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(72) HOBBS, JEFFREY WILLIAM, GB
(72) PARKES, PETER, GB
(71) BRITISH NUCLEAR FUELS PLC, GB

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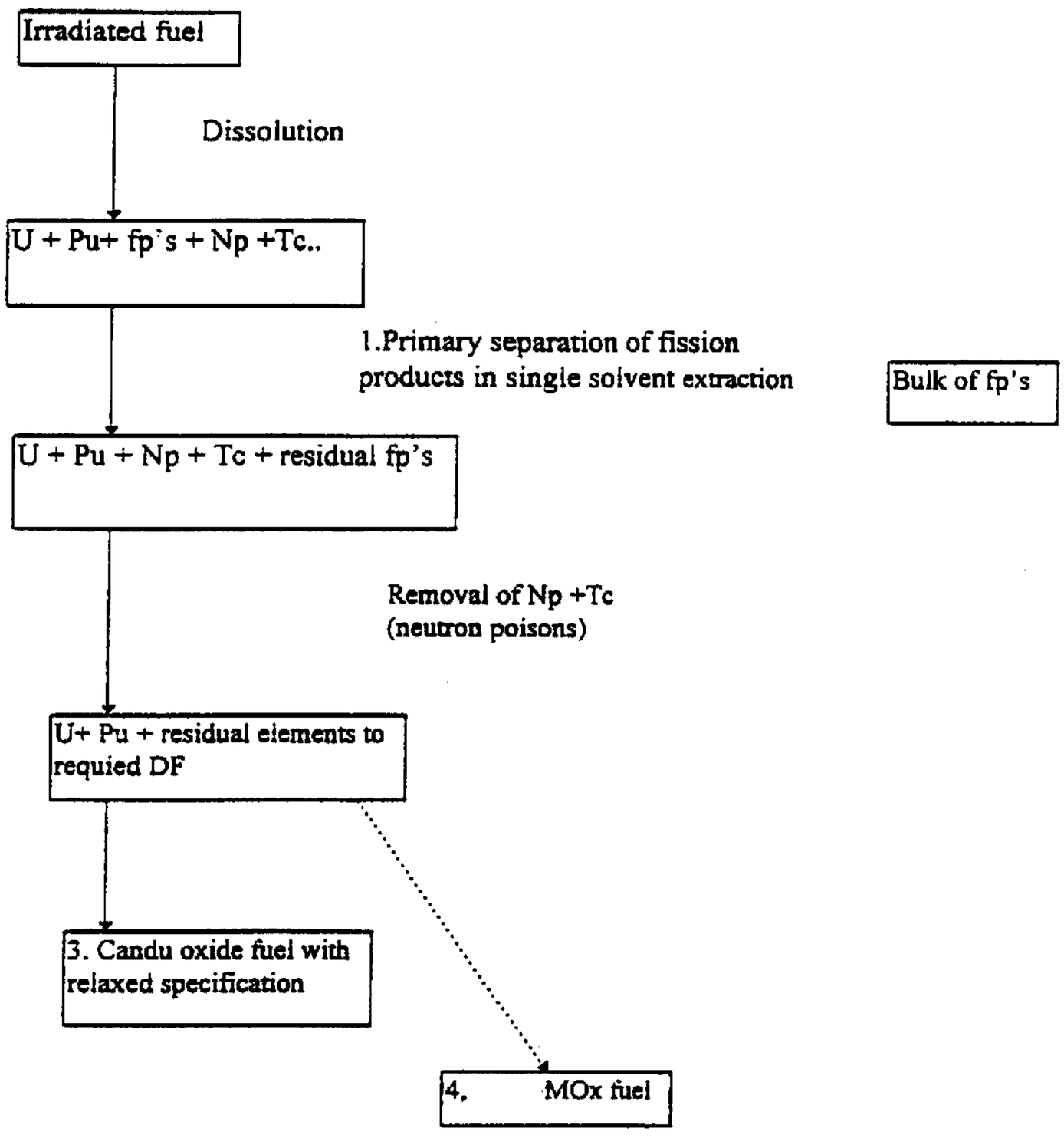
(54) **RETRAITEMENT DE COMBUSTIBLE NUCLEAIRE**
(54) **NUCLEAR FUEL REPROCESSING**



(57) L'invention concerne un procédé de retraitement de combustible nucléaire irradié dans lequel le combustible est dissous et soumis à un procédé d'extraction par solvant, mais où le plutonium et l'uranium ne sont pas séparés après séparation des produits de fission.

(57) Process for reprocessing irradiated nuclear fuel wherein the fuel is dissolved and submitted to a solvent extraction but the plutonium and uranium are not separated from each other after the separation of the fission products.

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(71) Applicant *(for all designated States except US)*: BRITISH NUCLEAR FUELS PLC [GB/GB]; Risley, Warrington, Cheshire WA3 6AS (GB).

(72) Inventors; and

(75) Inventors/Applicants *(for US only)*: HOBB, Jeffrey, William [GB/GB]; British Nuclear Fuels plc, Research & Technology, Sellafield, Seascale, Cumbria CA20 1PG (GB). PARKES, Peter [GB/GB]; British Nuclear Fuels plc, Research & Technology, Sellafield, Seascale, Cumbria CA20 1PG (GB).

(74) Agent: HARRISON GODDARD FOOTE; Belmont House, 20 Wood Lane, Leeds LS6 2AE (GB).

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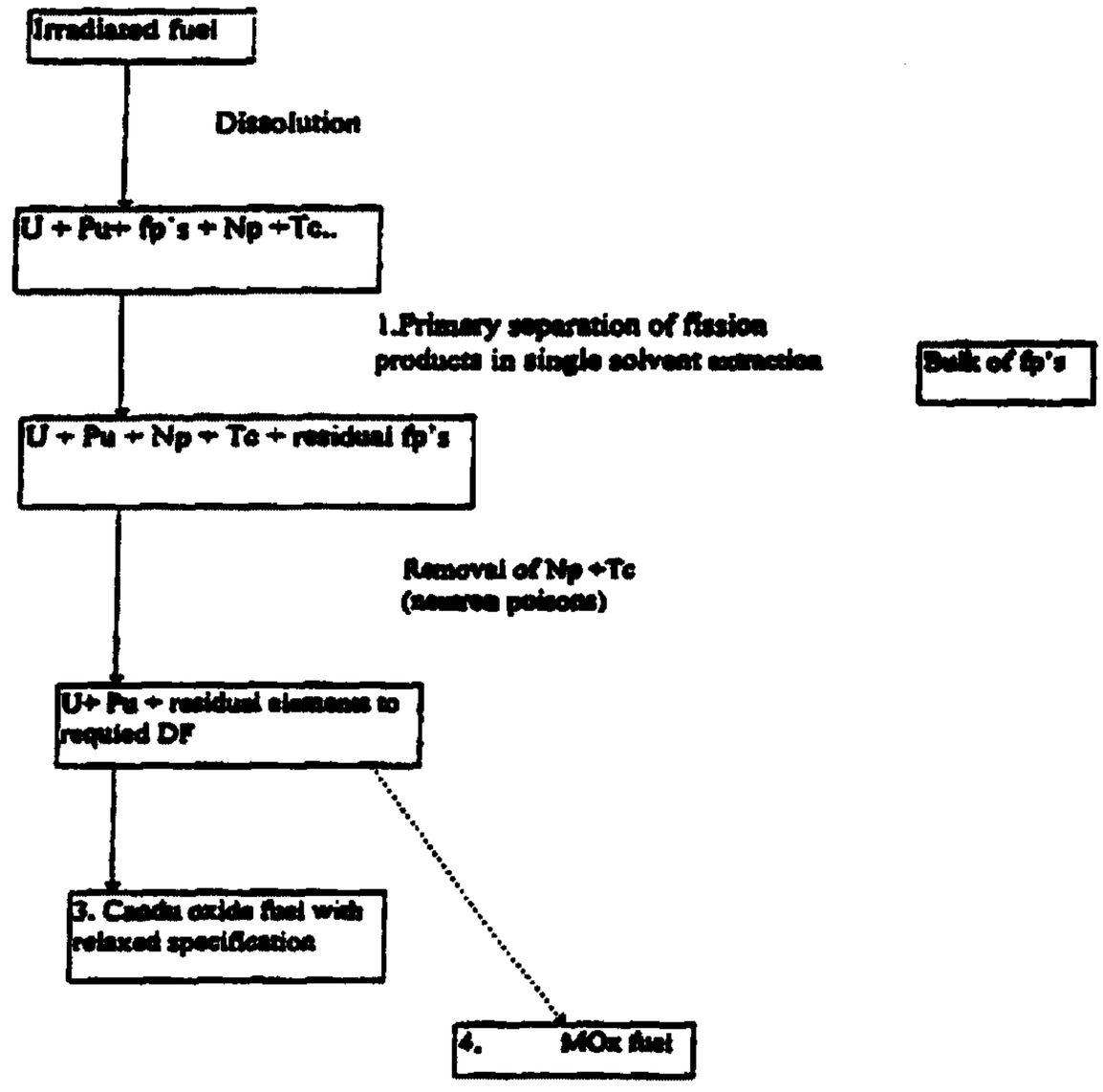
(57) Abstract

Process for reprocessing irradiated nuclear fuel wherein the fuel is dissolved and submitted to a solvent extraction but the plutonium and uranium are not separated from each other after the separation of the fission products.



NUCLEAR FUEL REPROCESSING

The present invention relates to the reprocessing of spent nuclear fuel.

The normal process used for recycling irradiated nuclear fuel is the PUREX process[1]. In this process, spent fuel rods are sheared into small pieces, which are contacted with nitric acid. The nitric acid dissolves uranium, plutonium and fission products. The uranium and plutonium are extracted into a solvent (tributyl phosphate diluted with paraffin). Thereafter, the plutonium is separated from the uranium by a further solvent extraction technique. The PUREX purification process has been operated commercially for many years and is well known to those in the nuclear industry.

CANDU reactors are moderated by heavy water and thus the most efficient reactors in terms of neutron usage. They can therefore use natural uranium fuel, even in the form of the oxide. It has been proposed to re-irradiate spent PWR fuel in CANDU reactors using a scheme avoiding the PUREX process. Such fuel cycles for using spent PWR fuel in CANDU reactors are usually referred to as DUPIC fuel cycles (DUPIC = Dual Use of spent PWR fuel In CANDU reactors). A major constraint on DUPIC fuel cycles is the requirement to minimise the potential for diversion of fissile material (notably plutonium). In prior art proposals, diversion of fissile material has been safeguarded against by avoiding chemical processing. In one proposed process, there is used for the recycle of oxide fuel a dry oxidation-reduction process which uses only gaseous and solid materials. The end product can either be blended with uranium or plutonium and pelletised for use in a thermal reactor or pelletised without blending and used in a CANDU reactor.

The proposed DUPIC fuel cycles are inflexible, in that they cannot be applied to the use of irradiated fuel in natural cored reactors other than CANDU reactors or modified for use in enriched cored reactors.

The present invention provides a process for reprocessing irradiated nuclear fuel comprising dissolution of the fuel followed by solvent extraction but in which the uranium and plutonium are not separated. The dissolved uranium and plutonium (plus in practice residual

fission products, Np and Tc) are then processed into a nuclear fuel product, this processing including the fabrication of fuel using gel precipitation.

In another aspect, the invention provides a modification of the PUREX process in which the dissolved uranium and plutonium are not separated after primary separation of fission products but are processed into a nuclear fuel product without being separated, the nuclear fuel product being obtained via gel precipitation.

Preferred embodiments of the invention are set forth in the accompanying dependent claims.

The present invention is further described by way of example only with reference to the accompanying drawings, in which:

Figure 1 is a simplified flow sheet of the PUREX process; and

Figure 2 is a flow sheet of a process of the invention.

Figure 1, therefore, illustrates the existing and well-known PUREX recycling procedure. No detailed explanation of the procedure is required here. However, it will be noted that it is a complex multi-stage procedure which involves separation and purification of uranium and plutonium. Such separation creates risk of diversion of the plutonium. The end product UO_2 and PuO_2 products can be blended back together to produce MOX fuel or stored.

The process of the invention involves primary separation of fission products, for example using a procedure similar to the primary separation step of the PUREX process, but thereafter departs from the PUREX flow sheet in not separating the uranium and plutonium and going through the UP and PP cycles. Rather, the primary separation step is followed, if necessary or desirable, by further removal of contaminants (especially of the neutron poisons Np and Tc). The solution remaining after any such removal of further contaminants may be processed into a natural cored reactor fuel or into an enriched cored reactor, for example using a procedure known *per se*. An exemplary natural cored reactor is the CANDU reactor.

The solution may be processed by gel-precipitation or even by conversion into oxide powder, for example for fabrication into fuel pellets.

An exemplary process of the invention, therefore, is illustrated by Figure 2. Irradiated fuel is dissolved, for example using nitric acid as in the PUREX process. Alternatively, the spent fuel may be dissolved in an aqueous carbonate-containing medium, for example as described in EP-A-282810.

Following dissolution, the resultant liquor is subjected to primary separation of fission products by solvent extraction, preferably but not necessarily in a single solvent extraction step.

The primary solvent extraction of the PUREX process gives decontamination factors (DFs) of at least 1,000 for all species except Np and Tc (the DF for a species is determined by dividing its activity in the feed solution by its activity in the final solution). Both these metals are neutron poisons and it may be necessary or desirable to remove them after the primary solvent extraction.

The solution remaining after the primary solvent extraction and any removal of Np and Tc may be processed into CANDU oxide fuel with a relaxed specification. Alternatively, the solution may be processed into a MOX fuel (MOX = mixed oxide (UO_2 + PuO_2)), by blending the U, Pu with additional fissile material (enriched uranium comprising U-235 or plutonium). The additional fissile material may be blended with product of the invention either as oxide powder or as a nitrate solution. An exemplary MOX fuel is light water reactor (LWR) fuel. The specification of LWR fuel is similar to that of CANDU fuel, which means that the process of the invention is flexible in being usable to make either fuel.

The solution may be processed into CANDU oxide fuel by, for example, one of the following methods (i) the oxide as powder or granules could be pressed into pellets; or (ii) the oxide as powder granules of optimised size distribution could be poured directly into a fuel pin using vibro-compaction. However, it has been found that optimum results are

achieved when the product stream is processed by gel-precipitation, for example to produce vibro-spherical fuel.

The metal nitrates in the solution may be converted to their oxides by concentrating the nitric acid product stream and converting the U, Pu nitrates into oxides. The concentrated solution may be finished to oxide powder by thermal denitration (TDN) or, less preferably, by precipitation, and then be fabricated into fuel pellets. The oxide powder will often require milling to ensure that pellets of high enough density are formed on pressing the powder.

Gel precipitation is particularly preferred as a processing technique, because the material inherently has relatively high radioactivity as a result of its low decontamination factor. Such high radioactivity means that it is very beneficial to use processes which offer the opportunity for operating with high levels of shielding, automation, low maintenance and a low number of process stages. Gel precipitation can offer such benefits as well as a dust free process. The product of gel precipitation is free flowing but not dusty.

Gel-precipitation is also a preferred technique for processing the solution into MOX fuel.

Gel-precipitation was introduced in the early 1970's primarily as a method of producing fast reactor fuel that could be loaded directly into the fuel pin. There are 2 principle methods. Internal gelation involves the formation of droplets in an immiscible liquid (silicone oil or a chlorinated hydrocarbon) or in air. Homogeneous precipitation occurs because hexamethylene tetramine in the feed solution undergoes thermal decomposition and releases ammonia internally as the sphere drops in the hot gelation media.

External gelation, or polymer supported precipitation, involves the addition of a water soluble organic polymer to a nitrate feed solution (eg a solution of $Pu(NO_3)_4$ and $UO_2(NO_3)_2$). This additive maintains the structure of the droplet as it precipitates when contacted with ammonia gas or an ammonium solution.

The gel spheres are typically washed in hot water to remove reaction products from the gelation stage and then dried, for example by extracting the water into hexanol or another

solvent. The spheres are calcined in, usually, a carbon dioxide atmosphere to debond them, i.e. to remove polymer and convert the spheres to microspheres of porous metal oxide.

The gel-precipitation process used is not critical. The skilled reader will not need instruction as to gel-precipitation techniques, but one technique which may be mentioned is the so-called KEMA internal gelation route, originally devised in 1974[2] but further developed subsequently. An alternative technique is an external gel-precipitation method developed by the UK Atomic Energy Authority[3-6]. More recently there has been developed another applicable process called total gelation, which in effect combines the internal and external routes[7,8].

The invention enables provision of a process for recycling nuclear fuel into CANDU or thermal reactors; of this latter category, LWRs are preferred. The invention has the potential to generate more electricity for the same volume of high-level waste than once-through irradiated fuel, and therefore represents a significant environmental advance. Nonetheless, the invention primarily uses PUREX technology, in which there is an abundance of industrial experience as well as ongoing development; other familiar techniques usable in the invention include gel-precipitation and vibro fuel production.

The invention is resistant to diversion of plutonium, not only because it is not separated from uranium but also because the intermediate and final products will in practice contain residual fission products, Np and Tc. These contaminants make the products more difficult to handle safely and require special facilities for their removal.

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CLAIMS

1. A process for reprocessing irradiated nuclear fuel comprising dissolution of the fuel followed by solvent extraction wherein the dissolved uranium and plutonium are not separated, which further includes processing the dissolved uranium and plutonium into a nuclear fuel product, characterised in that said further processing includes the fabrication of fuel using gel precipitation.

2. A process of Claim 1 wherein the fuel is dissolved in nitric acid.

3. A process of Claim 1 or Claim 2 wherein dissolution of the fuel is followed by separation from the solution of fission products.

4. A process of any of Claims 1 to 3 wherein dissolution of the fuel is followed by removal from the solution of one or both of Np and Tc.

5. A process of Claim 4 wherein the Np and Tc are removed after the fission products have been separated.

6. A modification of the PUREX process wherein the dissolved uranium and plutonium are not separated after primary separation of fission products but are processed into a nuclear fuel product without being separated, said process involving gel precipitation.

7. A process of any of Claims 1 to 6 which further includes combining extra fissile material with uranium and plutonium to make MOX fuel for LWRs.

8. A process of Claim 7 wherein the fissile material comprises Pu.

9. A process of Claim 7 wherein the fissile material comprises U-235.

10. A process of any of Claims 1 to 9 wherein the nuclear fuel product is for a natural cored reactor.

11. A process of any of Claims 1 to 9 wherein the nuclear fuel product is for an enriched cored reactor.

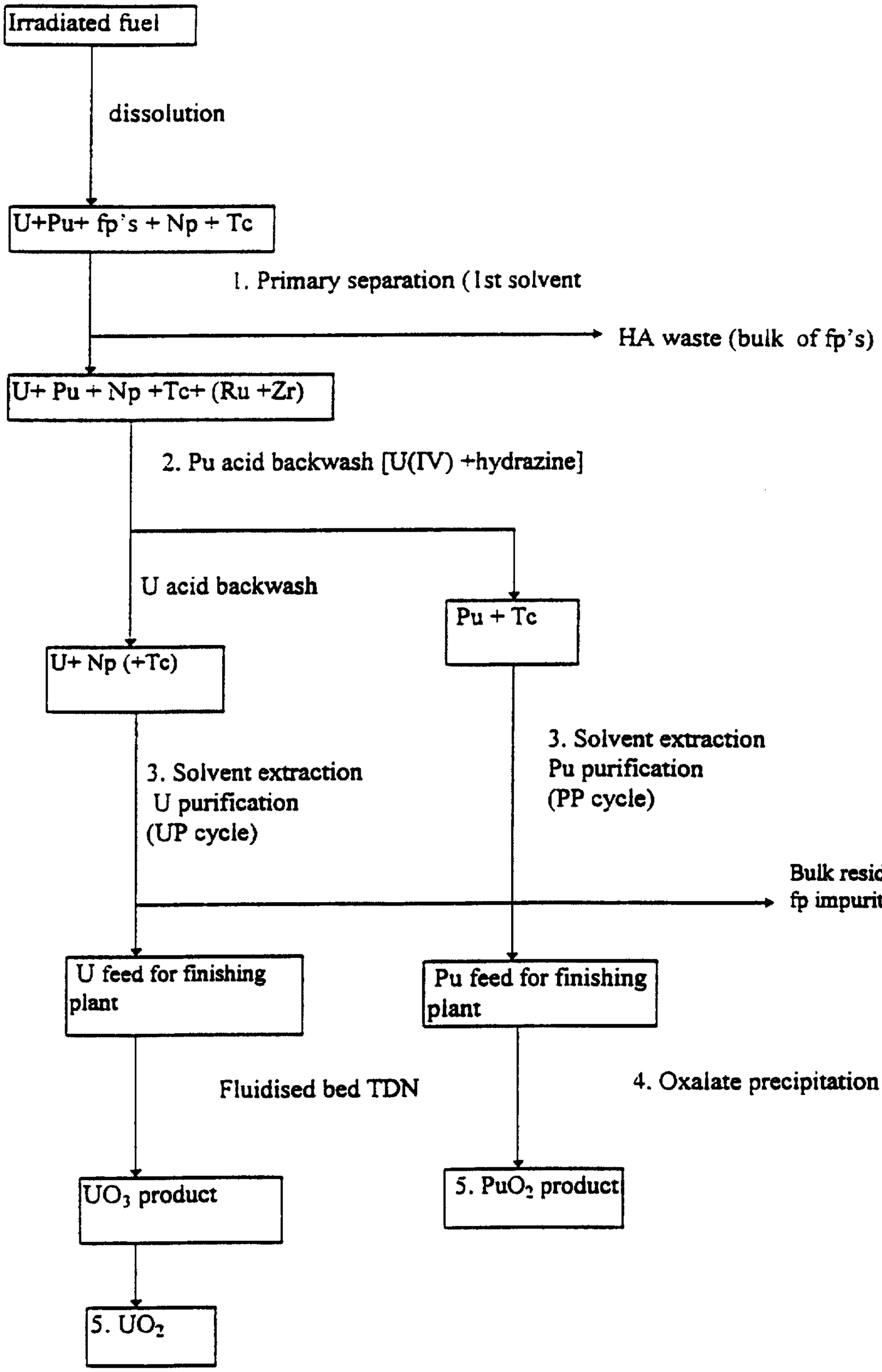
Irradiated fuel
dissolution
U+Pu+ fp's + Np + Tc
1. Primary separation (1st solvent
HA waste (bulk of fp's)
U+ Pu + Np +Tc+ (Ru +Zr)
2. Pu acid backwash [U(IV) +hydrazine]
U acid backwash
Pu + Tc
U+ Np (+Tc)
3. Solvent extraction
U purification
(UP cycle)
3. Solvent extraction
Pu purification
(PP cycle)
Bulk residual
fp impurities
U feed for finishing plant
Pu feed for finishing plant
Fluidised bed TDN
4. Oxalate precipitation
UO_3 product
5. PuO_2 product
5. UO_2

Figure 1

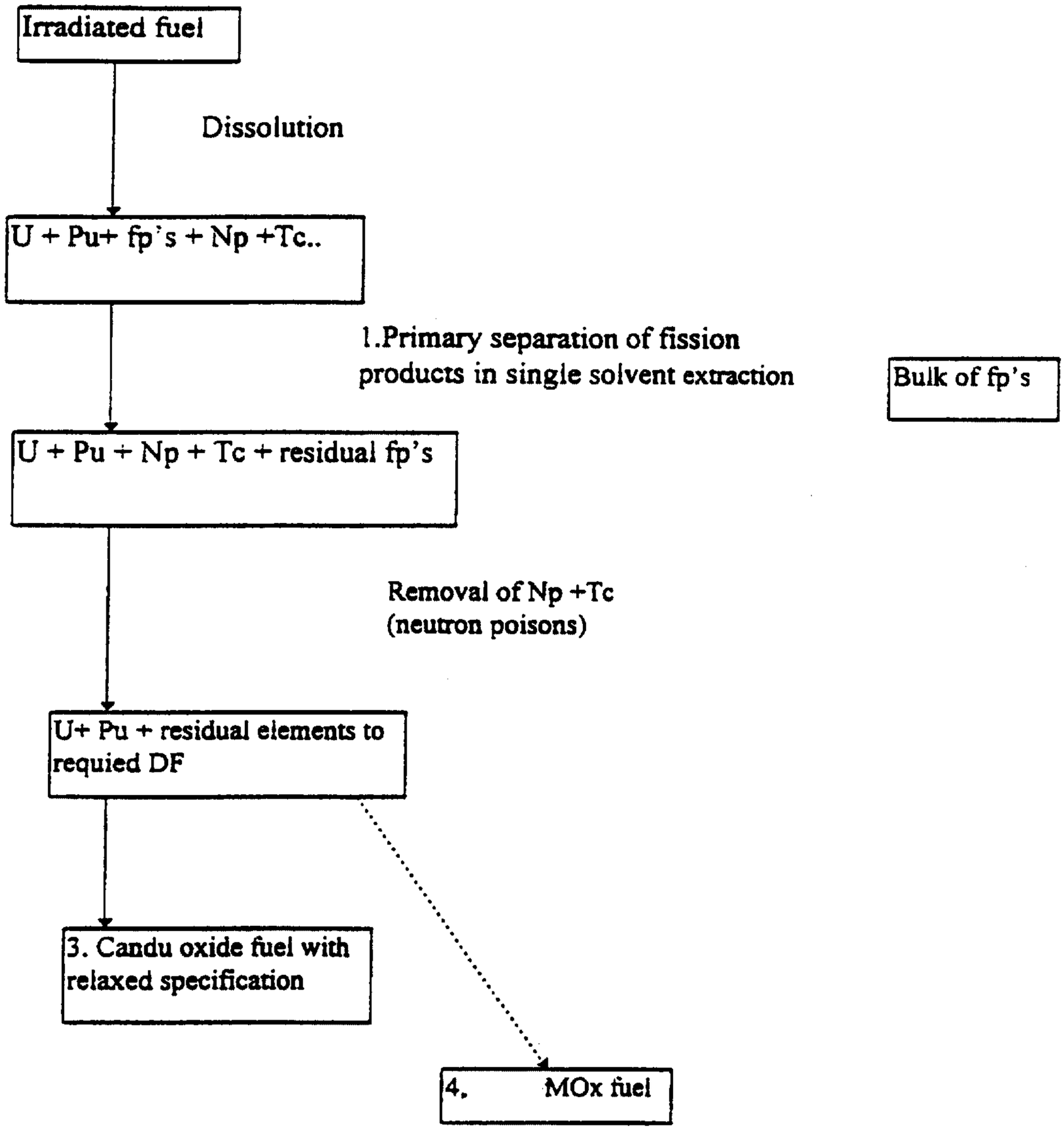
Irradiated fuel
Dissolution
U + Pu+ fp's + Np +Tc..
1.Primary separation of fission products in single solvent extraction
Bulk of fp's
U + Pu + Np + Tc + residual fp's
Removal of Np +Tc (neutron poisons)
U+ Pu + residual elements to requied DF
3. Candu oxide fuel with relaxed specification
4. MOx fuel

Figure 2