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MONONITRIDE URANIUM-PLUTONIUM FUEL OF FAST LEAD-COOLED REACTORS

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SUMMARY

The processes of producing mixed mononitride fuel of uranium-plutonium alloys and their oxides are developed by RIIM about forty years. Since it is proposed to use mixed nitride made of power-grade plutonium and uranium oxides as the initial fuel loads for the BREST-OD-300 core, the carbothermal process of mixed nitride fuel production has been studied and the major process parameters and subsequent investigation fields for the fuel quality improvement have been identified.

The process of synthesizing nitride fuel of metals has been studied and it has been shown that it is simpler, involves smaller number of operations, does not require complicated equipment, enables the continuity of the process and production of a powder suitable for the immediate manufacturing of fuel columns.

Radiation tests of nitride fuel in the SM-2, MIR, BR-10 and BOR-60 reactors at the heat rating of 350 to 1000 W/cm have demonstrated its high performance, compatibility with structural steels and metal coolants.

The properties of the U-Pu nitride fuel and the existing experience of electrochemical refining and production of metals have been the basis for studying and developing the electrochemical process of its reprocessing in molten haloid salts. The first stage of the studies has been completed and has showed the feasibility of this process.

1. INTRODUCTION

It was in the early 1990s that the integrity of the tasks concerned with highly efficient use of uranium and development of closed fuel cycle processes featuring self-protection properties against plutonium separation and theft and also leading to reduced radiation waste amounts, was shown as the condition of progressing large-scale fast reactor based nuclear power supported with resources /1,2/.

The selection of mononitride fuel out of high-temperature and high-density fuel compositions for fast reactors with intrinsic safety features depends on the unique combination of thermal, service and physical and chemical properties therein /1-5/. The drawback of mononitride fuel containing natural nitrogen is formation of C^{14} and tritium during irradiation. But the contribution of their activities to the total value is small and only requires C^{14} separation and retention during reprocessing. The fraction of tritium generated on nitrogen does not exceed 10% of its yield in mononitride, which largely depends on ternary fission of uranium and plutonium. The C^{14} generation can be further reduced considerably by enriching nitrogen with the N^{15} isotope.

To date, the number of technological studies and radiation tests of mononitride fuel has increased in Russia and abroad. Radiation tests of mononitride fuel at different heat rating and burnup values in EBRII, Phoenix, BR-10 and BOR-60 and other reactors have been successfully conducted or are underway /3-9/. It should be noted that the BR-10 reactor with a mononitride uranium core has been successfully operated for 18 years.

However, the scope of technological research and studies into properties of mononitride fuel is smaller than the scope of similar activities for oxide fuel.

The strategy of developing nuclear power in Russia to the mid-21st century seeks to build reactor and fuel technologies with their safety depending primarily on intrinsic safety features of components and regularities whose economic competitiveness would enable the development of large-scale nuclear power supported with resources.

These conditions, including the proliferation resistance support, are met by experimental and demonstration lead cooled fast reactor BREST-OD-300 under design. The blanket-free mononitride core with the full in-core fuel breeding operates in a closed U-Pu cycle with the actinide inclusion and users equilibrium fuel compozition. This enables the exclusion of weapons-grade plutonium generation during irradiation, plutonium separation and makeup to the reprocessed fuel, which provides for the closeness of the fuel state to the condition of the irradiated fuel at all handling and refabrication stages (the fuel radioactive level is 50-500 Ci/kg). Depleted uranium and plutonium dioxide separated during reprocessing of spent nuclear fuel (SNF) of thermal reactors are used as the initial fuel base.

Upgrading design reactor BN-800 schedules to reach the CBR ≅ 1 and use mononitride mixed fuel.

On-site fuel refabrication excluding transportation of fissionable radioactive materials and spent fuel contributes to the safety increase and is part of a complex of nonproliferation measures. In this connection, it is an urgent task to develop a safe compact dry reprocessing complex without in-depth removal of fission products and with small waste amounts. Among other such processes, electrochemical reprocessing is of a considerable interest.

Therefore, a more in-depth and consistent study of mononitride fuel is an urgent task.

2. STUDYING THE SYNTHESIS AND MANUFACTURING METHODS FOR FUEL PELLETS OF MIXED URANIUM AND PLUTONIUM NITRIDES

A considerable amount of energy-grade (reactor-grade) plutonium dioxide has been accumulated by now. The U-Pu alloy should be used as the parent material after the process of the UPuN fuel electrochemical reprocessing in molten haloid salts is developed. The use of these parent materials for manufacturing of the mixed nitride fuel is an important component of the BREST reactor and closed fuel cycle deployment efforts (Fig. 1) The basis for this are two most advanced flow diagrams:

- production out of the initial oxides by carbothermal method in the nitrogen atmosphere (using energy grade plutonium oxide);
- production out of the U, Pu parent metallic alloys (electrochemical reprocessing products) using their hydration with hydrogen and nitration.

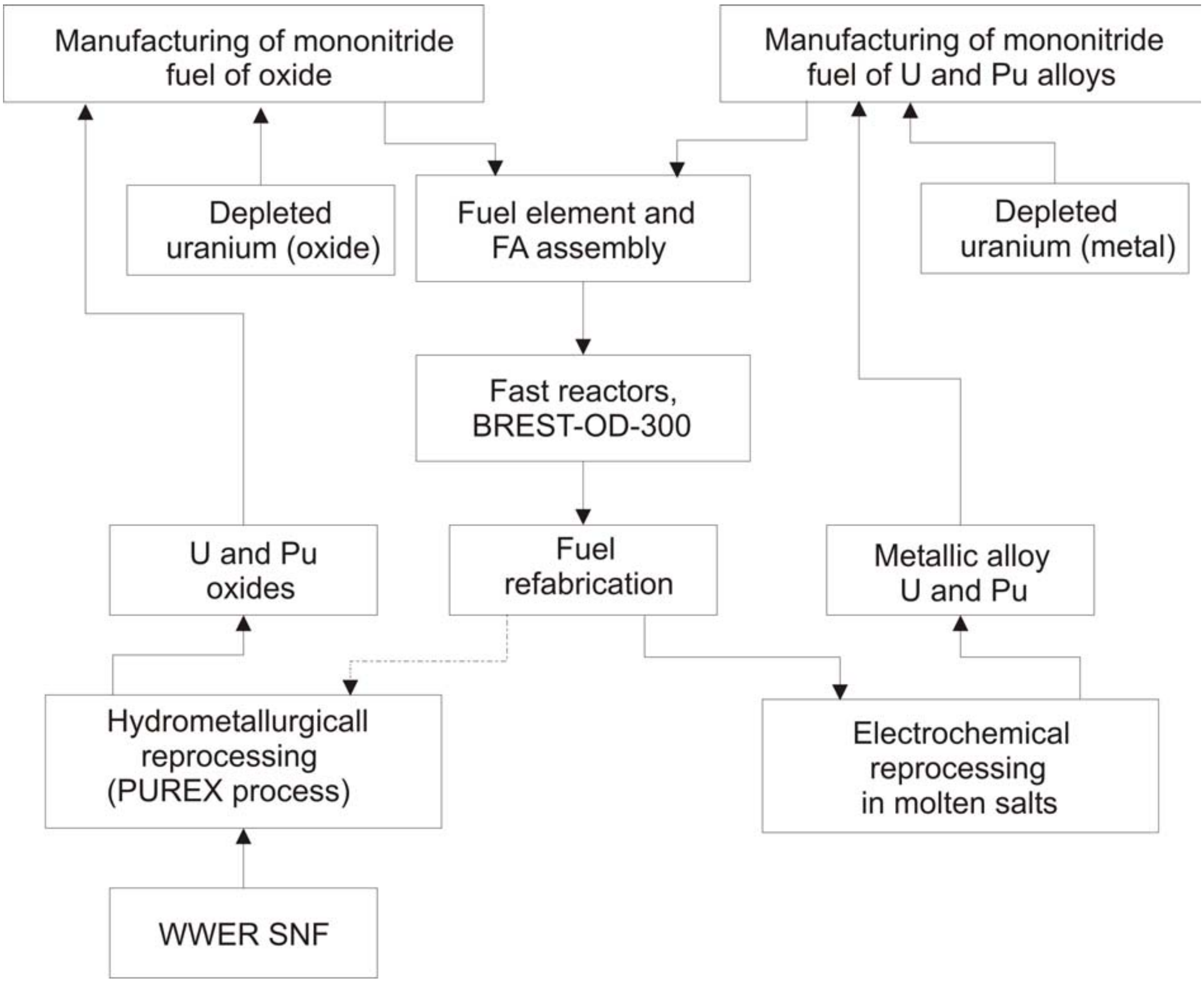

Figure 1 Closed fuel cycle diagram

2.1. STUDYING THE METHOD OF CARBOTHERMAL MIXED MONONITRIDE PRODUCTION

The studies /5, 8/ were conducted in the argon atmosphere using a multi-chamber facility and the adjoining process equipment.

Mixed mononitride with the PuN content of ~14.6% (its isotope composition conforms to the WWER SNF plutonium with the cooling period of 15-20 years) is proposed for use as the BREST-OD-300 reactor starting load.

Therefore, the charge was calculated using the following equation:

$0.854UO_2 + 0.146PuO_2 + 2.04C + 0.16N_2 + 0.162H_2 = U_{0.854}Pu_{0.146}N + 2CO + 0.04CH_4$ (1).

Up to 3% of the excessive carbon was introduced to bond the oxygen from the post-stoichiometric oxide and the oxygen and water vapor absorbed by carbon /5/.

Uranium and plutonium oxides and carbon as black, flakes, chemically pure argon, nitrogen and an argon-hydrogen mixture were used as the parent materials /9/.

The oxygen coefficient of uranium dioxide was in the limits of 2.01-2.05. The charge (oxides and carbon) was mixed in a ball mill for 4 to 5 hours /9/. It has been experimentally found that the mixing in an automatic vortex mixer (AVM) is a more advanced method.

The resultant charge was compacted at the pressure of 1.5 to 2.0 t/cm^2 to form briquettes with the diameter of 6 to 15 mm and the height of 4.0 to 6.0 mm. The density of the compacted charge using different carbon types slightly varied and was equal to around 40–42 % of the theoretical value. The briquetted charge was loaded into a vacuum-compression furnace with a graphite heater and was being gradually heated. Two heat treatment variants were tested:

- vacuum heat up to 1400 °C with the subsequent temperature increase in the nitrogen atmosphere and the holding at 1600–1700 °C for 5–7 hours and the cooling in the Ar atmosphere;
- heatup to 1800 °C in the chemically pure nitrogen flow, the holding at this temperature until to 12 hours and the cooling in argon.

To reduce the carbon content, hydrogen was introduced either in a mixture with argon ($Ar+8\%H_2$) or in a mixture with nitrogen ($N2+6\%H_2$) in the closing third of the holding period at 1600-1800°C.

No marked superiority of any carbidizing agent has been revealed in the studies. The contents of oxygen and carbon were in the limits of 0.2–0.3 % each.

The impact of the carbonitration time was studied at the temperature of 1750°C. It has been found that this temperature reduces the oxygen and carbon contents from 0.5 and 0.63 % to ~0.2 and ~0.3 % respectively as the heat treatment time is increased from 1 to 7 hours.

Introduction of hydrogen to nitrogen at the final stage contributes to a certain reduction in the content of primarily carbon to 0.15% of the mass. The parameter of the $U_{0.87}Pu_{0.13}N$ crystal lattice was 4.891±0.001A°.

Subsequently, it is proposed to determine the impact of the charge mass on the nitride composition and to study the optimization of the UPuN synthesis conditions.

Variant 1 of the mixed mononitride production using charge vacuum preheating at 1400 °C enables better results with a shorter total heat treatment time as compared to variant 2. However, variant 2 enables simplification of the furnace design, development of a continuous process and manufacturing of mixed mononitride on a larger scale.

2.2. STUDYING THE METHOD OF MIXED NITRIDE SYNTHESIS OF PARENT METALS

The mixed nitride synthesis is based on the following reactions:

$2Me(alloy) + 3H_2 = 2MeH_3$

$MeH_3 + ½ (1 + x)N_2 = MeN_{1+x} + 1.5H_2$ (2).

The U-Pu alloy will be synthesized in the fuel cycle (Fig. 1) after electrochemical reprocessing and refining melting. The alloy required for the studies was produced by vacuum remelting of the parent metals at the temperature of ~1200°C for 30 min.

The mixed nitride fuel was synthesized in a horizontal stainless steel apparatus that was heated by a pulled-on electric furnace. The source uranium and plutonium bars were placed on a steel tray that was put into the apparatus. The produced alloy was hydrogenated with purified hydrogen at the temperature of 180–220°C and was then nitrated with chemically pure nitrogen at 220–550°C.

The plutonium content in the parent alloy and a shorter nitration time reduce the amount of the produced uranium sesquinitride. The selected mode ensured the nitrogen content of 6.4–6.7% in the mixed nitride.

The produced mixed nitride is a powder with the particle size of up to 30–40 μm and is suitable for further fabrication of fuel columns.

It should be noted that the hydrogenation and nitration operations are performed in series in one apparatus without recharging of intermediate products. The process was tested on periodic action equipment. The production of uranium nitride by such method was tried on a continuous action facility with the capacity of 1.5-2.0 kg/h /9/. It creates confidence that such UPuN powder synthesis process and equipment are feasible.

2.3. STUDYING THE MANUFACTURING PROCESS OF FUEL PELLETS OF MIXED MONONITRIDE

The mononitride produced of oxides by carbothermal method required additional grinding and fractionation to the grain size of less than 40 μm. The mixed nitride powder synthesized of the parent metals (alloys) was suitable for the immediate fabrication of fuel columns.

Granulated and original UPuN powders were used for the selection of the initial powder grade. It was determined that the compaction in monocavity moulds did not necessarily required preliminary granulation of the powder.

The initial nitride powders were compacted under the pressure of 1-8 t/cm^2. The density of the column blanks produced was 45-55% of the theoretical density (TD). The impact of the blank compaction pressure was largely effective up to 3-4 t/cm^2 [3-7, 8, 20].

The pellets were sintered under vacuum and in the nitrogen atmosphere (Ar+N_2) at the temperature of 1550-1800°C depending on the initial nitride powder grain size and the required density. Fig. 2 shows the microstructure of a mixed mononitride fuel with the density of ~94% TD. The grain microhardness was 650-700 kg/mm^2 [3-7, 8, 20].

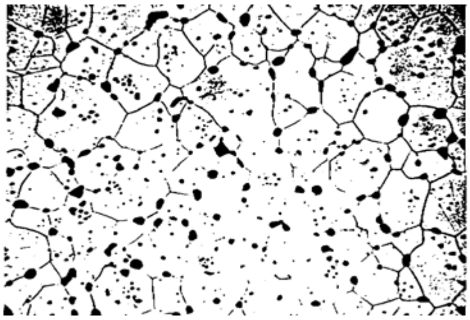

Figure 2 Microstructure of the UPuN fuel pellet

The UPuN pellets fabricated using oxides and metals as the parent materials featured high degree of the plutonium content homogeneity:

Simplified flow diagrams of the UPuN production of initial oxides and alloys and the co

lumn fabrication are presented in Figures 3 and 4.

3. STUDYING THE MONONITRIDE FUEL AND LEAD COMPATIBILITY WITH CLADDING MATERIALS

The compatibility of certain compositions under isothermal conditions was studied:

- mononitride fuel ($U_{0.9-0.85}Pu_{0.1-0.15}N$) – EP-823 and EP-450 steels;
- mononitride fuel ($U_{0.9-0.85}Pu_{0.1-0.15}N$) – lead - EP-823 steel.

The fuel and steel compatibility was studied using lead-filled container assemblies with fuel pellets and steel disks.

It was found as the result of the studies that the mononitride fuel with the above composition interacts neither with lead nor with the cladding steels EP-823 and EP-450 at the temperature of 650 and 800°C for up to 2000 hours (the measurement time) and at the temperature of 1200 and 1300°C (chosen for the simulation of emergency situations) for 5 hours. It has not also revealed any changes in the fuel surface layers washed by lead .

4. RADIATION STUDIES OF MONONITRIDE FUEL

Pallets with the oxygen and carbon content in the range of less than 0.1% to 0.4-0.5% each were fabricated of initial metals and oxides to study the radiation properties of the mononitride fuel.

Radiation tests of fuel elements with columns of uranium mononitride and mixed uranium and plutonium mononitride (with the density of 83-92% TD) produced of oxides and metals, were carried out in the SM-2, MIR, BR-10 and BOR-60 reactors at the linear rating of 350 to 1045 W/cm and the burnup of up to ~9% of heavy atoms (h.a.). Austenitic steel was mainly used as the cladding material. All fuel elements have remained leaktight. It should be noted that the lifetime of the initial BR-10 fuel load was some 5.5 years.

The fuel swelling and the yield of gaseous fission products (FP) depend on the temperature in the column center and the O and C contents in the fuel. The reduction in the oxygen and carbon contents from 0.4-0.5% to less than 0.15% each leads to the swelling decrease from 2.0-2.5% to 1.1-1.5% and in the gas release from 50-55% to 20-25% at the burnup of 9.0% h.a.

No carbonization of the internal cladding surface is observed in fuel elements with a helium sublayer at the oxygen and carbon concentration in the fuel of less than 0.15% each. A rise in the oxygen and carbon concentrations in the fuel by over 0.3% reveals (more etched) interaction areas on the internal fuel element cladding and the further growth in the impurities concentration leads to larger interaction areas and lower cladding ductility.

Mononitride fuel is efficient in retaining fission products, including iodine, cesium, selenium and tellurium. No examples of fuel element failures due to internal cladding corrosion are given in the published papers known to us.

After irradiation in the BOR-60 reactor at the linear rating of 1000-1045 W/cm, there were no changes in the plutonium concentration from the center to the periphery in the columns of mixed uranium and plutonium mononitride. No iodine or cesium corrosion of the cladding was found.

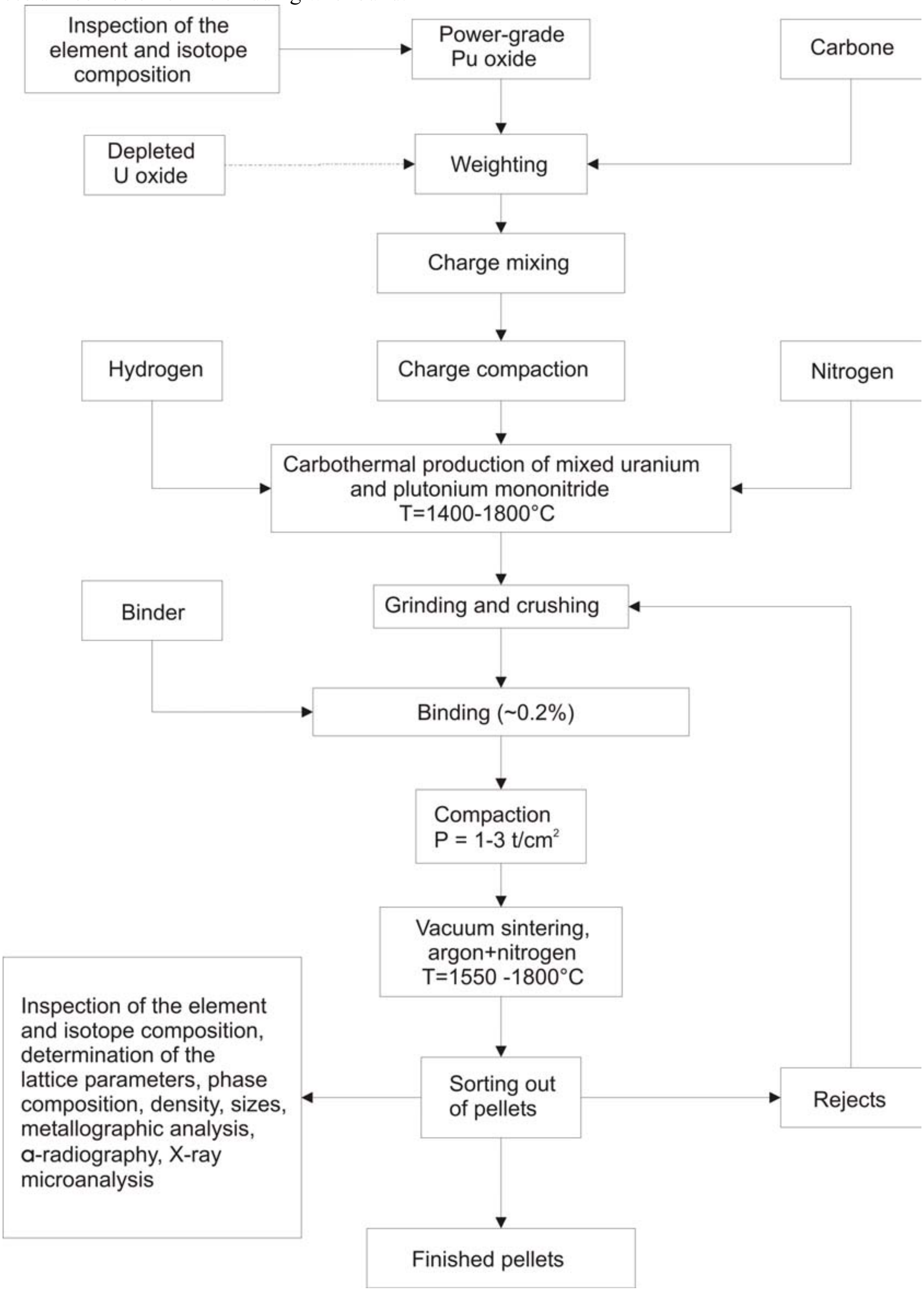

Figure 3 Flow diagram of the mixed mononitride fabrication out of U and Pu oxides for the initial load of the BREST-300 reactor fuel

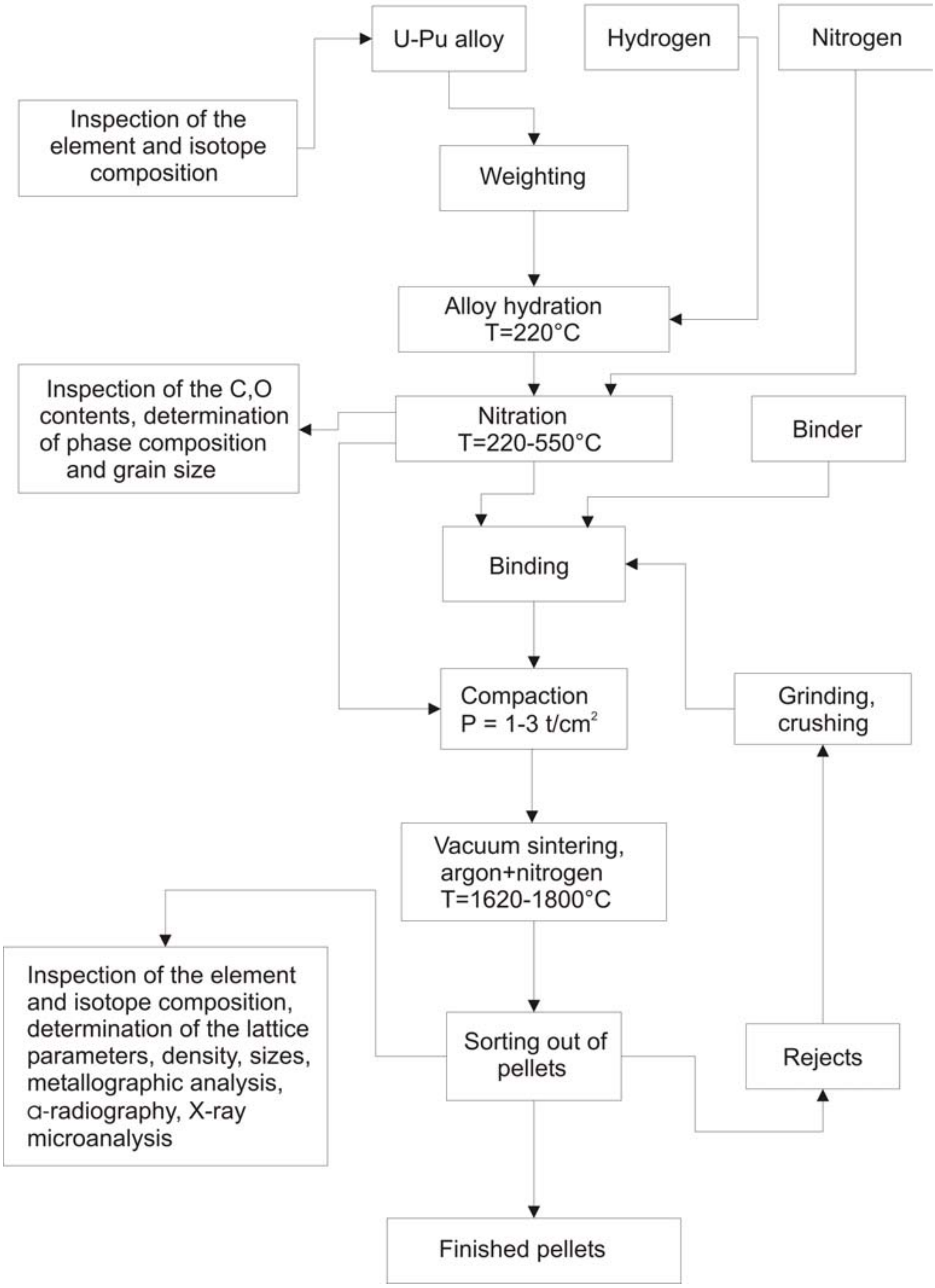

Figure 4 Flow diagram of the mixed mononitride fabrication out of U and Pu alloy

5. REPROCESSING OF MONONITRIDE FUEL

Reprocessing by water-chemical and pyrotechnic methods /7, 10-20/ has been studied and developed in many countries for more than 20 years.

The results of studying non-irradiated mononitride fuel water-chemical reprocessing (PUREX process) have shown that nitric acid dissolves mononitride fuel (UN, PuN) steadily, at a higher rate and more fully than oxide fuel as the plutonium and uranium quantity extraction is ensured /7, 19/. To implement such process (naturally after checks on irradiated fuel), hardware and process production lines for the oxide fuel reprocessing, e.g. of the RT-1 plant, can be used.

Physical-chemical and electrochemical properties of uranium and plutonium mononitrides and the existing experimental experience of refining metals and oxides /5, 7, 11, 13, 19, 21, 23/ have been the basis for developing a pyroelectrochemical mononitride fuel reprocessing. The major reasons in favor of the mononitride fuel electrochemical reprocessing in molten salts are as follows:

- nuclear, radiation, fire and explosion safety assurance;
- no necessity of long-term irradiated fuel cooling;
- the irradiated mononitride fuel consists of conducting nitrides, whose generation energy is less than the generation energy of their haloid salts; this should facilitate the anode dissolution of nitrides;
- electrochemical reprocessing in melts features the compactness of the hardware used and the possibility of the electrolyte multiple reuse after its periodic cleaning of fission products; the produced wastes are solid;
- the possibility of FP components with close separation potential values being fractionally separated;
- it is possible to run the electrochemical reprocessing in electrolyzers with solid and liquid cathodes;
- the possibility of building the process of spent nuclear fuel reprocessing in the on-site nuclear fuel cycle.

The process is studied largely in Russia (RIIM, Research Institute for Atomic Reactor) and Japan (JAERI). Investigations are underway to study several reprocessing options with potential end products as uranium and plutonium alloys or nitrides. The first stage of the studies is underway to:

- select the modes of non-irradiated mononitride fuel dissolution and joint uranium and plutonium deposition on the cathode;
- select the equipment design;
- analyze the possibility of uranium and plutonium nitrides being produced as the reprocessing end products /24/.

The parameters of the electrolysis process as recommended based on the RIIM studies are summarized in Table 1.

Table 1 The parameters of the electrolysis process

Parameter	Parameter value
Temperature, °C	550÷600
Current density anode, A/cm^2 cathode, A/cm^2	 0.05÷0.1 0.3÷0.6
Atmosphere over the electrolyte	Argon
Reprocessing rate, g/cm^2h	0.45÷0.6

Special experiments have confirmed the feasibility of the complete joint uranium and plutonium deposition on a solid cathode at the UCl_3 and $PuCl_3$ content in the melt of ~11% and ~2% of the mass respectively /25/.

Several consecutive cathode deposition cycles have been implemented with the result as the uranium content reduction in the electrolyte to 0.04% of the mass and the plutonium content in the fuel to 0.004 % of the mass. The uranium and plutonium relation in the cathode deposit was equal to their relation in the initial electrolyte.

The electrolyte is multiply reused for fuel reprocessing. After 10-15% of FP is accumulated, the electrolyte is cleaned and its composition is adjusted.

It has been shown that the removal of fission product simulators at the uranium cathode deposition took place at the Cs, Sr and La content of ~4%, 2.3% and 5.5% of the mass respectively, which is in conformity with the literature data evidencing that uranium can be cleaned of RZE when their concentration is not more than 3.6÷5.6% of the mass.

The advantage of the electrochemical reprocessing process is that there is no additional introduction of disposable reagents to the basic process except multiply reused electrolyte components, which excludes additional generation of wastes.

The positive results of studying the mononitride fuel electrochemical reprocessing process stimulate their continuation.

6. CONCLUSIONS

The currently available results of studying the thermochemical, thermal, mechanical, radiation and technological properties of mononitride uranium and mixed fuel have enabled the initiation of activities to design fast reactors with mononitride fuel.

1. The studies on the synthesis of mixed U-Pu mononitride fuel of initial oxides and metals have demonstrated that it is a promising process and it is possible to achieve low oxygen and carbon contents (less than 0.2% for fuel of oxides and 0.1% for fuel of metals). It has been shown that columns with the density of 92-94% TD can be fabricated by compaction and sintering at the temperature of 1600-1800°C.

2. The feasibility of a compact pyroelectrochemical process of nitride fuel reprocessing in molten haloid salts has been shown.

3. Studies of the fuel column synthesis, manufacturing and reprocessing should be continued to optimize respective modes.

4. To confirm the functionality of the fuel element with nitride fuel and build a model of the fuel element and fuel behavior in the lead cooled fast reactor under design, radiation tests of the FA mockups are required.

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