantial cost savings are realized by eliminating the expensive mechanical transport system. The separation of the present invention proceeds successfully at temperatures required to maintain the metal solvent in a molten state.

25   **Description of the drawing**   25
The accompanying drawing, which is incorporated and forms a part of the specification, illustrates an embodiment of the invention, and, together with the Description, serves to explain the principles of the invention.

Figure 1 is a schematic diagram of a spent nuclear fuel reprocessing and separation apparatus.

30   **Description**   30
Spent nuclear fuels, including actinide fuels in oxide, carbide or metal form, and volatile and non-volatile fission products are successfully reprocessed and separated utilizing a separation vessel housing a liquid metal solvent in a reaction region of the vessel, and a reflux region positioned above the reaction region. One embodiment of such a separation apparatus is illustrated in Figure 1.
35   Separation vessel 10 is formed from wall members 12 made of a carbon-containing material.   35
Preferably, the vessel walls 12 should: (1) be refractory and capable of containing the system at temperatures well above reaction temperatures; (2) inert to actinides at the reaction temperatures and solvent conditions employed; (3) possess low neutron absorption characteristics; and (4) be a source of carbon for the carbothermic reduction of actinide oxides. Exemplary carbon-containing materials
40   include carbides hydrocarbons and graphite. Graphite is the preferred material.   40
Separation vessel 10 defines various distinct regions: a reaction region 14 housing a molten metal solvent 16; a reflux region 18 positioned immediately adjacent to and above reaction region 14; and a condensation region 19 positioned adjacent to and immediately on top of reflux region 18. Condensation region 19 is a dome-like structure, removable from the remainder of vessel 10, defining
45   the top of the vessel. A reservoir 20 is formed within condensation region 19 at its intersection with   45
reflux region 18. Reservoir 20 is defined by a wall of insulation 22 and vessel wall member 12.

Establishment of different regions within vessel 10 permits the formation of a thermal gradient therein. Regions 14, 18 and 19 are all temperature independent. Generally reaction region 14 is maintained at a high temperature than regions 18 and 19. When reflux region 18 and condensation
50   region 19 are maintained at lower temperatures, evaporated metal from solvent 16 enters reflux region   50
18, condenses therein with the aid of a series of baffles 24, preferably formed of graphite, and returns to the solvent in reaction region 14. Regions 14 and 18 can be maintained at the same temperature, while condensation region 19 is maintained at a lower temperature. In this event, evaporated metal solvent continues through region 18 and into region 19. The evaporated solvent condenses therein and
55   is collected in reservoir 20 as a liquid.   55
For purposes of the present invention, the metal solvent should have good solubility for uranium and other actinide metals; possess a vapor pressure suitably low to allow condensation of the solvent in the reflux region; not readily form nitrides; and have a suitably low neutron cross section. Suitable solvents include lead, zinc, bismuth, and tin, as well as combinations thereof. The preferred solvent is
60   tin.   60
In order to effect the thermal gradient within vessel 10, a first induction heater 26, connected to a suitable power source, is disposed at the exterior of the vessel in surrounding relationship with reaction region 14. A second induction heater 28, also connected to a suitable power source, is disposed at the

exterior of the vessel in surrounding relationship with reflux region 18. Each induction heater is independent of the other, permitting the formation of the thermal gradient within vessel 10. Other heating and cooling means can be employed to establish the temperature gradient. Such means include, but are not limited to, radiation heating with resistance heaters, electron impact heating of the

5 vessel, and direct electrical resistance heating applied to the vessel. A plurality of cooling coils 30 are   5
disposed at the exterior of condensation section 19 in surrounding relationship thereto. A circulating medium such as water, steam, air, helium and the like flows through coils 30 to cool the condensation section of vessel 10.

Separation vessel 10, induction heaters 26, 28, and cooling coils 30 are all housed within a metal

10 vacuum tank 32. Tank 32 can alternatively be an inert gas chamber. The tank serves as a secondary   10
containment vessel for the spent nuclear fuels and provides additional safety in the event of a spill. Top lid 34 of tank 32 is removable, providing a means for introducing spent fuel into vessel 10. To this end, a flange 36 joining the vessel wall members which define reflux region 18 and condensation region 19, is removed from condensation region 19, e.g., the dome is separated from the remaining sections of

15 vessel 10. Spent fuel is then added mechanically or in batch-style to the vessel. A removable graphite   15
liner 38 may optionally be included and disposed within the interior of vessel 10. Before each new batch of spent fuel is reprocessed, a new graphite liner is placed within vessel 10. This reduces wear on the interior walls of vessel 10.

A conduit 40 pierces bottom wall 42 of tank 32 and wall member 12 of vessel 10. A valve 43 is

20 disposed within conduit 40. Conduit 40 is in communication with pump and vacuum systems (not   20
shown), various collection vessels (not shown) to collect and hold CO, volatile fission products, etc. removed from vessel 10, and also a containment vessel (not shown) housing a non-oxidizing nitrogen-containing gas such as N_2. Electrical leads for induction heaters 26 and 28, piping for coils 30, and other required auxiliaries are also provided through bottom wall 42.

25 Disposed within separation vessel wall member 12 in a position defining the bottom of vessel 10,   25
and specifically the bottom of reaction region 14, is a porous filter member 46 which is in a supporting relationship with metal solvent 16. Filter 46 can vary in size, and may extend to the full width of the bottom of vessel 10. The porosity of filter 46 must be sufficient to provide support for molten metal solvent 16 when a pressure of about 1 atm is applied to reflux region 10 and condensation region 19,

30 but yet permit the flow of the solvent and any compositions soluble therein when a pressure of about   30
1.1 to 1.2 atm is applied to the reflux region. Preferably, the pore size of the filter material should be about 10 to 100 microns.

If graphite liner 38 is included in separation vessel 10, the bottom of liner 38 is of a porous nature similar to filter 46. Preferred filter materials include graphite, stainless steel, and other materials which

35 may be used to form separation vessel 10. The preferred material is graphite. When a pressure of about   35
1.1 to 1.2 atm of the non-oxidizing nitrogen-containing gas, or an inert gas such as argon, is applied to separation vessel 10 above metal solvent 16, the solvent is caused to flow from the vessel and into a recycle chamber 48 positioned under vessel 10. Recycle chamber 48 is cooled by the circulation of a cooling medium such as water through its chamber walls. Metal solvent 16 as well as the non-volatile

40 fission products are collected in recycle chamber 48, cooled, and thereafter removed therefrom. A   40
drainage tube 49 is removably attached to separation vessel 10, and also removably attached to recycle chamber 48. Inclusion of drainage tube 49 is necessary to separate the cooler recycle chamber from the much hotter separation vessel.

Metal solvent 16, as well as reaction region 14, are maintained at a temperature sufficient to

45 solubilize the actinide fuels, e.g., the reaction region temperature. Preferably, the reaction region   45
temperature is about 1450 to 1800°C. More preferably, the temperature is from 1550 to 1700°C, and most preferably it is about 1630°C. Reflux region 18 is initially maintained at a lower temperature than the reaction region e.g., the reflux temperature. This reflux temperature must be low enough to effect the refluxing of the metal solvent. When tin is employed as the solvent, a temperature of about 800 to

50 1200°C is maintained. Preferably, the temperature range is about 950 to 1050°C. The temperature of   50
condensation region 19 is lower than reflux region 18, but above the melting point of the metal solvent. A temperature range of about 232 to about 1050°C is suitable. At these lower temperatures, residual metal solvent or volatile fission products that pass through reflux region 18 are trapped and collected.

55 The rate of carbothermic reduction of actinide oxides is dependent on the amount of available   55
carbon within the metal solvent of choice. Carbon is only very slightly soluble in the metals of choice, especially in tin. However, with the addition of the first catalyst to the metal to increase carbon solubility and permeability, the carbothermic reduction rate is greatly increased. To be effective, the catalyst must increase carbon solubility in the metal and not readily form stable carbides. Suitable

60 catalysts include cobalt, nickel, iron, and combinations thereof. The preferred catalyst is cobalt. These   60
elements are also particularly useful because they are relatively non-volatile and do not evaporate from the molten tin solvent. For example, tin is more than one thousand times as volatile as cobalt. The catalyst is present in an amount wherein the weight percent of catalyst in molten metal solvent is in the range of from about 0.1 to 20. Preferably, the percentage is from about 1 to 20. Most preferably,

65 the percentage is about 5 to 10.   65

Prior to introducing the spent nuclear fuel into vessel 10, it may be necessary to declad the fuel, either mechanically or chemically. The spent fuel is broken up to relatively small pieces (3 mm or less) and then introduced into vessel 10 with a mechanical feeder or in batch style.

Once the spent nuclear fuels have been added to solvent 16 at the appropriate temperature,
5 carbon from graphite wall 12, dispenser 52 and liner 38 reduces the actinide oxides, evolving CO. As the spent fuels dissolve in the metal solvent, volatile fission products are released, while non-volatile fission products remain in solution. At the reaction region temperature, a significant amount of solvent from molten solvent 16 in reaction region 14 is evaporated and enters reflux region 18 along with the volatile fission products and CO. Within reflux region 18 the evaporated solvent is caused to condense,
10 due to the lower temperature, and is returned to reaction region 14. Very little of the evaporated solvent reaches condensation region 19.

Volatile fission products, as defined herein, include those fission products listed in Table I which have very high, moderate, or low volatilities. Volatilities of the volatile fission products within reflux region 18 vary tremendously. At the reflux region temperature the less volatile fission products
15 condense and return to molten tin solvent 16. These volatile fission products are defined as condensable fission products. The more volatile fission products do not condense in reflux region 18 or condensation region 19. Instead, they flow from the vessel through conduit 40 and are separately collected in a collection vessel. By definition herein, the more volatile fission products are called non-condensable fission products.
20 Table I lists the estimated volatilities of fission products at about 1627°C.

Table I
Volatilities of fission products

A.	Very high (10^{-3} to 1 atm)	
	I, Br, Cd, Cs, Rb, and Se	
B.	Moderate (10^{-6} to 10^{-4} atm)	
	Sb and Te	
C.	Low (10^{-8} to 10^{-6} atm)	
	Ba, Eu, Sm, and Sr	
D.	Very low (less than 10^{-9} atm)	
	Ce, Gd, La, Mo, Nd, Pd, Pm, Pr, Rh, Ru, Tc, Y, and Zr	

During the removal of CO and the non-condensable volatile fission products, a sparging gas is introduced into vessel 10 through conduit 50 to accelerate carbothermic reduction. An inert gas such as argon is suitable for this purpose. After the non-condensable volatile fission products have been removed from vessel 10, the temperature of reflux region 18 is increased from its reflux temperature,
35 e.g., 950 to 1050°C, to the temperature of the reaction region. However, condensation region 19 is maintained by cooling as necessary at a temperature within the range of 232 to 1050°C. At the elevated reaction region temperature, the condensable volatile fission products will generally not condense in reflux region 18. Essentially, the condensable volatile fission products, as well as some evaporated tin, are distilled, condensed and collected in condensation region 19 at reservoir 20.
40 Although some tin is lost in this distillation process, far less is lost than if the removal of all volatile fission products occurred without the existence of reflux and condensation regions which establish the temperature gradient. Additionally, the temperature gradient permits selective separation of the volatile fission products based on their different volatilities, e.g., condensable and non-condensable volatile fission products are separated.
45 When distillation is complete, the temperature of reflux region 18 is decreased back to the defined reflux temperature of about 950 to 1050°C. Thereafter, a non-oxidizing nitrogen containing atmosphere is introduced into vessel 10. An example of such an atmosphere is pure nitrogen. The nitrogen atmosphere enters metal solvent 16 and is dispersed therein with the aid of dispenser 52. Additionally, the nitrogen containing atmosphere may also be introduced through conduit 40. A
50 nitrogen pressure of about 1 atm is preferred. Within solvent 16, solid actinide nitrides including UN, U_2N_3, PuN, Pu_2N_3, ThN and the like, are formed. The original soluble fission products remain in solution.

To separate the solid actinide nitrides from the molten solvent and soluble fission products, reflux and condensation regions 18 and 19 are pressured with a gas such as nitrogen, and introduced through conduit 40 at a pressure of about 1.1 to 1.2 atm. The pressurizing effect forces the molten
55 solvent and soluble fission products to flow from reaction region 14, through filter 46 and into recycle chamber 48. Vessel 10 is then cooled and dismantled e.g., a separation of the vessel at flange 36 occurs. The remaining solid actinide nitrides are taken from vessel 10 and processed to yield usable fuels, e.g., ignited in the presence of oxygen to produce actinide oxides.

To accelerate the nitriding process, a second catalyst is included. The second catalyst chosen
60 must increase the affinity of nitrogen in the metal solvent, but not form stable nitrides. Suitable catalysts include magnesium, calcium, strontium, barium, aluminum, manganese, vanadium,

chromium, and mixtures thereof. The preferred second catalyst is calcium. Depending on the choice of second catalyst, it may be added to the metal solvent initially as part of the solvent, or just prior to the nitriding process. This is due to varying volatilities of the catalysts and the potential loss of the more volatile second catalysts during the carbothermic reduction process. The weight percentage of second
5   catalyst to metal solvent is about 1.0 to 10.0. Preferably, the weight percentage is about 1.8 to 2.5.   5
Most preferably, it is about 1.0. By including the second catalyst, an approximate threefold rate increase in nitriding is observed.
        The following examples are illustrative of the process and apparatus of the present invention, and are not to be regarded as limiting its scope.

10   **Example 1**   10
        50 g of reagent grade shot tin, 10 g of UO_2 (depleted) and 1 g of cobalt are added to the separation vessel shown in Figure 1. Cobalt is added as a catalyst otherwise the carbothermic reduction will take about 20—30 hours. The reaction region of the vessel and the tin are then heated and maintained at a temperature of about 1600°C, creating a molten tin solvent. The reflux region of
15   the vessel is maintained at a temperature of about 1000°C. 10 grams of UO_2 (depleted) are added to   15
the molten tin solvent. Argon is then bubbled through the molten tin as a sparging gas for two hours: Non-condensable volatile fission products are collected in suitable cold traps, CO is oxidized to CO_2 and trapped in an alkaline matrix such as the commercial product Ascarite. The temperature of the reflux region is then increased to about 1600°C. Evaporated tin and the previously condensable volatile
20   fission products are collected in a collection vessel. After about 15 minutes, the temperature of the   20
reflux region is decreased to about 1000°C. N_2 (at 1 atm) is introduced into the vessel through the porous filter, forming actinide nitrides. After about one hour, the flow of N_2 through the filter is discontinued while N_2 is introduced into the reflux region at about 1.2 atm. The molten tin and the non-volatile fission products in the molten tin flow out of the vessel through the porous filter and collected
25   in a recycle chamber, leaving the solid actinide nitrides behind.   25

Example 2
        10 g of ThO_2 is separated from other spent nuclear fuels under the same conditions as in Example 1.

Example 3
30   10 g of $(U,Pu)O_2$, i.e., a mixture of UO_2 and PuO_2, is separated from other spent nuclear fuels   30
under the same conditions as in Example 1.
        The foregoing description of a preferred embodiment of the invention has been presented for purposes of illustration and description. It is intended that the scope of the invention be defined by the claims appended hereto.

35   **Claims**   35
        1. An apparatus for reprocessing and separating spent nuclear fuels including actinide fuels and fission products, in a molten metal solvent, comprising:
        a vessel having a reaction region housing said molten metal solvent, and a reflux region positioned above said reaction region, said reflux region being capable of operating at a different temperature than
40   said reaction region, said vessel including an aperture disposed within a wall member of said vessel in   40
a position adjacent said reflux region;
        means for heating said vessel;
        means for evacuating said vessel; and
        means for introducing a non-oxidizing nitrogen containing atmosphere into said vessel, to form
45   actinide nitrides within said molten metal solvent.   45
        2. The apparatus according to Claim 1 wherein said vessel further comprises a condensation region positioned above and adjacent said reflux region, said condensation region being capable of operating at a different temperature than said reflux and reaction regions.

        3. The apparatus according to Claim 1 further comprising a liner disposed within said vessel.
50   4. The apparatus according to Claim 1 wherein said vessel is substantially formed of graphite.   50

        5. The apparatus according to Claim 1, wherein a porous filter member is disposed within a separation vessel wall member defining the bottom of said separation vessel in supporting relationship to said metal solvent.
        6. The apparatus according to Claim 1, wherein said filter member has a pore size of about 10 to
55   100 microns.   55
        7. A method for reprocessing and separating spent nuclear fuels including actinide fuels, volatile and non-volatile fission products, comprising:
        providing a separation vessel formed of a carbon-containing material, and a molten metal solvent disposed within said separation vessel, said separation vessel including an aperture disposed within a
60   vessel wall member;   60

adding a first catalyst to said metal solvent which increases the solubility of carbon in said solvent;

introducing spent nuclear fuels into said separation vessel; and

forming CO within said molten metal solvent from actinide fuels which are oxides.

8. The method according to Claim 7, further comprising removing said volatile fission products and CO from said reaction vessel.

9. The method according to Claim 8, further comprising:

introducing a non-oxidizing nitrogen-containing atmosphere into said separation vessel after said volatile fission products and CO have been removed therefrom;

forming solid nitrides from said actinide fuels within said molten metal solvent; and

separating said solid actinide nitrides from said molten metal solvent and non-volatile fission products.

10. A method for reprocessing and separating spent nuclear fuels including actinide fuels, volatile and non-volatile fission products, in a molten tin solvent, comprising:

providing a separation vessel formed of a carbon-containing material, said separation vessel defining a reaction region housing said molten tin solvent and a reflux region positioned adjacent to and above said reaction region, said separation vessel including an aperture disposed within a vessel wall member in a position adjacent said reflux region;

adding a first catalyst to said tin solvent which increases the solubility of carbon in said solvent;

maintaining said reaction region at a temperature sufficient to convert said actinide fuels to a solubilized state;

maintaining said reflux region at a temperature sufficient to reflux said tin solvent, said reflux temperature being lower than said reaction region temperature;

introducing spent nuclear fuels into said tin solvent;

forming CO within said molten tin solvent from actinide fuels which are oxides;

allowing CO and volatile fission products within said molten tin solvent, along with some molten tin, to evaporate and flow into said reflux region;

removing CO and volatile fission products which are not condensable at said reflux temperature from said reflux region through said vessel aperture, leaving evaporated tin and less volatile fission products in said reflux region; and

allowing evaporated tin and less volatile fission products disposed within said reflux region which are condensable at said reflux temperature to cool sufficiently and return to said molten tin solvent in said reaction region.

11. The method according to Claim 7, wherein the temperature of the reaction vessel is varied; the said separation vessel is evacuated and various atmospheres are introduced into said separation vessel and said separation vessel contents are filtered through a porous filter disposed within a separation vessel wall member.

12. The method according to Claim 11, wherein said filter has a pore size of about 10 to 100 microns.