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SOME ASPECTS OF A TECHNOLOGY OF PROCESSING WEAPONS GRADE PLUTONIUM TO NUCLEAR FUEL

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The concept of Russia to use fissile weapons-grade materials, which are being recovered from nuclear pits in the process of disarmament, is based on an assessment of weapons-grade plutonium as an important energy stuff intended for the usage in view of fuel of nuclear power facilities. This valuable raw-material is the national property of Russia and its effective usage on behalf of people is a significant statuary task.

However, at the path of involving plutonium excessive from the purposes of national safety into industrial power engineering there are a lot of problems, from which effectiveness and terms of its disposition are being dependent upon. Those problems have political, economical, financial and environmental character.

In this report we would like to outline several technological problems of processing weapons-grade metallic plutonium into MOX-fuel for reactors based on thermal and fast neutrons. In particular, we will touch the issue of conversion of the metal into dioxide from the viewpoint of fabrication of pelletized MOX-fuel.

The processing of metallic weapons-grade plutonium into nuclear fuel is rather complicated and multi-stage process, every stage of which is own production. Some of the stages is absent while production of MOX-fuel, for instance a stage of the conversion, i.e. transferring of metallic plutonium into dioxide of the ceramic quality.

At this stage of plutonium utilization some tasks must be resolved as follows:

- As a result of the conversion, a material purified from ballast and radiogenic admixtures have to be obtained. This one will be applied to fabricate pelletized MOX-fuel going from morphological, physico-mechanical and technological properties.

- It is well known that metallic gallium, which is used as an alloying addition in weapons-grade plutonium, actively reacts with multiple metals. Therefore, an important issue is to study the effect of gallium on the technology of MOX-fuel production, quality of the pellets, as well as the interaction of gallium oxide with zirconium and steel shells of fuel elements depending upon the content of gallium in the fuel.

The rate of the interaction of gallium oxide, containing in MOX-fuel, with zirconium alloys and stainless steels is not known.

The knowledge of its acceptable content in fuel will permit to choose an optimum variant of plutonium purification from gallium. Therewith, it is possible to guess that the acceptable content of gallium in plutonium dioxide used for fabrication of the fuel for the reactors on fast and thermal neutrons can be different.

To furnish the environmental safety of MOX-fuel during its storage, transportation and production, plutonium dioxide must be refined from americium-241, which is being accumulated in weapons-grade plutonium on the account of decaying plutonium-24 1.

At present, in Russia there are several technologies under study, which principally allow to resolve the tasks mentioned above.

All technologies have advantages and disadvantages, but neither of them can be considered complete since in each one there are fields not to be studied sufficiently, and mainly in many processes a final result, the quality of plutonium dioxide, is not assessed.

I would like to tell you more in detail about some technologies of the conversion, which are under study by scientific centers of MINATOM.

Aqueouschemical technology

The technology is separated upon several variants hinging upon the way of the dioxide fabrication.

Oxalate precipitation (Appendix I). This technology has been studied better; its merits and limitations are given in the Table 1.

Merits and limitations of the technology of oxalate precipitation

Merits	Limitations
1. Long-term experience of dioxide plutonium fabrication at a full scale for the eventual metallurgical production (the presence of equipment). 2. The availability of experience of using such plutonium for fabrication of MOX-fuel for the reactors BN-350 and BN-600 by a selective choice of the material parties. 3. The feasibility of correcting properties of an initial dioxide powder at the stage of fabrication of a press-powder.	1. Not sufficient amount of R&D works with the purpose of selecting modes of fabrication of a powder of the ceramic sort. 2. High dispersity, dust formation, high adhesive ability. 3.Instability of the powder features that is defined by the complexity of parameters of the technological modes (composition of solutions, temperature, duration of the process, intensity of blending, etc.) 4. Liquid waste enriched by plutonium (up to 200 mg/l) must be processed additionally.

Ammonium precipitation (Appendix 2a) or co-precipitation of plutonium and uranium *(Appendix-t* 2b) with parallel involving of surface-active substances into the solution. This method allows to obtain a prepared master-blend with a regulatory ratio of PuO_2 and U02.

The technology of ammonium precipitation and especially co-precipitation from our point of view is more favorable because it allows to obtain the prepared master-blend. On the account of adding the surface-active substances, the powders do not raise dust. Basic merits and drawbacks of the technology of ammonium precipitation are presented in the Table 2, but the technology of ammonium co-precipitation - in the Table 3.

Table 2.

Merits and limitations of the technology of ammonium precipitation with the surface-active substances

Merits	Limitations
1. Obtaining of agglomerates of plutonium dioxide with the given dimension and having insignificant dust-formation, good fluidity, low adhesion and technological characteristics according to technical requirements. 2. Intensification of the process of powders fabrication due to formation of quickly filtrating sediments. 3. The possibility of using regular radiochemical equipment. 4. Low content of plutonium in the master taps (<5 mg/l) 5. Preliminary positive experience of using such powders to fabricate pellets by the technology of mechanical blending.	1. Not sufficient amount of R&D works with the purpose of selecting modes of fabrication of a powder of the ceramic sort. 2. The absence of special equipment for the stages of precipitation and granulation. 3 The necessity of a stage of preliminary reduction of plutonium in the solution by the tree-valent condition before precipitation.

Table 3.

Merits and limitations of the technology of ammonium co-precipitation with the surface-active substances

Merits	Limitations
1. The availability of using such a type of the material to fabricate MOX-fuel for the reactors BN-350 and BN-600 at the PA "Mayak". 2. A great volume of R&D works to define technological modes providing fabrication of the powders with required features. 3. Obtaining of agglomerates of plutonium dioxide with the given dimension and having insignificant dust-formation, good fluidity, low adhesion and technological characteristics according to technical requirements. 4. Intensification of the process of the powders fabrication due to formation of quickly filtrating sediments. 5. The possibility of using regular radiochemical equipment. 6. Low content of plutonium in the master taps (<5 mg/l). 7. The presence of positive laboratory experience of using the co-precipitated master-blend to fabricate fuel for the reactors VVER. 8. Reduction of a technological process of pellets fabrication.	1. The absence of special equipment for the stages of precipitation and granulation. 2. Some difficulties of correcting properties of an initial granulate of the mixed oxides at the stage of pellets fabrication.

The aqueouschemical technology in all variants permits to purify plutonium from gallium, americium and considerable amounts of ballast admixtures almost completely. It is the effective, but rather expensive technology. Along with a complicated process of metallic plutonium dissolution, its main limitation is the formation of considerable amounts of waste basically in view of aqueous tailings. But, one should note that the technology of their disposal is developed and mastered by the Russian atomic industry.

A pyrochemical technology (Appendix 3) is more reliable from the viewpoint of the absence of such waste. It is proposed to use hydration of metallic plutonium and the conversion of hydride into dioxide. Therewith, the hydride can be transferred to nitride and then to dioxide.

However, the pyrochemical process has a lot of uncertainties ("white blots") related to the absence of purification from americium and ballast admixtures, ineffective purification from gallium and high potential explosive danger caused by the presence of hydrogen and oxygen in the same process. Properties of the dioxide obtained by means of a dry technology must be studied more diligently. Our Institute does the very subject. In particular, we began to study the behavior of gallium at all stages of MOX-fuel production with the aim to work off the technological regimes that allow to disregard gallium from the fuel and on the other hand to provide therewith the formation of the dioxide of the ceramic sort.

By the way, **a combined method of the conversion of metallic plutonium,** in which merits of the pyrochemical and aqueouschemical technologies are being used, is under research (Appendix +). A poor place of the aqueouschemical technology is the preparation of metallic plutonium to its dissolution (the necessity of plutonium grinding) and also a labor-intensive process of the dissolution.

Finally dispersed products of the pyrochemical process is easily dissolved in nitric acid with an addition of fluoride ion. In the forth, a merit of the aqueous processes as effective extraction of plutonium from gallium, americium and the ballast admixtures and warranty fabrication of plutonium dioxide of the ceramic sort is used.

A pyrochemical technology of the conversion of metallic plutonium in molten salts (Appendix 5) is being developed. This technology is worked off by RIAR (Dimitrovgrad) well in order to obtain vibro-pressed fuel, which is applied in the reactor BOR-60 and has been tested in the industrial reactor BN-600. However, the technology of fabrication of the dioxide of the ceramic sort for pelletized MOX-fuel whilst is absent. Works are under way in RIAR to realize this technology. A complicated problem of the pyrochemical technology of the conversion is to develop a technology of the complex utilization of wastes of this process from the standpoint of its economical and environmental optimization.

MOX-fuel obtained by different technologies of weapons-grade plutonium proessing must pass pre-reactor tests, a reactor verification in a research reactor, and post-reactor investigations. Without this complex of the works it is impossible to get a license of GAN to load the fuel into commercial reactors. By this time, tests have been made in breeders of pelletized MOX-fuel, fabricated by the aqueouschemical technology (oxalate precipitation, ammonium co-precipitation), and vibro-pressed MOX-fuel, fabricated by the pyrochemical technology. Preparation to test pelletized MOX-fuel for light-water reactors is in progress.

The technologies, which are under study and will furnish fabrication of the dioxide met to the requirements of the ceramic sort with purification from gallium, americium and the ballast admixtures, will be assessed as potentially applicable for industrial aims. However, to select one or two of the technologies it will be necessary to make comparable objective appraisals of the technologies upon the following parameters:

- Economical efficiency of the process;
- The presence and complexity of equipment;
- The volume of waste and difficulty of its reprocessing;

- Environmental safety of the technology, -including nuclear, radiation, explosive and fire ones;

- The availability of engineering infrastructure needed to realize the technology at industrial sites;

- The necessity and volume of investments needed to create an industrial production of the technology;

- A series of other aspects, including social.

Going from those deliberations, before making any decision about enacting MOX-fuel production it is necessary to scrutinize variants of the technologies, to assess the quality of the product, a possibility and terms of licensing the fuel, to implement engineering feasibility studies of the technologies and finally upon the results of these works to accept an optimum decision.

We believe that it is advisable to tell a few words about another aspect of plutonium reprocessing into the nuclear fuel. To utilize plutonium more reasonably there is a need to develop new types of the fuel, which would allow to decrease reproduction of plutonium dramatically or rather completely. A technology of the fuel with the content of plutonium up to 45-50 wt% (and without uranium) with inert matrices (diluants) of the ceramic or carcass type is being studied in our institute.

The fuel of the ceramic sort is plutonium mononitride or monocarbide. Zirconium mononitride or monocarbide can be used as a diluant.

A laboratory scale technology of fabrication of fuel cores from 54.5 % wt PuC and 45.5 % wt ZrC has been developed. An experimental set of the cores was produced; reactor tests of experimental fuel elements in the reactor BOR-60 at the linear output of 400-500 Wt/cm up to burn-up equals to 8 % wt. All elements retained hermetically and were permissible for eventual exploitation. Swelling did not exceed 1 % wt per percent of the burn-up. The yield of gaseous products of decaying was less then 20 % wt of the forming quantity. Only local fields of carbonizing the shell were identified.

At present, an experimental set of fuel elements made from plutonium mononitride and zirconium nitride (40 % wt PuN, 60 % wt ZrN) for reactor tests in the BOR-60 are being fabricated.

A material on the base of a porous carcass from different refractory high-temperature compositions (zirconium carbide or nitride, or its solid solutions with incorporation of plutonium) is the other variant of the fuel with the high content of plutonium intended for its use in reactors-burners.

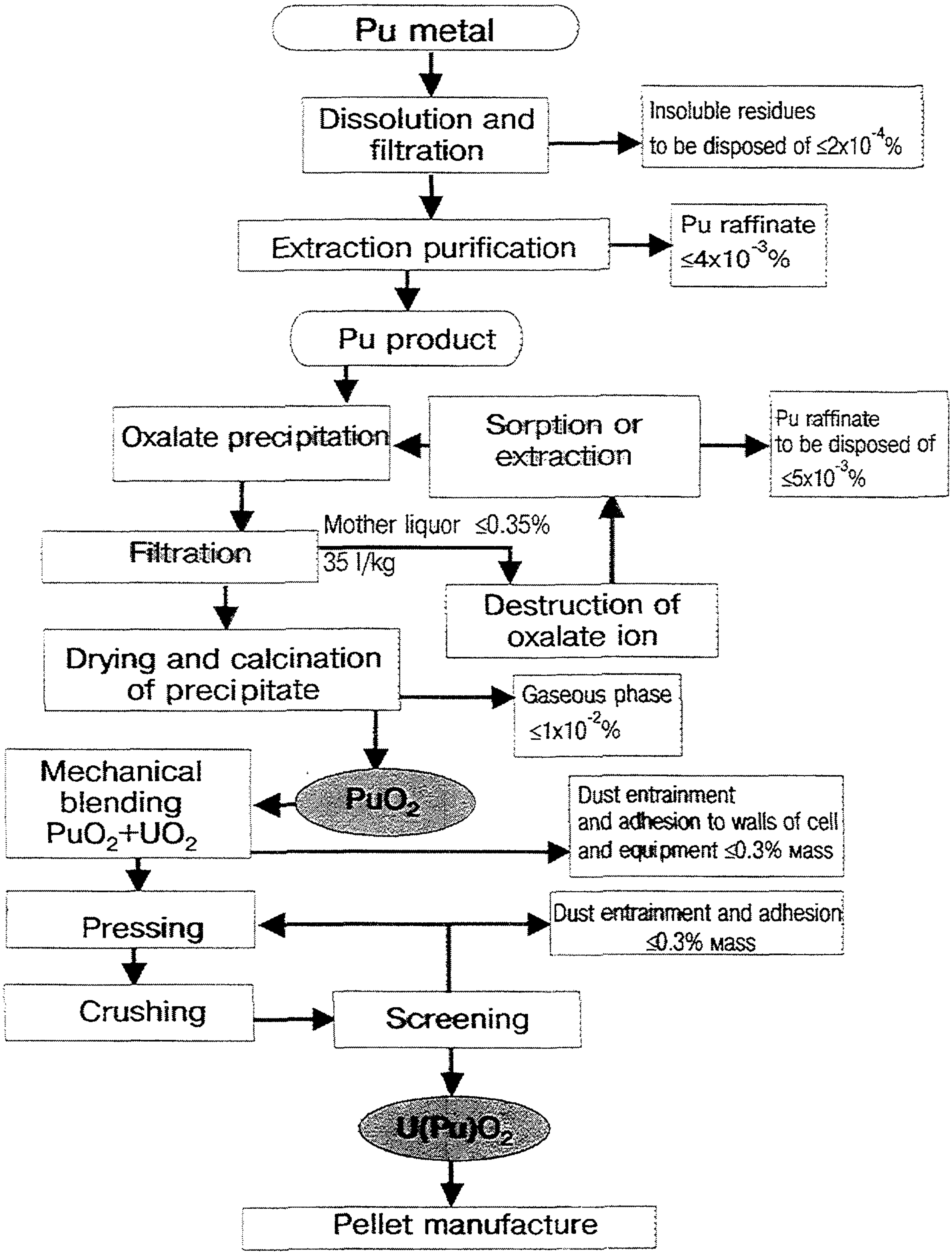
The porous carcass from zirconium carbide or nitride (porosity is within 20-80 % vol) is impregnated by plutonium-containing solutions, from which plutonium dioxide is being formed during the thermal destruction.

This way can also be used for transmutation of long-term high-radioactive wastes, containing minor actinides (neptunium, americium, and curium). It permits to obtain in porous of the cores a mixture of oxides of different nuclides. By the way, there is a possibility of profiling of the content of a fissile component upon the radius and longevity of the core, therewith accurate dosing of the fissile material is provided by parameters of the technology.

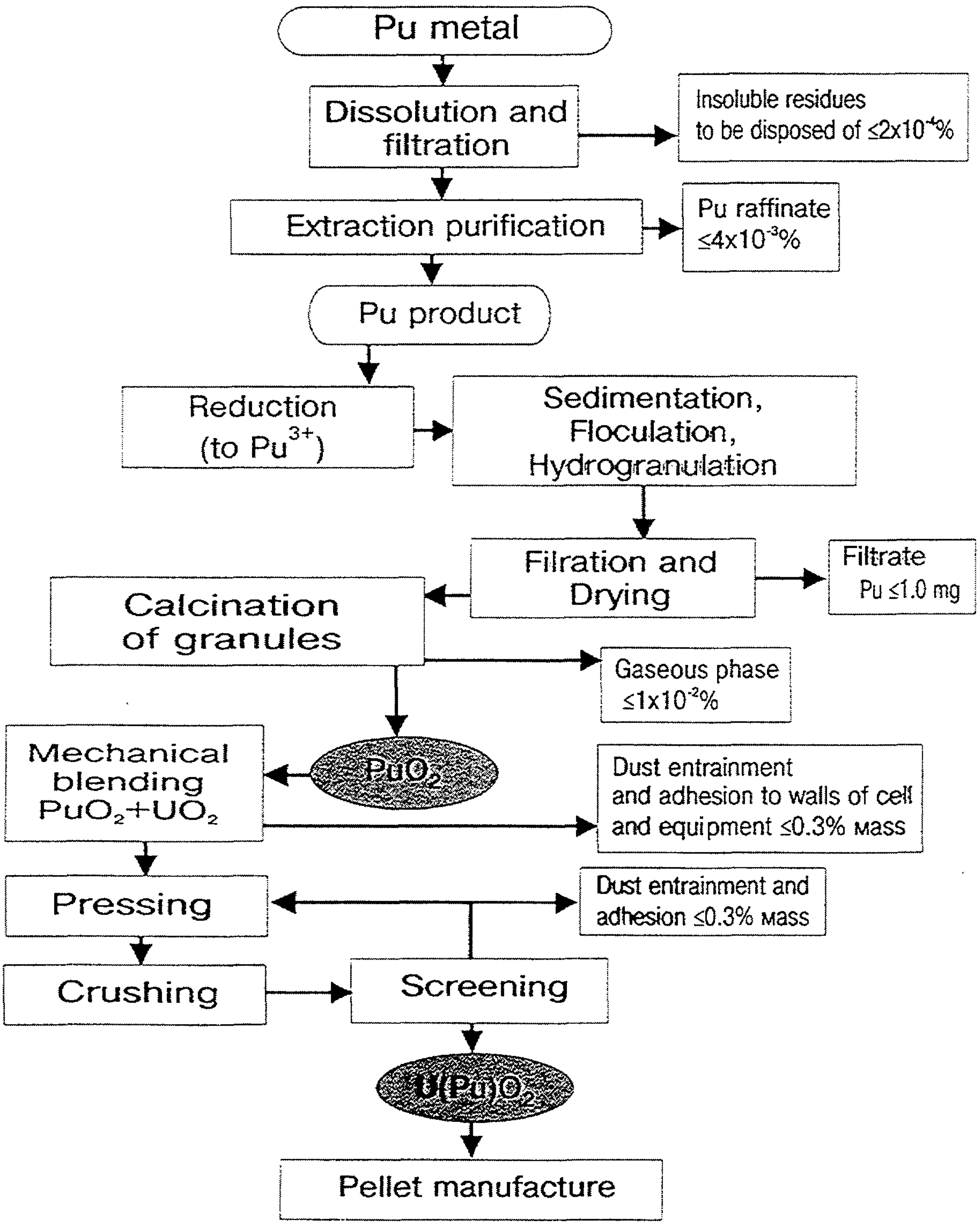
Right now we are conducting the works on the post-reactor study of these compositions.

Appendix 1

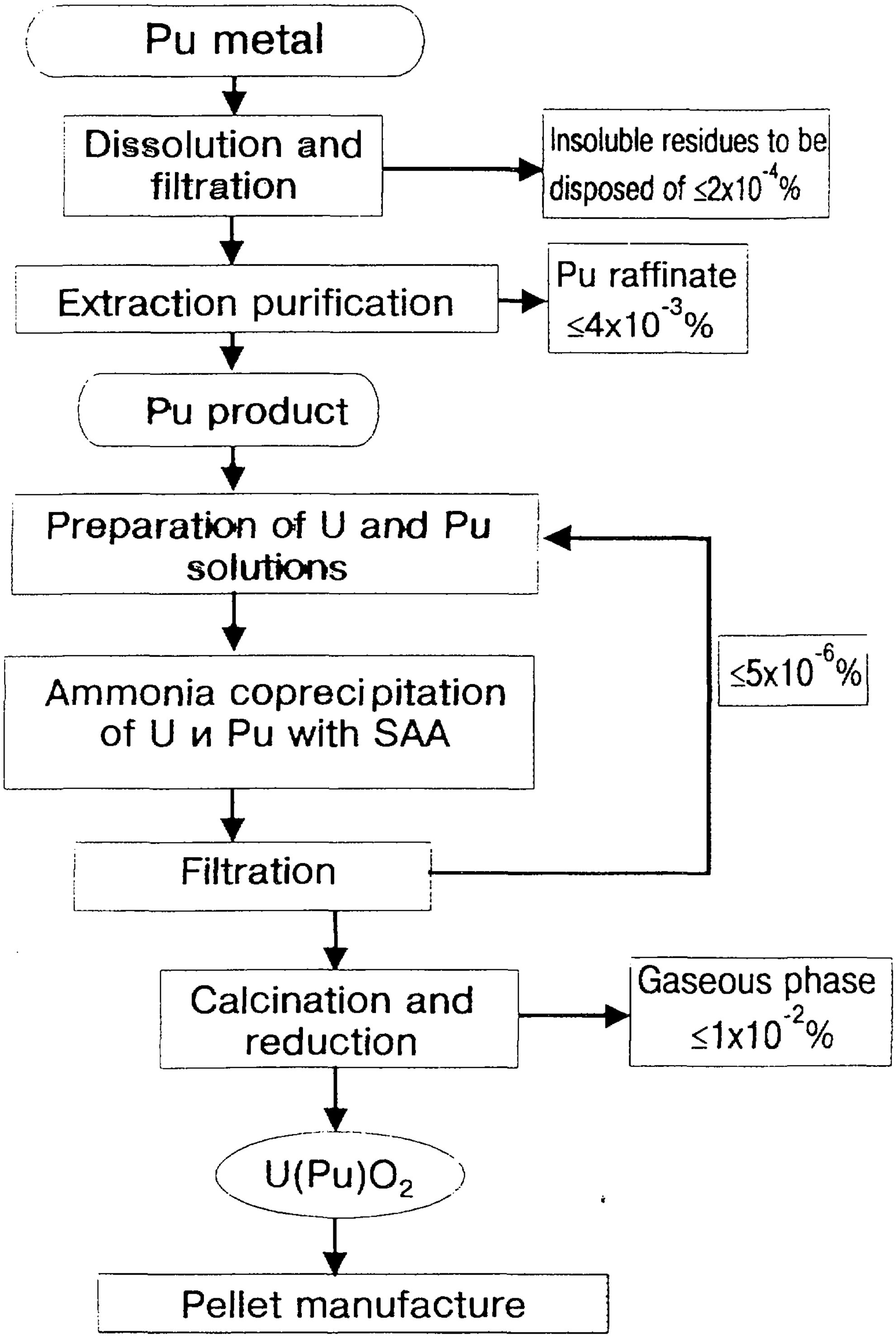
OXALATE PROCESS SCHEMA OF WEAPON'S PU METAL CONVERSION TO MOX-FUEL



AMMONIUM PROCESS SCHEMA OF WEAPON'S PU METAL CONVERSION TO MOX-FUEL

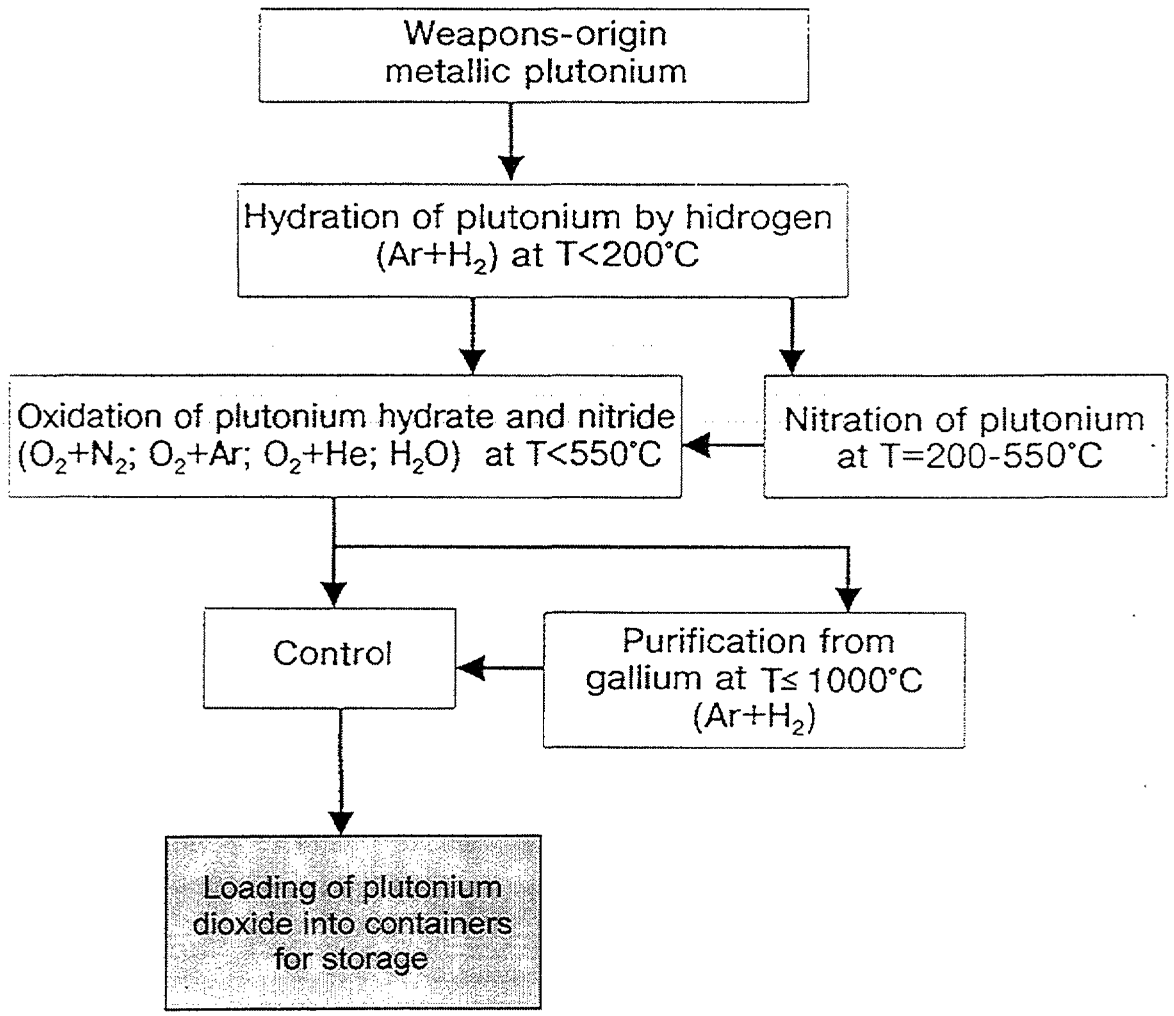


«GRANAT» PROCESS FLOW SHEET OF WEAPON'S GRADE PU CONVERSION TO MOX-FUEL (ammonia coprecipitation of U and Pu with SAA)



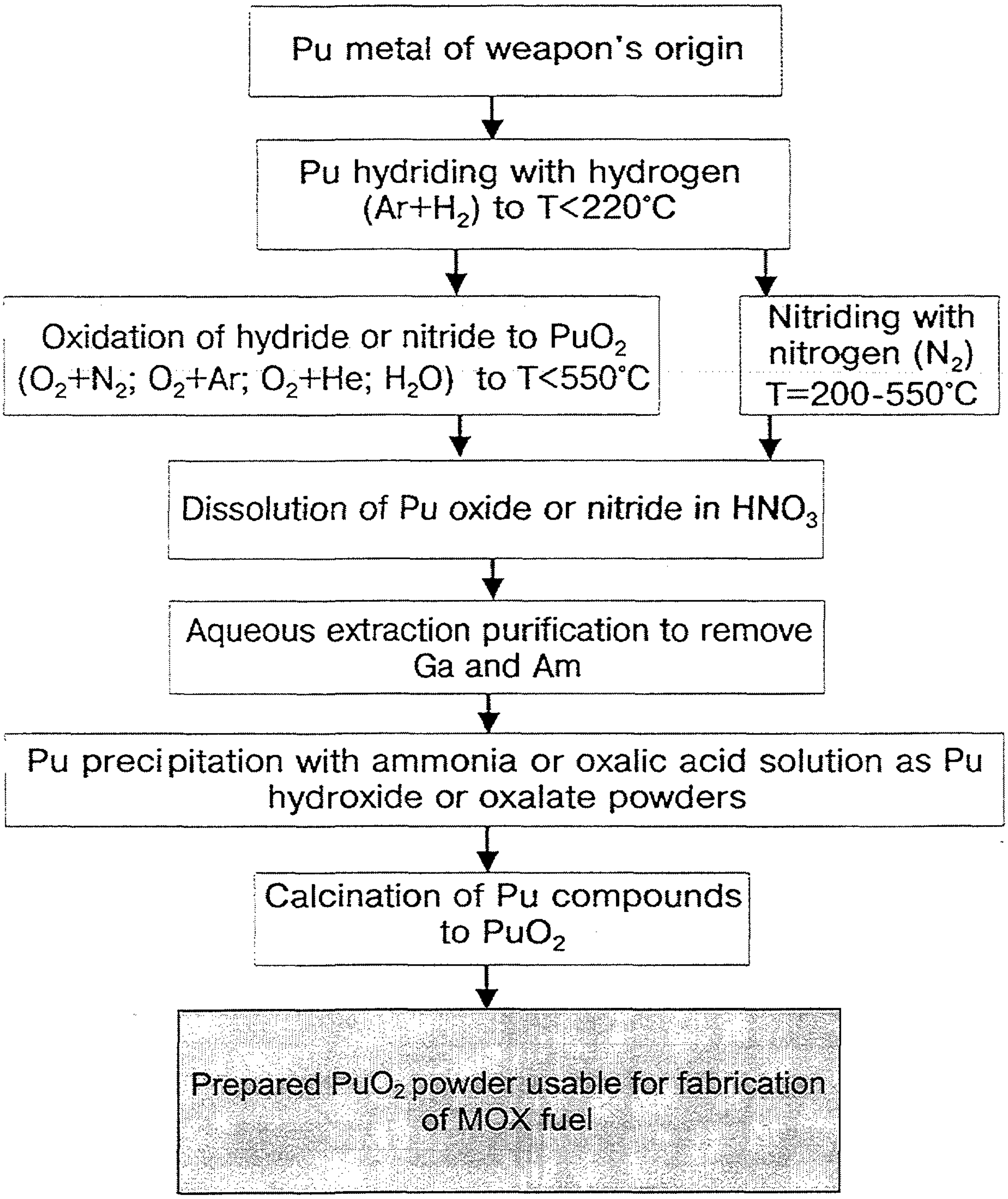
Appendix-3

A FLOW SHEET OF A PYROMETALLURGICAL PROCESS OF TRANSFERRING WEAPONS-GRADE PLUTONIUM INTO PLUTONIUM DIOXIDE

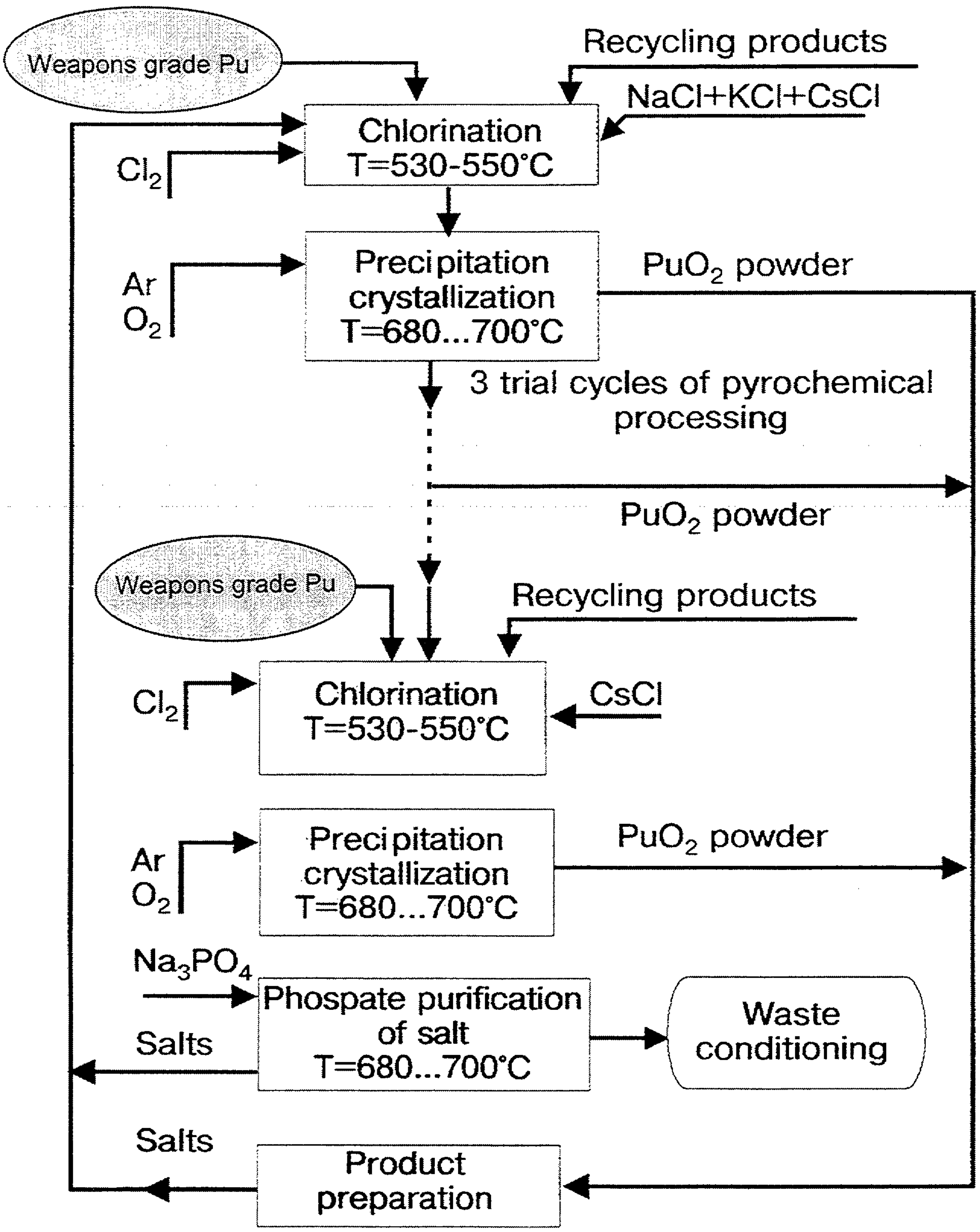


Appendix 4

PROCESS FLOW SHEET of PuO_2 preparation from weapon's crade Pu (combined schema)



Appendix 5



Weapons grade Pu
Recycling products
NaCl+KCl+CsCl
Chlorination
T=530-550°C
Cl2
Ar
O2
Preci pitation
crystallization
T=680...700°C
PuO2 powder
3 trial cycles of pyrochemical
processing
PuO2 powder
Weapons grade Pu
Recycling products
Cl2
Chlorination
T=530-550°C
CsCl
Ar
O2
Preci pitation
crystallization
T=680...700°C
PuO2 powder
Na3PO4
Phospate purification
of salt
T=680...700°C
Salts
Waste
conditioning
Salts
Product
preparation