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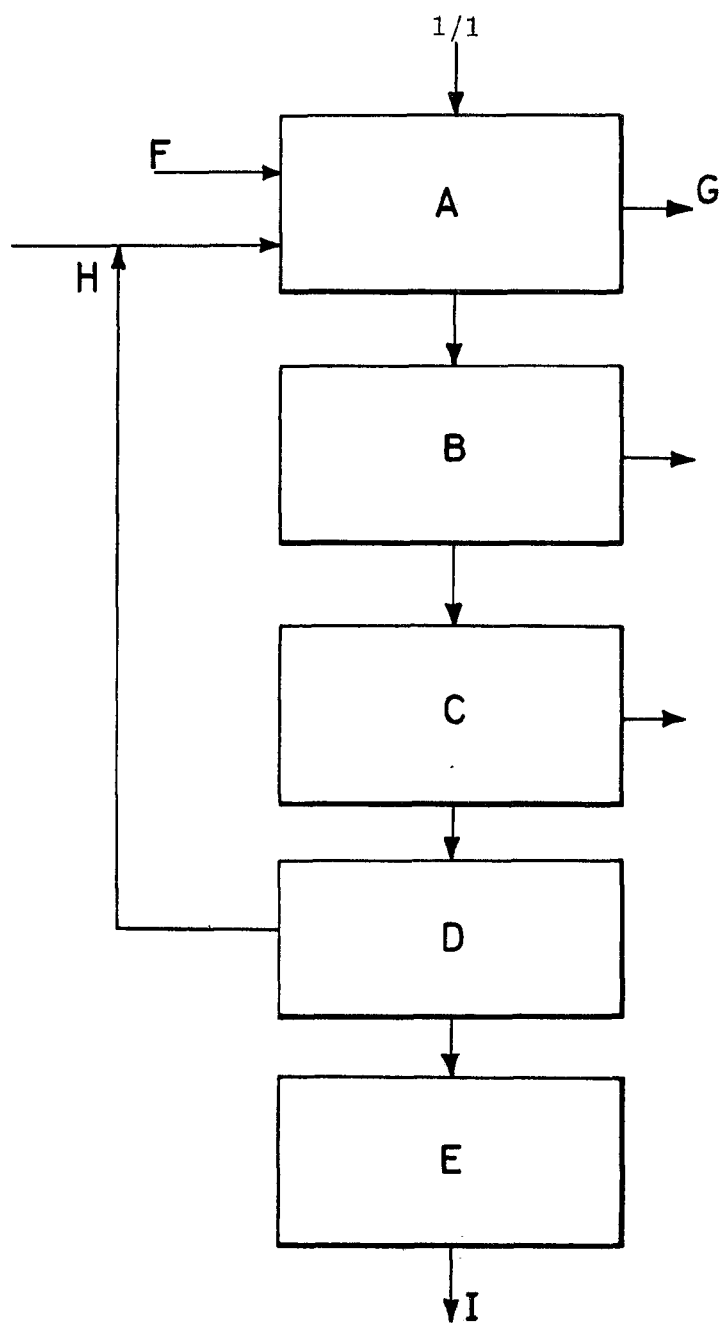
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(54) **Electrochemical reprocessing
of nuclear fuels**

(57) A method for the reprocessing of irradiated nuclear fuel which is particularly suitable for use with fuel from fast reactors and has the advantage of being a dry process in which there is no danger of radiation damage to a solvent medium as in a wet process, comprises the steps of dissolving the fuel in a salt melt under such conditions that uranium and plutonium therein are converted to sulphate form. The plutonium sulphate may then be thermally decomposed to PuO₂ and removed. The salt melt is then subjected to electrolysis conditions to achieve cathodic deposition of UO₂ (and possibly PuO₂). The salt melt can then be recycled or conditioned for final disposal.

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SPECIFICATION

Electrochemical process for reprocessing irradiated nuclear fuels

- 5 This invention relates to an electrochemical process for the reprocessing of irradiated nuclear fuels and in particular, but not exclusively, nuclear fuels from fast nuclear reactors. 5
- The reprocessing of irradiated fuels from fast nuclear reactors, hereinafter termed fast fuel, poses a number of characteristic problems which are not normally encountered with irradiated nuclear fuels obtained from other types of nuclear reactor. Thus, if wet processing is carried out, 10 the high residual activity of the fuel should not impair, as a result of radiolysis, the stability of the organic phase. Hence comparatively long cooling times (6 to 9 months) are required to enable the activity of the fuel to drop. 10
- When dry reprocessing is carried out, reactants are employed which are insensitive to irradiation and hence the cooling time can be considerably reduced (1 month). This reduction in 15 the time fast fuel which has a high residual activity has to be stored before reprocessing is also of high economic value because it enables costly plutonium to be re-used with minimised delay. 15
- Since fast reactors are likely to come to be of increasing importance, the burn-up ratings of fast fuels and the number of recycling operations to be carried out thereon will in the future become of increasing importance and need to be improved as is far as possible. For example, 20 until such time as plutonium is recovered from fuels having a short exposure time, the alpha activity alone of Pu_{239} makes it imperative to employ, in the subsequent reprocessing, alpha-proof glove boxes without any need for gamma- and neutron opaque shields. At higher radiation levels, however, considerable amounts of plutonium isotopes are formed due to subsequent reactions (n, γ) as well as not inconsiderable amounts of neptunium and transplutonic 25 elements. Under those circumstances it will become necessary to resort to remote- or semi-remote handling equipment and it shall not be feasible even to consider achieving a complete separation of plutonium and uranium from the fission products, such as is required with normal fast fuels. 25
- Thus, in general the dry procedures for processing fast fuel will not be impaired by the 30 problem of inability to achieve high decontamination factors, which problem has generally militated against development of the dry procedure. 30
- Those fast fuels which undergo a high level of combustion (high burn-up fast fuels) yield a large amount (10%) of fission products. Tritium is a particularly important fission product since it is produced in an amount 2 to 3 times greater with fuel from a fast reactor than with fuel 35 from a thermal reactor for the same degree of burn-up. The presence of considerable amounts of tritium would be a particular problem with wet processes when it would be necessary, in order to prevent its irreversible dilution in water, for constant passage of water to occur. 35
- In the dry processes, conversely, gaseous fission products (iodine, xenon, krypton, tritium) are released completely in a concentrated form without undergoing dilution by other gases and 40 liquids. 40
- Fuel from a fast reactor retains a specific residual activity which, for the same cooling time, is about 10 times greater than that of fuel from a thermal reactor. In the wet process, it is thus vital, if cooling times reduced to the order of a few months are to be achieved, to reprocess the fuel and the fertile material of the cladding of the reactor, in order to dilute the residual heat of 45 the former. In this case, it is necessary to remove about 3 times the amount of heat as is generated by thermal-reactor fuel. In the case of the dry processes, in which the reactants which are used are thermally stable, this necessity no longer arises and it is possible to process the core material and the cladding material separately, thus avoiding, in the head-end procedure, 50 the additional step of regrouping the dismantling rods prior to cutting. 45
- According to the present invention, there is provided an electrochemical method for the reprocessing of irradiated nuclear fuel, which comprises the steps of: 50
- (a) dissolving the fuel in a salt melt under conditions such that uranium and plutonium therein are converted to sulphate form;
 - (b) subjecting the salt melt to electrolysis conditions to effect electrodeposition of uranium 55 and of plutonium present in solution the salt melt at the time of the electrodeposition; and
 - (c) optionally thermally decomposing plutonium sulphate present in the salt melt prior to said electrodeposition and removing the plutonium oxide thereby formed from the salt melt. 55
- The electrochemical process of the present invention, which, in its preferred operating mode in which occurs thermal decomposition of PuSO_4 in the melt to form PuO_2 which may be 60 separated off, may be termed a pyroelectrochemical process, is a development of previously devised electrochemical processes involving the physical degradation and dissolution in nitrate 60 melts of fuel from fast reactors and which usually consist of mixed oxides or carbides of uranium and plutonium, which carbides may additionally comprise thorium carbide, as described for example in Italian Patents Nos. 959,654 and 951,873 owned by AGIP NUCLEARE S.p.A., 65 followed by the separation of plutonium by thermal decomposition. 65

In the procedure of Italian Patent No. 951,873 fused salts or solutions containing active fission products which cannot be re-used and need to be conditioned are supplemented by addition of silica and titanium dioxide and water which may be present is then evaporated off.

The mixture is fired at 800°C to 850°C, melted at 900 to 1200°C and the molten mass is rapidly cooled to yield a glassy solid which is both infrangible and insoluble. 5

As with other dry procedures, the electro-chemical method of this invention makes it possible to process the irradiated fuels after a short decay, since the problem of thermal and radiolytic degradation of solvents does not arise and it is not necessary to effect the complete extraction of the fission gases in a concentrated form. In addition, the electrochemical process enables 10 comparatively high decontamination factors to be attained, even though these cannot be compared with those which can be obtained with wet aqueous procedures. 10

Due to the overall simplicity of the method of this invention in comparison with the wet methods, the present method can be carried out, moreover, in a small compact facility integrated with a reactor or a group of reactors. Hence, there is no need to transport highly 15 active fuels to recovery plant. 15

The electrochemical method of this invention requires the use of a salt melt and conditions under which uranium and plutonium will be converted to sulphate form. Hence it is preferred to use an eutectic mixture of alkali metal sulphates and effect the dissolution preferably with the assistance of absolute H₂SO₄. Typical eutectic mixtures of salts which may be used are the 20 following: 20

	EUTECTIC MIXTURES OF SULPHATE	MELTING POINT	
25	Li ₂ SO ₄ -K ₂ SO ₄ -Na ₂ SO ₄ (78-8.5-13.5 molar %)	512°C	25
	Li ₂ SO ₄ -K ₂ SO ₄ (80-20 molar %)	535°C	

Various steps of the method of this invention are summarised in the block diagram of the accompanying drawing, which steps are indicated by a series of reference letters whose 30 significance will now be summarised. 30

A UO₂-PuO₂ fuel is dissolved, at A in a molten sulphate bath which is preferably at 550°C to 600°C under the action of H₂SO₄ (absolute) supplied through line F. Fission products in the gaseous state and ruthenium are discharged at G.

The second stage (B) involves the thermal decomposition of plutonium sulphate (Pu(SO₄)₂) to 35 PuO₂ and is carried out by raising the temperature (preferably up to 750°C to 800°C) of the plutonium sulphate. Electrodeposition of uranium and any plutonium which is present takes place at C. 35

The molten salts at (D) are recycled to the starting end of the method at H. Molten salts which 40 contain a high concentration of fission products are conditioned at E, for example by encasing them in a metallic matrix, whereafter the fission products are forwarded to final storage at I. 40

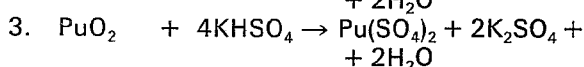
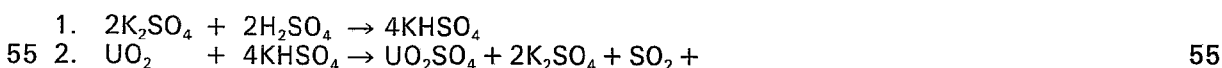
The individual steps will now be discussed in greater detail.

A. DISSOLUTION OF NUCLEAR FUEL

The fuel is preferably dissolved in a molten sulphate bath by dropping of absolute sulphuric 45 acid (100%) on the bath to which the fuel is added, the bath being kept at a temperature of 500°C to 600°C. 45

When sintered oxides UO₂-PuO₂ are immersed in the molten sulphates, they do not disaggregate but, rather, enter directly into solution as a result of attack by bisulphates which 50 are formed by reaction between the sulphates of the bath and the added sulphuric acid. 50

For example, for potassium:



The sintered carbides UC-PuC, in contrast, undergo rapid disaggregation when they are 60 immersed in the fused-salt bath. 60

Alkali metal bisulphates can be employed directly, although this is not advisable because of their effect on the proportion by weight of components present in the bath.

Inasmuch as the dissolution is not selective with respect to the cladding material from a 65 nuclear fuel reactor, the removal of the cladding from the fuel elements when these are formed 65

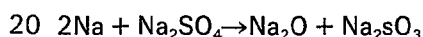
of oxides, by attack with liquid metals enables the mechanical cutting of the cladding of the electrodes to be dispensed with. Dissolution of the cladding takes place if the fuel element is immersed in zinc or an alloy, such as Cu-Sb, at a temperature of 800°C to 950°C. The dissolution ratings which may be attained by stainless steel may be varied from 10% to 40% by weight.

The fuel which was left undissolved at the bottom of the melt crucible is separated off from the liquid metal solvent by filtration.

In the case of the carbides, the claddings can easily be separated from the fuel. For example the fuel-containing claddings may be immersed in a fused-salt bath in a metal basket. The stainless steel claddings are not attacked whereas the fuel proper, which is in powder form, are removed from the claddings and deposited in the melt vessel. Once the fuel is completely separated from the cladding, the basket with the empty cladding shells is withdrawn from the molten bath.

The oxides are of substantially massive form and can be broken up in the molten sulphate bath by the action of appropriate oxidizing agents such as oxygen, air and molten nitrates.

The presence of residual elemental sodium on the claddings is not a source of difficulty since sodium dissolves in the molten sulphates without bursting into flame and Na₂O with Na₂SO₃ are formed; these are subsequently converted into sulphates by the action of the sulphuric acid:



High concentrations of oxides of up to 50% by weight can be obtained in the molten salts.

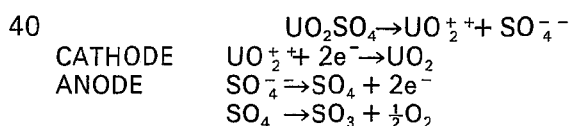
B. THERMAL DECOMPOSITION OF PLUTONIUM

By raising the temperature of the molten bath to 750°C to 800°C, thermally unstable plutonium sulphate Pu(SO₄)₂, is decomposed with the attendant formation of uranium-free, black, crystalline PuO₂ which collects on the bottom of the melt vessel; in contrast uranyl sulfate UO₂SO₄ remains unaffected.

The recovery of plutonium to an extent of up to 98% has been found to be possible after a residence time of the fuel in the molten bath at a temperature between 800°C and 760°C for a time of from 90 to 120 hours.

C. ELECTRODEPOSITION OF URANIUM

When a pair of electrodes are immersed in the molten sulphate bath containing UO₂SO₄ from the fuel being reprocessed, the bath being at a temperature of 550°C to 600°C and the electrodes being maintained at a potential of 0.5 to 1.0 volts, an electrolytic cell is formed on the cathode of which UO₂ is deposited whilst sulphur trioxide and oxygen are evolved at the anode. The half cell reactions which occur are as follows:



Metallic uranium cannot be produced with this type of electrolysis since this would require an oxygen-free source of uranium ions and a higher potential.

The electrodeposited UO₂ is crystalline, compact and adheres to the cathode. The O/U atomic ratio is exactly stoichiometric.

In a set of tests which have been carried out on irradiated UO₂ supplemented by additional UO₂ which contained a synthetic mixture of stable isotopes so as to simulate a 5% burn-up, the behaviour of a few fission products (cerium, caesium, zirconium, niobium, ruthenium) was determined by gamma-spectrometry analysis, and that of certain other elements (strontium, molybdenum, samarium) was determined by atomic absorption analysis.

The following decontamination factors were obtained for the electrodeposited UO₂; a decontamination factor denotes the ratio of initial to final contamination of an element in UO₂:

ELEMENT OF NUCLIDE	DECONTAMINATION FACTOR	
5 $^{144}_{\text{Ce}}$	∞ (*)	5
$^{137}_{\text{Cs}}$	∞	
$^{95}_{\text{Zr}}-^{95}_{\text{Nb}}$	0	
$^{103}_{\text{Ru}}$	50	
Sr	1,000	
10 Mo	500	10
Sm	100	

(*) The symbol ∞ means that no activity of the electrodeposited UO_2 could be detected.

15 The overall decontamination factor as determined on the irradiated UO_2 was of the order of 30 to 50. 15

The uranium remaining in solution in the melt after the electrolysis stage has been found to amount to less than 15 parts per million (ppm) of U.

20 ELECTRODEPOSITION OF URANIUM-PLUTONIUM 20

By dispensing with the plutonium thermal decomposition stage and carrying out electrodeposition of a melt containing both Pu and U in solution in the melt as sulphates, it is possible electrolytically to deposit on the cathode a solid solution of $\text{UO}_2\text{-PuO}_2$.

The amount of plutonium remaining in solution in the melt following electrolysis in this case amounts to less than 30 ppm of Pu. 25

D. RECYCLING THE MELTED SALTS

Possibly after subjecting them to a decontamination treatment to precipitate fission products from solution therein using basic agents such as oxides, carbonates or hydroxides, the molten salts can be recycled to the starting point in the overall process. 30

The only element which cannot be removed from the molten salts is caesium, which will accumulate in the molten salts and make it necessary periodically to dispose of the salts in a conditioning operation and effect partial renewal with fresh salts.

35 E. CONDITIONING OF THE MOLTEN SALTS CONTAINING FISSION PRODUCTS 35

The molten salts which contain fission products are conditioned by embedding them in a metallic matrix. This is a procedure which is already known for fired materials and glasses, and comprises the following steps:

(a) reduction of the molten sulphates to a solid granular form by dropping onto a cold surface, 40

(b) dispersion of those particles obtained which have a diameter of 3 to 5 mm in a metallic matrix by casting a liquid alloy onto the granular material which is disposed in a preferably cylindrical vessel at the time.

The granules are distributed evenly in the metallic matrix which fills the voids between them and, in practice, occupies about 30% of the overall volume of the casting obtained. 45

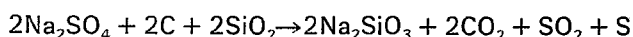
The thermal conductivity of the final product (10 to $30 \text{ W.m}^{-1}.\text{°C}^{-1}$) exceeds by a factor of 15 to 30 that of glasses. Hence it is possible to maintain a maximum temperature of 450°C to 500°C at the centre of a cylindrical block having a diameter of 50 cm and a height of 1.5 m and a power density of 100 W litre^{-1} . This maximum temperature is lower than the melting point of the alkali metal sulphate eutectic. 50

Two kinds of alloys can, for example, be used for this purpose, viz.:

(1) Lead alloys, melting in the range 250°C to 320°C

(2) Zinc alloys, melting in the range 400°C to 500°C .

As an alternative to using alloys, the molten sulphates can be vitrified to silico-titanates, by subjecting them to decomposition at temperatures higher than 800°C after adding coal and silica thereto and subsequently effecting melting at 1000°C to 1200°C after the addition of titanium dioxide. With, for example, sodium sulphate, the following reactions take place: 55



60 $2\text{Na}_2\text{SiO}_3 + 2\text{TiO}_2 + 2\text{SiO}_2 \rightarrow 2(\text{Na}_2\text{O} \cdot 2\text{SiO}_2 \cdot \text{TiO}_2)$ 60

The silico-titanate glasses, while containing a high amount of alkali metals (sodium, potassium and lithium), have dissolution ratings in water which can be compared to those of phosphate and borosilicate glasses.

Prior to being reprocessed, the electrolytically deposited oxides must be subjected to the following operations: 65

- (a) mechanical removal from the electrode
 (b) milling
 (c) washing with water, to remove the adhering salt from the melt
 (d) classification
 5 (e) admixture with further oxide to restore the original composition. 5
- The process of the present invention can be carried out so as to ensure an overall decontamination factor which is never below 10 and which can be improved by carrying out a multi-step electrolysis in which the UO_2 -coated cathode of a cell acts as the soluble anode in a following cell by virtue of polarity inversion.
- 10 While the decontamination factors which can be attained cannot be compared with those 10 obtained in wet processes, as outlined above, the isotopic composition of the fast fuel as such will require the adoption of remote-handling manufacturing procedures which will make the complete separation of the fission products from plutonium or from uranium unnecessary.
- The volume of the final waste materials which are composed of solidified sulphates and fission
 15 products embedded in metallic matrices or of silico-titanate glasses incorporating the fission 15 products is about 1.6 to 1.8 times higher than the volume of the vitrified wastes obtained in wet procedures, assuming that there are performed:
- (a) 5 recyclings of the molten sulphates in the treatment of fuel material from reactor cores
 (UO_2 -20% PuO_2), and
 20 (b) 100 recyclings of the molten sulphates obtained in the treatment of the fertile material of 20 the reactor cladding (UO_2 2% PuO_2).
- According to updated estimates (1977) a wet processing plant, to be commercially competi-
 tive, should have a potential output of reprocessed fuel of some 1500 tonnes a year, which is
 equivalent to the amount dumped yearly by about 50 nuclear power stations of 1,000 MW_e.
 25 each. 25
- The dry methods, in contrast, permit the unit costs of the reprocessed fuel to be substantially
 of the same order of magnitude as with reprocessed fuel obtained by wet methods by virtue of
 the reduced number of processing stages and greater compactness of the apparatus resulting
 from the fact that small closed loop plant integrated with the reactor or a pool of reactors can be
 30 adopted. 30
- The potential advantages, in general, of an integrated plant in which reprocessing of fuel is by
 a dry method, are the lesser initial investment cost, the absence of need to transport the
 irradiated fuel with the attendant improvement in public safety, and the reduction in cooling
 times and thus poor utilisation of a costly asset, namely the plutonium.
- 35 The advantages, more particularly, of a pyroelectrochemical process are: 35
- (a) Fairly high decontamination factors for plutonium and uranium are obtained thereby
 reducing the parasitic capture of neutrons to acceptable levels;
 (b) The quantitative recovery of plutonium and uranium in the form of oxides, PuO and UO_2 ,
 either separately or admixed, which oxides are of reactor-grade and are ready for reprocessing
 40 into fuel elements, is made possible; 40
- (c) The suppression of the loss of plutonium which occurs in the aqueous procedures due to
 its incomplete dissolution and which may reach 3% by weight.
- The efficient solubilizing effect of molten sulphates and sulphuric acid ensures total dissolution
 of the oxides.
- 45 The following examples illustrate this invention: 45
- EXAMPLE 1 (Dissolution of UO_2)
- A sintered UO_2 -pellet weighing 20.0 g was immersed in 100 g of a ternary eutectic mixture
 Li_2SO_4 - K_2SO_4 - Na_2SO_4 (mol %: 78.0-8.5-13.5, respectively) which was maintained in the
 50 liquid state at a temperature of 575°C. 50
- 18.0 g of 100% H_2SO_4 obtained by admixing 98% sulphuric acid with fuming sulphuric acid
 (oleum) and containing 20% by weight of SO_3 were added cautiously in dropwise fashion to the
 melt.
- After a period of 5 hours, the pellet had completely dissolved in the salt melt.
 55 55
- EXAMPLE 2 (Dissolution and separation of Pu/U by thermal decomposition to PuO_2 of Pu)
- A sintered UO_2 -18% PuO_2 -pellet weighing 1.6 g was dissolved in a molten bath constituted
 by 100 g of a ternary eutectic mixture Li_2SO_4 - K_2SO_4 - Na_2SO_4 to which 5.0 g of 100%- H_2SO_4
 produced as in Example 1, were added, the bath having a temperature of 575°C. The
 60 temperature of the fused salt bath which contained 24.8 mg of Pu per g of salt, was 60
 subsequently raised to 800°C and maintained at this temperature for 50 hours, before being
 reduced to 760°C at which it was kept for 140 hours.
- Under these conditions, the plutonium sulphate, $\text{Pu}(\text{SO}_4)_2$, which was unstable, was decom-
 posed to PuO_2 which slowly and gradually deposited at the base of the melt vessel.
- 65 On completion of the decomposition, only 0.05 mg of Pu per g of salt remained in solution 65

together with all the uranium which was initially present. This means that 98% by weight of the initial plutonium had been precipitated free from uranium.

EXAMPLE 3 (Electrodeposition of UO_2)

5 A sintered UO_2 -pellet weighing 1.8 g was irradiated for one hour with a flux of 5
63.10¹²n/cm²/sec and allowed to undergo decay for about 2 months. This pellet together with
0.2 g of "fissium" (non-irradiated UO_2 containing, however, a synthetic mixture of stable
isotopes to simulate a 5% burnup) was dissolved in 100 g of a melt consisting of a eutectic
mixture (ternary) of Li_2SO_4 - K_2SO_4 - Na_2SO_4 to which was added 100% H_2SO_4 as in Example 1.
10 The melt was maintained at a temperature of 575°C. 10

The fused salts were subjected to gamma-counting to determine the initial activity of the
individual fission products which were present. Two electrodes of platinum wire of a diameter of
1.0 mm connected with a DC source were immersed in the fused salt bath. A potential
difference of 1.3 V was applied between the two electrodes, this potential difference correspond-
15 ing to the current flow of 10 milliamperes. 15

After about 25 hours, the electrolysis was complete as was indicated shown by the absence of
uranium in solution in the melted salts. The salts and the UO_2 deposited on the cathode were
subjected to gamma-counting again, the results being as follows:

Element	Initial activity in salt (disint./minute)	Final activity in salt (disint./minute)	% Element in salt at end of electrolysis	% Activity in the electro- deposited UO_2 (disint/ minute)	% Element in UO_2 at end of electrolysis
144 _{Cerium}	4418.0	4519.0	100	0	0
137 _{Cerium}	475.4	490.0	100	0	0
101 _{Ruthenium}	199.6	12.0	44	1.9	25
95 _{Zirc. Niobium}	103.6	0	0	104.3	100
235 _{Uranium}	273.0	0	0	271.0	100

35 EXAMPLE 4 (Electrodeposition of UO_2 - PuO_2) 35

A sintered UO_2 -18% PuO_2 pellet weighing 0.6 g was dissolved together with 100%- H_2SO_4
produced as in Example 1, in 100 g of a ternary eutectic mixture of Li_2SO_4 - K_2SO_4 - Na_2SO_4
having a temperature of 575°C.

The melt contained 91 mg of Pu per g of salt and 430 mg of U per g of salt. After immersing
40 two platinum electrodes in the molten bath as in Example 3, electrolysis was carried out at 1.5 40
V and 30 mA with the melt at a temperature of 600°C. After 45 hours, 0.03 mg of Pu per
gram of salt and 0.015 mg of U per g of salt were left in solution, thus indicating that 99.7%
of uranium and 99.9% of plutonium which were initially present in the melted salts had been
co-deposited on the cathode in the form of mixed oxides.
45

CLAIMS 45

1. An electrochemical method for the reprocessing of irradiated nuclear fuel, which
comprises the steps of:

(a) dissolving the fuel in a salt melt under conditions such that uranium and plutonium
50 therein are converted to sulphate form; 50

(b) subjecting the salt melt to electrolysis conditions to effect electrodeposition of uranium
and of plutonium present in solution in the salt melt at the time of the electrodeposition, and

(c) optionally thermally decomposing plutonium sulphate present in the salt melt prior to said
electrodeposition and removing the plutonium oxide thereby formed from the salt melt.

2. A method as claimed in claim 1, wherein said nuclear fuel comprises the mixed oxides
55 UO_2 - PuO_2 . 55

3. A method as claimed in claim 1, wherein said nuclear fuel comprises the mixed carbides
 UC - PuC .

4. A method as claimed in claim 3, wherein said nuclear fuel additionally comprises thorium
60 carbide. 60

5. A method as claimed in any one of the preceding claims, wherein a metal sulphate salt
melt is employed and dissolution of the nuclear fuel in the salt melt takes place under the action
of absolute sulphuric acid.

6. A method as claimed in any one of claims 1 to 4, wherein a sulphate salt melt bath is
65 employed containing a metal bisulphate for enabling dissolution of the nuclear fuel in the melt 65

to take place.

7. A method as claimed in any one of the preceding claims, wherein the salt melt has a temperature of 550 to 600°C.

8. A method as claimed in any one of the preceding claims, wherein the salt melt is constituted by the system $\text{Li}_2\text{SO}_4\text{-K}_2\text{SO}_4\text{-Na}_2\text{SO}_4$ (78-8.5-13.5 molar %). 5

9. A method as claimed in any one of claims 1 to 7, wherein the salt melt is constituted by the system $\text{Li}_2\text{SO}_4\text{-K}_2\text{SO}_4$ (80-20 molar%).

10. A method as claimed in any one of the preceding claims, wherein step (c) is effected, the temperature of the salt melt being raised to from 750 to 800°C from a temperature at which the fuel has been dissolved in the salt melt thereby to thermally decompose plutonium sulphate present in the salt melt while leaving uranyl sulphate in solution in the salt melt. 10

11. A method as claimed in any one of the preceding claims, wherein, to effect step (b), a pair of electrodes is immersed in the salt melt while the salt melt is at a temperature of 550 to 600°C and deposition of UO_2 or UO_2 and PuO_2 takes place at the cathode with sulphur trioxide and oxygen being evolved at the anode. 15

12. A method as claimed in claim 11, wherein a potential difference of from 0.5 to 1.0 volts is applied between the electrodes.

13. A method as claimed in any one of the preceding claims, wherein, after the electrolysis of step (b), at least part of the salt melt is recycled to step (a).

14. A method as claimed in claim 13, wherein salt melt is decontaminated before being recycled to step (a). 20

15. A method as claimed in any one of the preceding claims, wherein salt melt which is not recycled to step (a) and which contains fission products is conditioned by encasement in a metallic matrix, the encasement being effected by converting the salt melt to a solid granulate by dropping it onto a cold surface and dispersing the granulate in a liquid alloy which is then cast in a container therefor. 25

16. A method as claimed in any one of claims 1 to 14, wherein salt melt which is not recycled to step (a) and which contains fission products is conditioned by conversion to a vitreous silico-titanate in a procedure in which coal and silica are added to the salt melt which is heated to above 800°C, the reaction product obtained being melted at 1000 to 1200°C after the addition of titanium dioxide and then allowed to cool to form a glass. 30

17. A method as claimed in any one of the preceding claims, wherein the nuclear fuel to be reprocessed is fuel from a fast reactor.

18. An electrochemical method for the reprocessing of irradiated nuclear fuel, substantially as described in either of the foregoing Examples 3 and 4. 35

19. An electrochemical method for the reprocessing of irradiated nuclear fuel as claimed in claim 1, substantially as described herein.

20. An irradiated nuclear fuel which has been reprocessed by the method claimed in any one of the preceding claims.