Using an Induction Melter With a Cold Crucible for the Immobilization of Plutonium

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Significant reductions in defense programs and the dismantling of large numbers of nuclear weapons have placed on today's agenda questions of safe handling, utilization and burial of weapons-grade plutonium. The optimum approaches to the management of surplus plutonium stocks in the United States and Russia may differ because of the differences in policies of the two countries on the plutonium fuel cycle, the infrastructure of reactors and the reprocessing of radioactive waste.

There are two approaches to this issue that are closest to satisfying safety criteria for handling of surplus plutonium:

- utilization of the plutonium as fuel in existing or modified reactors
- vitrification of plutonium along with high level waste.

This report looks at the possibilities for immobilizing weapons-grade plutonium in glass-type materials that satisfy requirements for eventual burial in deep geological formations and correspond to the standards set for spent fuel.

The proposed plan for the reprocessing of weapons-grade plutonium (Figure 1) and its immobilization in glass-type materials may be regarded as a vitrification operation for separated high level waste, produced during the regeneration of spent fuel.

High level waste separation calls for the extraction of the cesium and strontium fraction (total concentration 12 g/l), the transplutonium elements and rare earth fraction (maximum concentrations for transplutonium elements - 2.5 g/l; for rare earth elements - 26 g/l) and the residual solution that contains the rest of the radionuclides and the equipment corrosion products.

The process of vitrifying plutonium with the necessary admixtures so that it satisfies the standards set for spent fuel and provides for a decrease in criticality, will be, to a great extent, quite similar to the vitrification process for the transplutonium and rare earth fractions.

After warheads are dismantled the fissile material must be ground up and oxidized producing a chemically highly active plutonium dioxide with the necessary physio-chemical properties that will simplify its further reprocessing. We all know that the dissolution of metallic plutonium in mineral acids poses significant problems and as a rule the plutonium is never completely dissolved. If hydrochloric or hydrofluoric acid is used to achieve greater dissolution of plutonium, problems are created further down the line in the reprocessing because of the presence of Cl and F ions in the solution. However, we can significantly reduce the volume of secondary waste products containing plutonium (hard to dissolve residues formed when metallic plutonium is dissolved) from plutonium reprocessing if we use the chemically active plutonium dioxide (dissolution in nitric acids, direct solidification of the dioxide as it fuses with fluxing admixtures).

At the next stage we can consider two options as further steps in the handling of the plutonium dioxide.

The first option calls for "wet processing" and requires at the initial stage the preparation of a solution of plutonium dioxide in nitric acid. This is necessary if technology used for the vitrification of high level liquid waste is applied to plutonium. The choice of this option means that the vitrification process will produce glass containing plutonium together with fission products (prior to vitrification the solution with plutonium is mixed with high level liquid waste from the reprocessing of spent fuel). Fluxing admixtures are added at the high temperature synthesis stage to satisfy specifications for the final product and to produce materials with the desired properties.

The second option is the "dry processing" of plutonium, which makes it possible to avoid the dissolution step and, on the one hand, simplifies the technological process, while, on the other hand, makes possible the production of either an intermediate material with both plutonium and high concentrations of fissile materials (for later synthesis with the proper admixtures into a glass-type composite material) or the direct production of a vitrified product.

The molten glass products are poured into containers of the proper shape and after decontamination are transferred to interim storage facilities or to burial sites.

The proposed approach for the reprocessing of weapons-grade plutonium makes it possible to use the vitrification technology to produce vitrified materials with a plutonium concentration up to 20%.

The proposed technology for the vitrification of weapons-grade plutonium could be based on the two-stage solidification process used in the reprocessing of high level waste that uses an induction melter with a cold crucible (IMCC). The melting process in the IMCC is based on the ability of high frequency electromagnetic fields to penetrate the whole thickness of the material and to release their energy as they subside. The special feature of IMCC is that it allows synthesis of different types of material with a broad range of compositions. It becomes possible to apply the IMCC melting technology for the purpose of solidifying waste from various technologies used in the reprocessing of spent fuel, including waste produced as a result of the separation of high level waste, as well as toxic waste, containing heavy metals.

Most of the work in the development of the IMCC technology was connected to the solidification of high level liquid waste at Mayak. Most of our experience was with the vitrification of simulated high level waste that was immobilized in phosphate and borosilicate glass.

Composition 1

Na_2 o	-	5.0
Cs_2O	-	5.3
K_2O	-	3.7
NiO	-	0.2
CaO	-	0.3
Fe_2O_3	-	1.3
Cr_2O_3	-	0.3
MnO_2	-	0.5
SrO	-	2.1
BaO	-	2.2
ZrO_2	-	6.8
MoO_3	-	4.5
RuO_2	-	4.0
Rh_2O_3	-	1.7
PdO	-	1.7
oxides		
REE	-	16.7

Composition 2

Al_2O_3	-	41.9
Na_2O	-	3.1
NiO	-	0.1
Fe_2O_3	-	1.2
Cr_2O_3	-	0.3
oxides		
REE	-	0.2

Composition 3

Al_2O_3	-	18.9
Na_2O	-	54.0
CsO	-	0.5
NiO	-	7.6
CaO	-	5.6
Fe O	-	14.3
Cr O	-	3.0
MnO	-	3.2
SrO	-	0.4

Composition 4

Al O	-	19.0
Na O	-	67.4
NiO	-	6.3
Fe O	-	7.0
Cr o	-	3.0

Composition 5

Cs_2O	-	9.2
SrO	-	4.5
BaO	-	4.8
PbO	-	1.1

Composition 6

Oxides		
Fe, Cr, Ni		64.3

Composition 7

Oxides		
REE	-	30.3
ZrO	-	8.5

We also applied the IMCC technology and studied some of the physio-chemical properties of pyroxene type mineral matrices (aegirine, jadeite, aegirin-augite, orthite and others) and of orthosilicates (andradite, sphene, lovchorrite and others). The total concentrations of oxides and other waste components that were incorporated onto these materials were equal to 15% of the mass: Cs_2O - 5.0; SrO - 3.0; CeO_2 - 5.0; Fe_2O_3 - 2.0.

Based on the results of a series of studies of solidification technologies for various simulated high level wastes, a pilot facility was created at the Mayak vitrification site with a two-stage process for the immobilization of high level waste on phosphate and borosilicate glass as well mineral type materials.

The solidification process for liquid high level waste at the pilot facility includes the following steps:

- selection of the composition of liquid high level waste and their fluxing with orthophosphoric acid for the production of phosphate glass of a given composition;
- processing of the initial simulated high level waste in a concurrent flow evaporator in order to produce a highly concentrated solution;
- vitrification of the concentrated salty melt;
- drainage of the melted glass into cans;
- placement of cans in cases;
- decontamination from gas releases.

The fluxed solution of high level waste (Figure 2 and 3) continuously enters the concurrent flow evaporator at a rate of 100 dm^3/hr. The evaporator, heated by steam at 0.5 MPa, is co-located with the separator. Monitoring of the concentration process is done through the measurement of pressure at the point the solution enters into the evaporator (EV), and the pressure of the steam used for heating, by flow-meters, and through the measurement of the salt concentrations in the initial solution.

The concurrent flow evaporator is a vertical assembly with the main working section in the form of a spiral enclosed in a steam jacket. The solution enters in countercurrent to the steam. The stationary zone for the separation of the vapor-gas phase and the salt melt is the interior shell of the steam jacket - the separator.

The processes in the concurrent flow evaporator develop in the following order as the material progresses though the system: heating, evaporation and the removal of nitric acid, partial interaction of the nitrates with the orthophosphoric acid, and drying. After the separation of the vapor-gas phase and the melt in the separator unit, the melt flows by gravity into the IMCC at the rate of 30 - 40 kg/hr. The vapor-gas phase enters the first purification stage - the sparger-cooler.

Dehydration and denitration occur in the IMCC at 1100 - 1200^{0}C with the production of phosphate glass at the rate of 15 - 18 kg/hr.

The IMCC is a rigid construction composed of cooling pipes mounted around a water collector. The crucible is surrounded by the water-cooled inductor that is connected to a high frequency generator (Figure 4).

The molten product is discharged from the crucible into 200 dm^3 containers placed on a circular conveyer belt.

Scales will be used to monitor the filling operation.

During the normal mode of operation of the IMCC, the crucible will continuously discharge the melt into containers. A special device will interrupt the drainage operation until a new container is in place.

Start up of the IMCC requires initial heating, which is achieved through the interaction between the high frequency field and the conductive material brought into the crucible. The start-up operation can be accomplished by a mixture of materials or broken glass (initial), or with glass that has hardened in the crucible (secondary).

The filled containers are cooled and placed in cases, which are sealed by welding and, after the seals are tested, are transferred to storage.

The first step of the gas purification process during the vapor-gas phase separation stage is the sparger-cooler, where the vapors are condensed, the condensate is cooled, and the first phase of aerosol capture takes place.

When tests are conducted at the EV-IMCC facility with simulated solutions, the second gas purification step is an adsorption column for the capture of nitric oxide, after which the decontaminated gas is released into the atmosphere.

During the vitrification of actual high level liquid waste in the EV-IMCC, the gas purification system of the direct heating electrical furnace will be used. The radionuclide removal coefficient for the gas purification system is 10^8 - 10^9.

A closed cooling system with demineralized water or condensate with a specific resistance below 20,000 Om*cm is used for cooling both the IMCC and the high frequency generator in order to provide for accident-free operations.

The two-stage pilot facility is located in a hot chamber and is being used for the following tasks:

- perfecting solidification technology for high level waste on the basis of simulated and actual waste products;
- perfecting the functioning of connectors and mechanical assemblies for operation in a remote mode;
- perfecting transfer technology for the removal of containers with solidified waste and other equipment.

The dimensions of the chamber are given in mm:

Length	8750
Height	4100
Width	2000

The chamber where the EV-IMCC is located has a space for a conveyer for containers with the following dimension:

Length	2000
Height	1200
Width	2000

The technical parameters of the simulated HLW solidification facility are:

- throughput volume of initial solution, l/hr 100
- temperature of the glass melt entering the containers, ^{0}C 1200

Concurrent flow evaporator:

- throughput volume of initial solution of simulated HLW 100
- steam consumption, kg/hr 120
- steam temperature at evaporator inlet, ^{0}C 150
- operating pressure at evaporator inlet, MPa 0.4-0.5
- dimensions, m
  diameter 0.55
  height 1.2

Cold crucible (a two-zone crucible):

- nominal capacity of the tank 22.6
- melting mirror area, dm^2
  boiling zone 6.7
  processing zone 1.8
- maximum operating temperature, ^{0}C 1500
- glass throughput by mass, kg/hr up to 18
- oscillating power of the high frequency generator, kW 160
- power consumption, kW 240
- operating frequency, Mhz 1.76
- inductor cooling water consumption, m^3/hr 2
- crucible water consumption, m^3/hr 4
- cooling water temperature, ^{0}C
  inlet
  outlet 25±5
- cooling water pressure, MPa 45±5

0.4

Sparger-cooler:	210
• vapor-gas mixture consumption, m^3/hr	
• vapor-gas mixture temperature, ^{0}C	
inlet	300-600
outlet	55
• cooling water flow rate, m^3/hr	25
• cooling water temperature, ^{0}C	
inlet	25
outlet	30
• cooling water pressure, Mpa	0.4
Circular conveyer:	
• load-carrying capacity, N	$1.2*10^3$
• number of slots for containers, pc	2
• angle of swing, degrees	180
Weighing equipment:	
• weight of container filled with glass, kg	600
• the weighing device is an electronic tensometer	
The phosphate glass product is placed in containers with the following parameters:	
• diameter, mm	575
• height	1000
• material	steel #3
Weight of empty container, kg	95±10
Volume of container, dm^3	
• total	220
• useable capacity	200
Volume of glass mixture poured into container, dm^3	190±10

The pilot two-stage vitrification facility for high level waste will be the prototype for the industrial scale technologies of that type to be installed at the second production line at Mayak (Figure 5 and 6).

The unique features of the melting process in the IMCC make it possible to use this technology for the reprocessing of liquid radioactive waste and toxic waste.

Induction melting in a cold crucible offers the following unique possibilities not available with melters of other types:

- the possibility to vary melting temperatures within a wide range of both temperatures and solidification mixtures from glass to mineral-type;

- high intensity of convective mixing of the melt making it possible to significantly increase the throughput capacity without preliminary mixing of the waste components with the fluxing admixtures;
- the possibility to provide for remote installation and dismantling of the equipment, as well as its removal for storage and reprocessing in accordance with provisions for the transportation of vitrified waste;
- a layer of slag is formed on the water-cooled walls of the crucible during the melting process, thereby protecting the crucible from the corrosive action of the melt products;
- the fact that the parameters of the generator are dependent on the condition in the tank with the melt makes it possible to automate the process using indicators from standard measurements from the generator instruments;
- the capability to stop and start the melter if any problems should rise in any part of the process.

The limitations and deficiencies of the IMCC process also need to be addressed. They are as follows:

- the complexity of the manufacture of the power equipment, which decreases the reliability of melters as a whole;
- the complexity of the equipment itself mandated by the requirements of the induction heating process;
- because of the small dimension of the crucible, although the specific melting capacity is high, the output of glass is measured in kilograms per hour;
- low positive thermal coefficient (0.2 - 0.35) and its dependence on the thermal and physical properties of the processed mixture;
- the need for specially formulated water for the cooling of the generator and crucible.

However, the obvious advantages of the IMCC technology in comparison to its disadvantages made this process quite attractive for the vitrification of weapons-grade plutonium.

DISCLAIMER

The views expressed in this paper are those of the author(s) and do not necessarily reflect any biases, proposed actions, or decisions of the United States Government or any agency thereof.

Equipment list (Figures 2 and 3):

1. Concurrent flow evaporator
3. Cold crucible
5. Circular conveyer
7. Mechanism for closing the tops of the cases
8. Case with containers
10. Bin for fluxing admixtures
11. Sparger-cooled

2. Flux tube
4. Container with glass melt
6. Weighing equipment
8. Crane
9. Welding equipment for sealing the top of the case

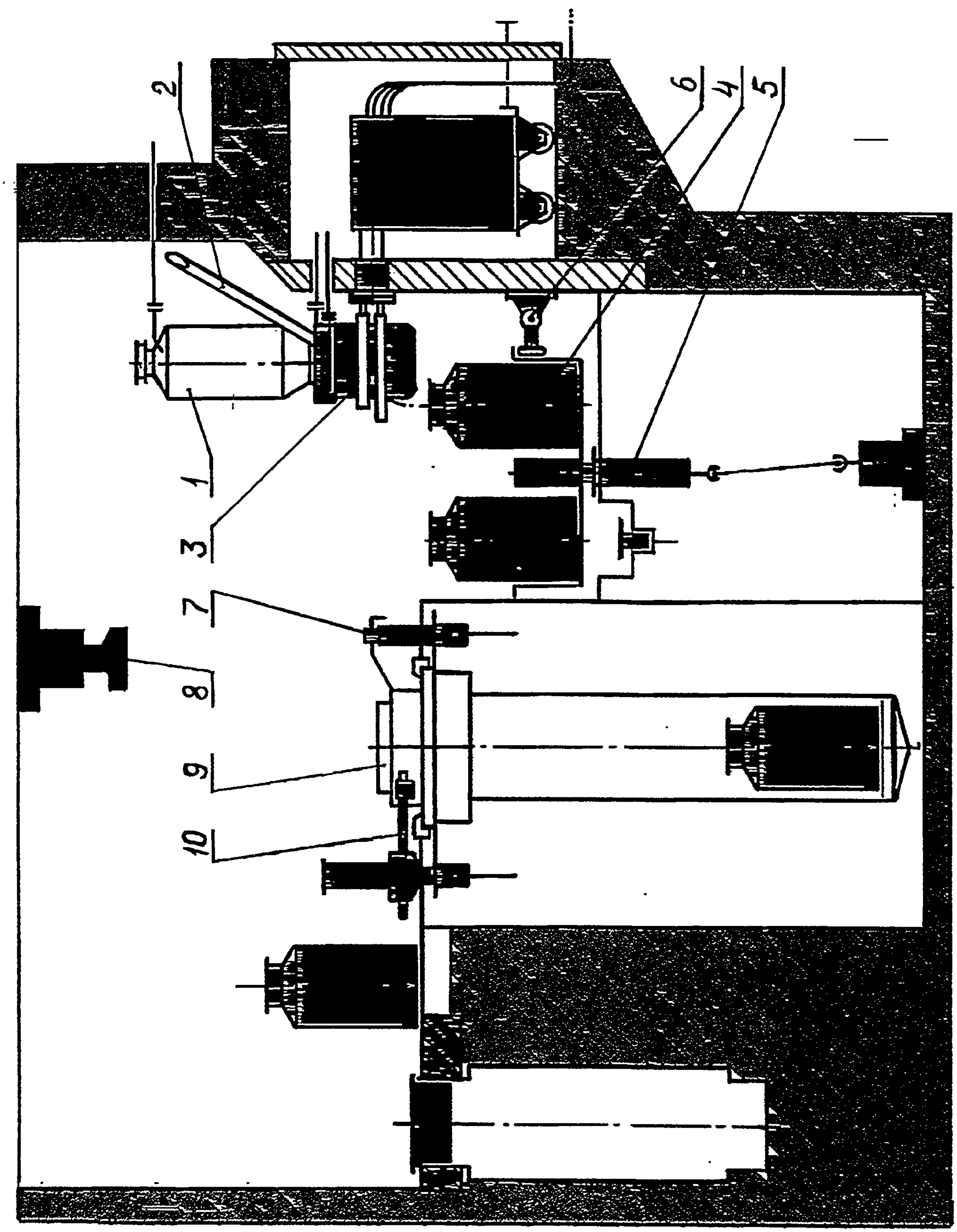


Рис. 2

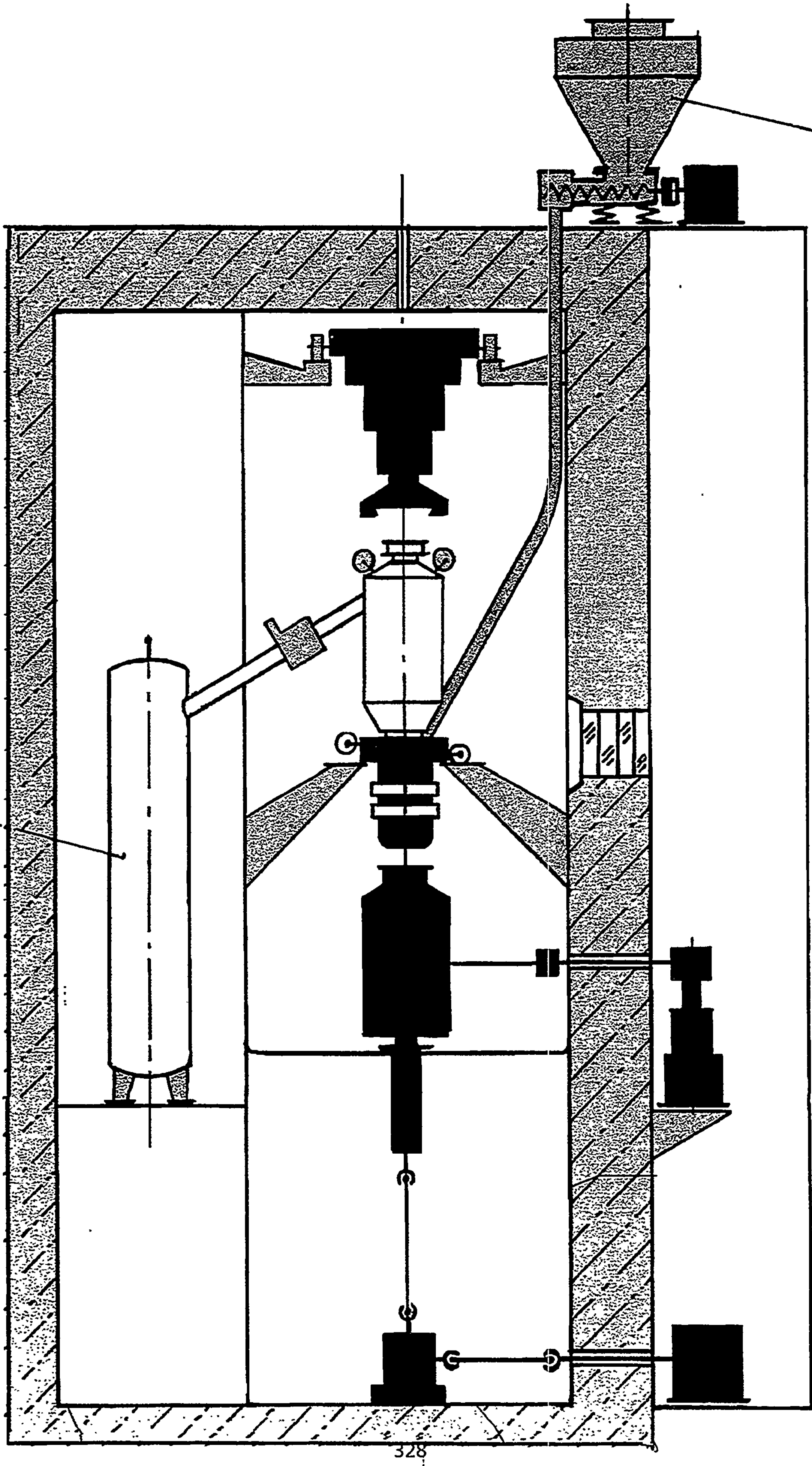


Рис. 3

INDUCTION MELTER WITH A COLD CRUCIBLE (IMCC)

1. Fusible connector
2. Pipe section cooling water
3. Inductor
4. Melter inlet for material
5. Pipe section of crucible
6. Melt
7. Partition
8. Outlet for melt

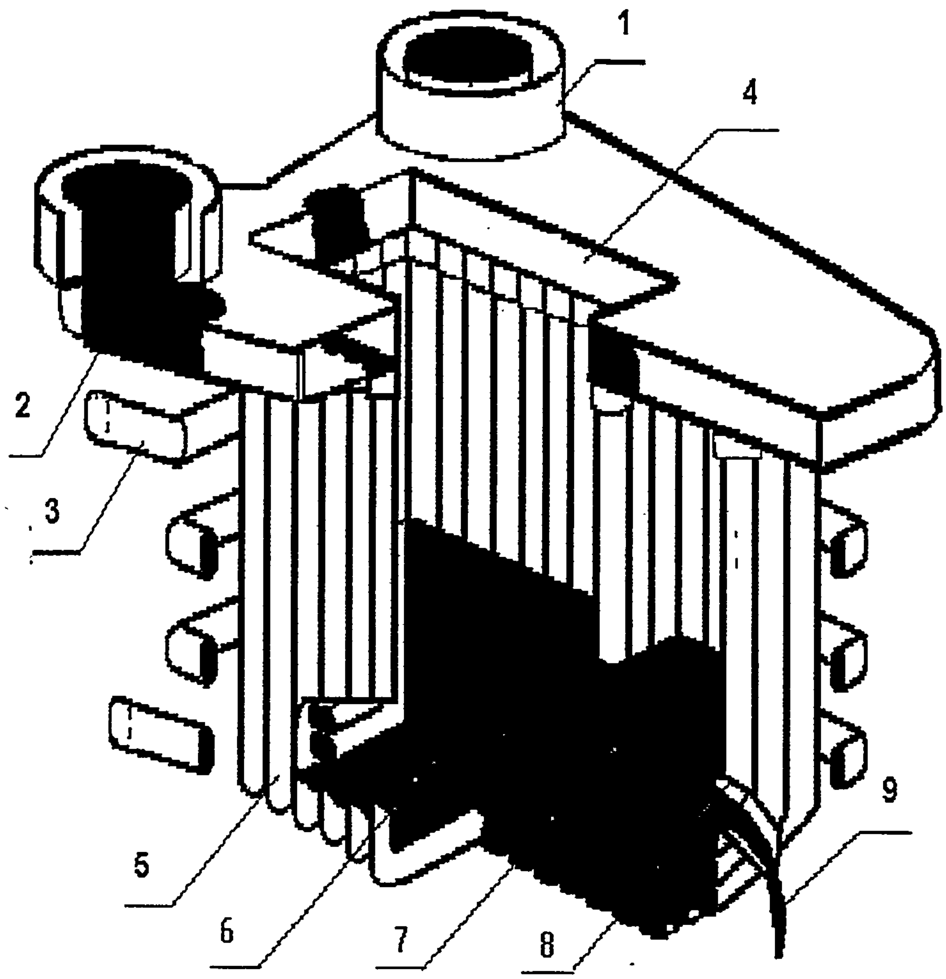
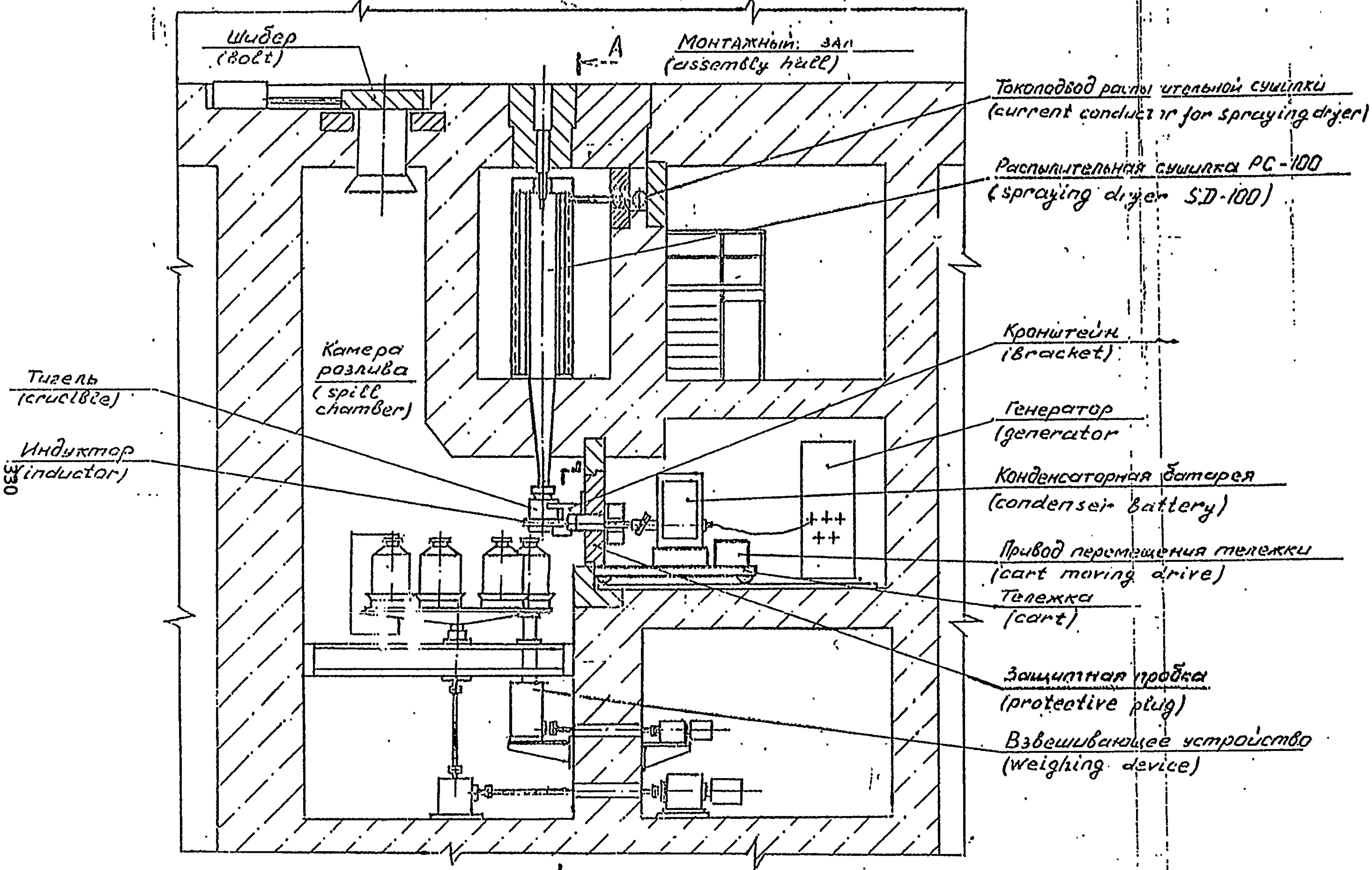


Рис. 4



Двухстадийная установка РС-ИПХТ
(Double-stage SD-IMCC installation)

Рис.5

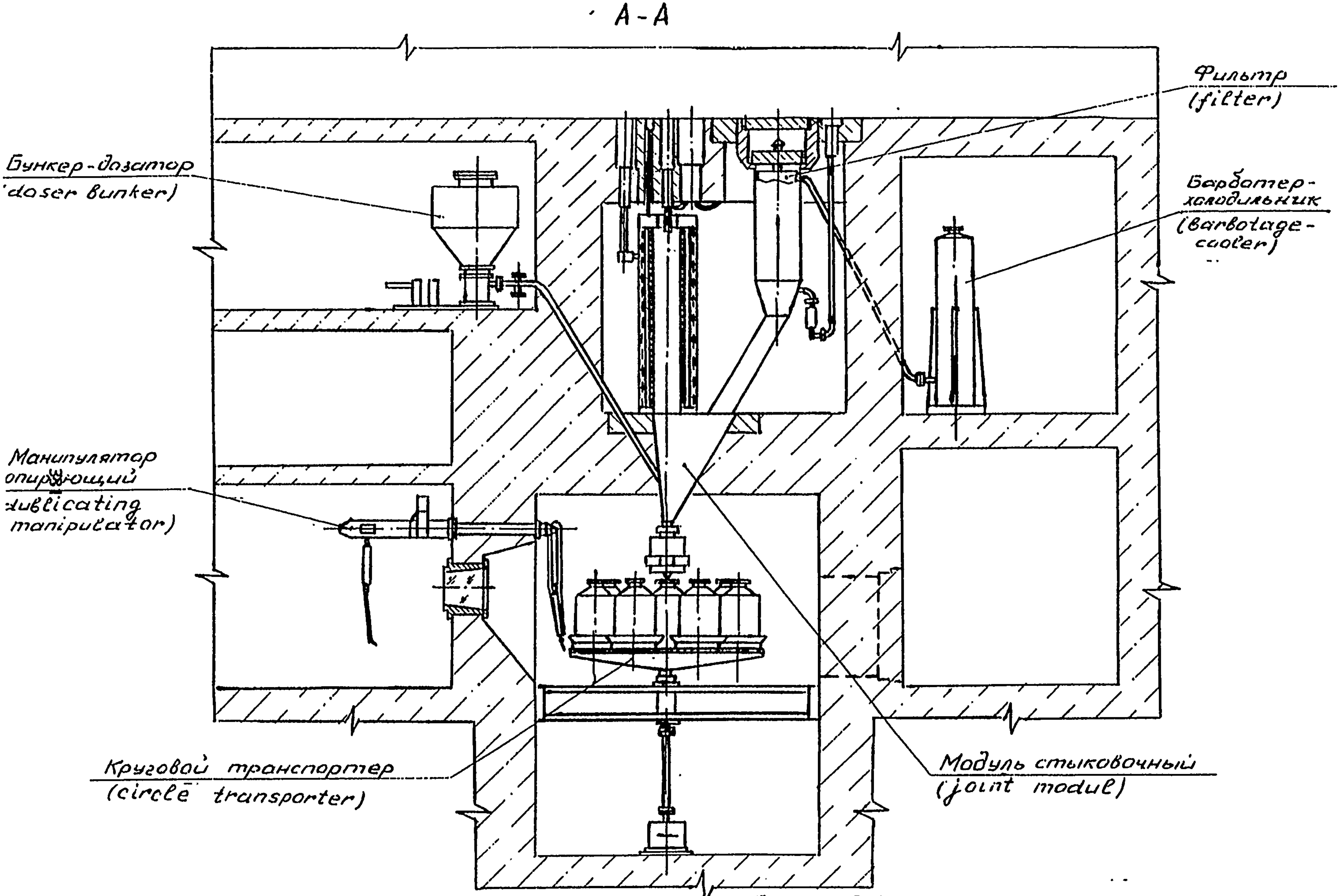


Рис. 6

Двухстадийная установка РС-ИПХТ
(Double-stage SD-IMCC installation)