# HIGH TEMPERATURE TREATMENT OF INTERMEDIATE-LEVEL RADIOACTIVE WASTES – SIA RADON EXPERIENCE

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## ABSTRACT

This review describes high temperature methods of low- and intermediate-level radioactive waste (LILW) treatment currently used at SIA Radon. Solid and liquid organic and mixed organic and inorganic wastes are subjected to plasma heating in a shaft furnace with formation of stable leach resistant slag suitable for disposal in near-surface repositories. Liquid inorganic radioactive waste is vitrified in a cold crucible based plant with borosilicate glass productivity up to 75 kg/h. Radioactive silts from settlers are heat-treated at 500-700 $^0$C in electric furnace forming cake following by cake crushing, charging into 200 L barrels and soaking with cement grout. Various thermochemical technologies for decontamination of metallic, asphalt, and concrete surfaces, treatment of organic wastes (spent ion-exchange resins, polymers, medical and biological wastes), batch vitrification of incinerator ashes, calcines, spent inorganic sorbents, contaminated soil, treatment of carbon containing $^{14}$C nuclide, reactor graphite, lubricants have been developed and implemented.

## INTRODUCTION

State Unitary Enterprise Scientific & Industrial Association "Radon" (SIA Radon) is a company involved in the system of the Administration of Moscow and responsible for management of radioactive wastes from non-nuclear applications. Moreover, SIA Radon collaborates with Russian Nuclear Power Plants and other institutions and plants of the Ministry of Atomic Energy of Russia (Minatom). So, SIA Radon deals with collection, transportation, treatment, conditioning, and disposal or long-term storage of conditioned LILW, develops technologies for treatment of wastes from both non-nuclear and nuclear applications including waste from nuclear fuel cycle in cooperation with Minatom institutions.

SIA Radon processes both solid and liquid organic, inorganic and mixed wastes and has forty-years experience in this area. Existing technologies are permanently updated and new high effective processes are developed. Previous experience was reviewed in our papers [1-4]. This paper summarizes recent advantages of SIA Radon in development and implementation of high temperature technologies for LILW treatment.

## PLASMA TECHNOLOGIES FOR SOLID WASTE TREATMENT

The most efficient method of solid and liquid burnable low- and intermediate level wastes volume reduction is considered to be an incineration. However, incinerator ash is powdered dusting and easily leachable material, which is not suitable for final disposal and requires an additional conditioning. Several methods of high-temperature treatment of incinerator ash were developed at SIA Radon. One of them is melting of incinerator ash using a plasma torch. The method is realized as follows. Incinerator ash is crushed, intermixed with fluxing additives and fed into inclinable melter heated by two arc plasmatrones to temperature of ~1600 $^{0}$C. Molten slag is periodically poured into containers. Solidified slag is solid monolithic, and high chemically durable and suitable for disposal in near-surface repositories.

The other technology avoids incinerator ash production. A plasma-heated shaft furnace based plant with liquid slagging (Figure 1) was developed, designed, constructed, tested, and it is under operation now. This plant (Figure 2) is meant for joint treatment of solid organic waste and spent heat-insulators, concrete, glass breakage, construction garbage, and other high-fusible materials. Waste packages are charged into the shaft until filling and this level is kept constant during operation. The shaft is heated by arc plasmatrones. Waste in the shaft is subjected step-by-step to drying, gasification, burning, slag formation and melting. Molten slag is accumulated and homogenized in the bottom part of the shaft, and poured through stopper unit into containers. Off-gas is purified in an off-gas system, which involves high-temperature post-combustor, chemical and catalytic toxic gases neutralization units, and two-step radioactive aerosols trapping unit.

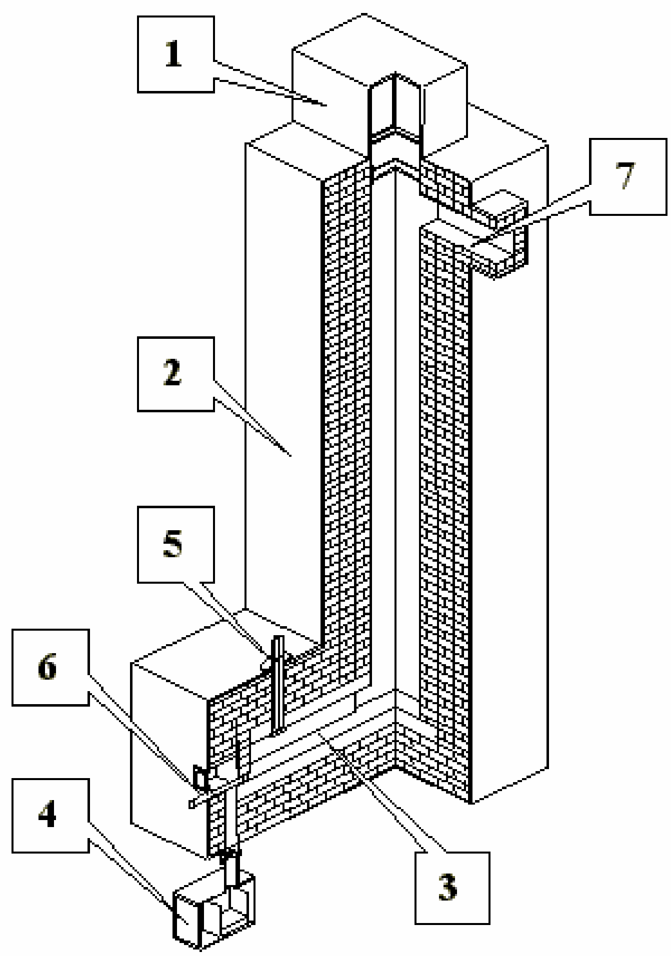


Figure 1. Shaft furnace.

1 – waste charging unit, 2 – shaft, 3 – bottom, 4 – slag collector, 5 – plasmatrone, 6 – gate, 7 – off-gas pipe.

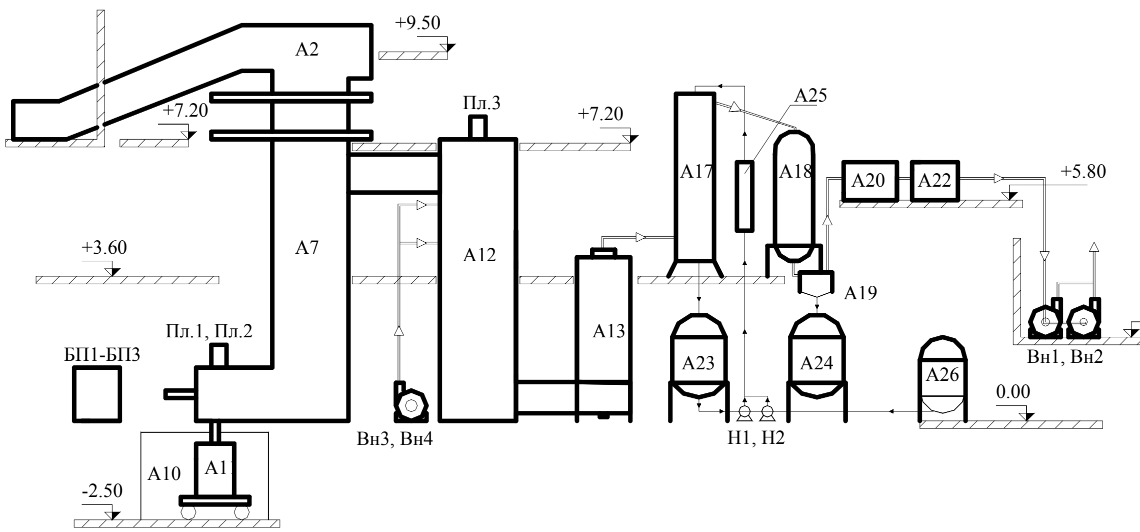


Figure 2. Plasma treatment unit flow sheet.

A2 – waste charging unit, A7 – plasma furnace, A10 – slag collector, A12 – post-combustion chamber, A13 – evaporating heat-exchanger, A18, A25 – heat-exchangers, A20, A22 – filters, A23, A24, A26 – vessels, Pl1, Pl2 – plasmatrones, Bn1, Bn2 – ventilators, H1, H2 – pumps.

The main advantages of the shaft furnace are possibility to treat unsorted waste - high temperature furnace is able to process waste containing 20-40% of non-burnable constituents (glass, metal, constructing garbage, heat-insulators) and formation of conditioned final product in the same plant without interim steps. Currently the “Pyrolis” plant with waste capacity 40-50 kg/h is under operation and full-scale commercial “Pluton” plant with waste capacity up to 250 kg/h is under testing now. Process variables are given in Table I.

Table I. Plasma plants parameters.

| Parameter | “Pyrolis” | “Pluton” |
|---|---|---|
| Solid waste capacity, kg/h | 40-50 | 200-250 |
| Overall dimensions, m | 8 × 8 × 10 | 12 × 18 × 12 |
| Number of plasmatrones | 1 | 2 |
| Electric power of plasmatrone, kW | 70-120 | 100-150 |
| Time to get steady-state conditions | 3-4 | 6-8 |
| Specific power expenses, kW·h/kg | 1-2 | 0.5-1 |
| $^{137}Cs$ loss, %* | 5-11 | 7-9 |

* depends on waste composition

The other method of incinerator ash conditioning is soaking with high-penetrating cement grout. Ash is placed into container following by soaking with liquid cement slurry. The method has some advantages over conventional cementation such as higher productivity and product quality.

## LIQUID LILW VITRIFICATION

To process evaporator concentrates and reboiler residues a vitrification plant (Figure 3) has been designed and constructed. It is under operation since 1999 including vitrification of actual waste for the last two years. Liquid waste is concentrated in a rotary film evaporator to salt content ~1000 g/L. Salt concentrate is intermixed with glass forming additives (quartz sand, datolite, bentonite) in a batch mixer and feed in the form of slurry with ~25% water content is fed into a melter using a peristaltic pump. Melter is high-productive small-sized unit – cold crucible (Figure 4). Due to skull formation, no contact of high temperature melt with the cold crucible walls takes place that avoids corrosion problem. Process temperature is ~1200 $^0$C. Molten glass is poured into containers, annealed and containers are sent to repository. Off-gas is purified from radionuclides and hazardous components such as $NO_x$, $SO_x$ using a system of coarse and HEPA filters, $NO_x$ absorption columns for nitric acid recycling, and catalytic decomposition of residual $NO_x$ to $N_2$ and $O_2$. Vitrification process variables are as follows: glass productivity – up to 75 kg/h (three crucibles with productivity each of up to 25 kg/h), total power supply – 1000 kW, operation frequency – 1.76 MHz, single generator’s vibrating power – 160 kW, melting ratio – 5-6 kW·h/kg of glass, $^{137}$Cs loss –3-5%.

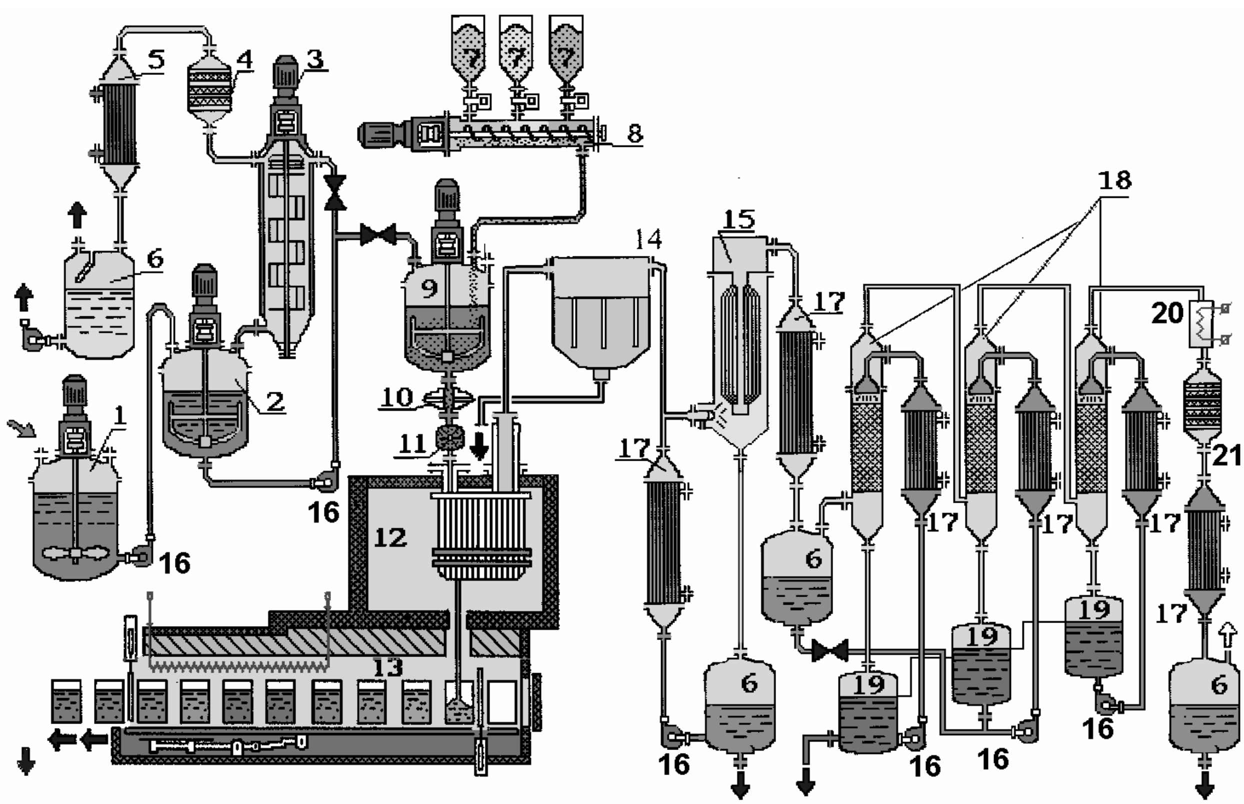


Figure 3. Vitrification plant flowsheet.

1 – waste storage tank, 2 – evaporator collector, 3 – rotary evaporator, 4 – filter, 5, 17 – heat exchangers, 6 – condensate collector, 7 – glass formers hoppers, 8 – screw, 9 – batch mixer, 10 – inductive mixer (dismantled), 11 – peristaltic pump, 12 – melter, 13 – annealing furnace, 14 – coarse filter, 15 – HEPA-filter, 16 – pump, 18 – $NO_x$ absorption column, 19 – nitric acid colletor, 20 – heater, 21 – catalytic reactor.

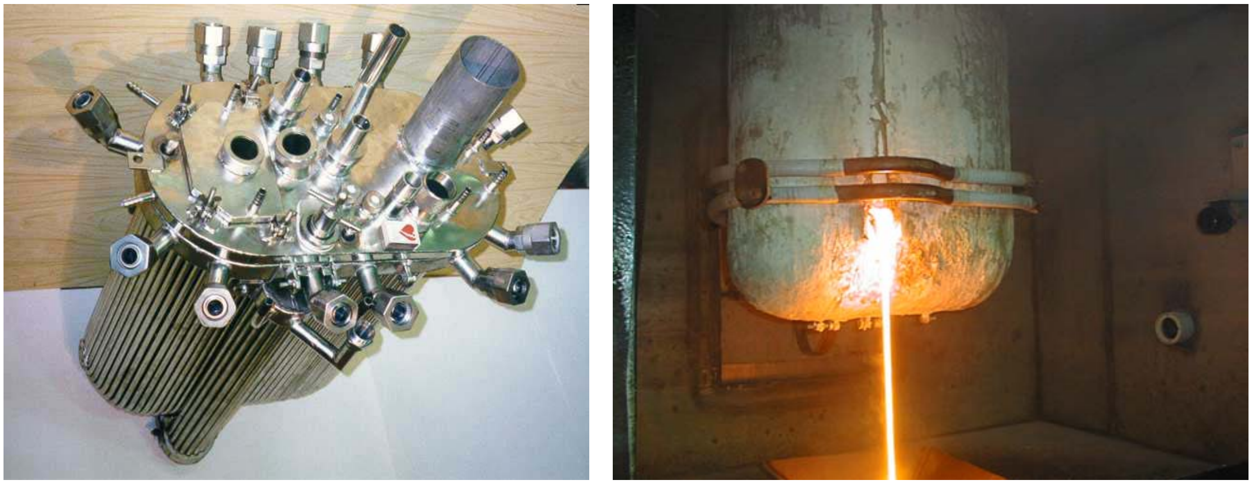

Figure 4. The cold crucible (general view – left, installed in a process box - right).

Works on development, calculation, and design of large-diameter (up to 1 m) cold crucibles are being also performed. Such crucible may reach glass productivity up to 500 kg/h (at dry feeding) at quite low melting ratio (2-3 kW·h/kg). The major problem is availability of powerful high frequency generators with vibrating power of several MW. This will also require lower operation frequency – 100-440 kHz. The high frequency generator with vibrating power 250 kW and operation frequency 440 kHz energizing cold crucible ~0.5 m in diameter has been successfully tested.

## TREATMENT OF RADIOACTIVE SILTS

A special method was developed to treat silts contaminated with radionuclides. Silt is fed onto a conveyer of electric furnace and treated at temperature of 500-700 $^{0}$C. Organic constituent is burned-off and water is evaporated. Off-gas and aerosols are sent to off-gas system. Cake of radioactive silt is crushed and discharged into 200 L barrels following by soaking with high-penetrating cement grout, exposure for solidification, and final disposal. Currently a bench-scale plant with silt capacity of 20 kg/h is under operation at SIA Radon. Process flowsheet is shown on Figure 5.

## THERMOCHEMICAL TECHNOLOGIES OF WASTE TREATMENT

One of the promising methods for low-level waste treatment is application of thermochemical reactions. A powdered metal fuel (PMF) is selected especially for the specified waste composition. At that, energy of exothermal reaction is used. The metal fuel is composed of metal or intermetallic powders, stabilizers, surfactants and other constituents. Up to date the following thermochemical technologies have been developed:

- Decontamination of metallic, asphalt, and concrete surfaces;
- Treatment of organic wastes such as spent ion-exchange resins, polymers, medial and biological wastes;

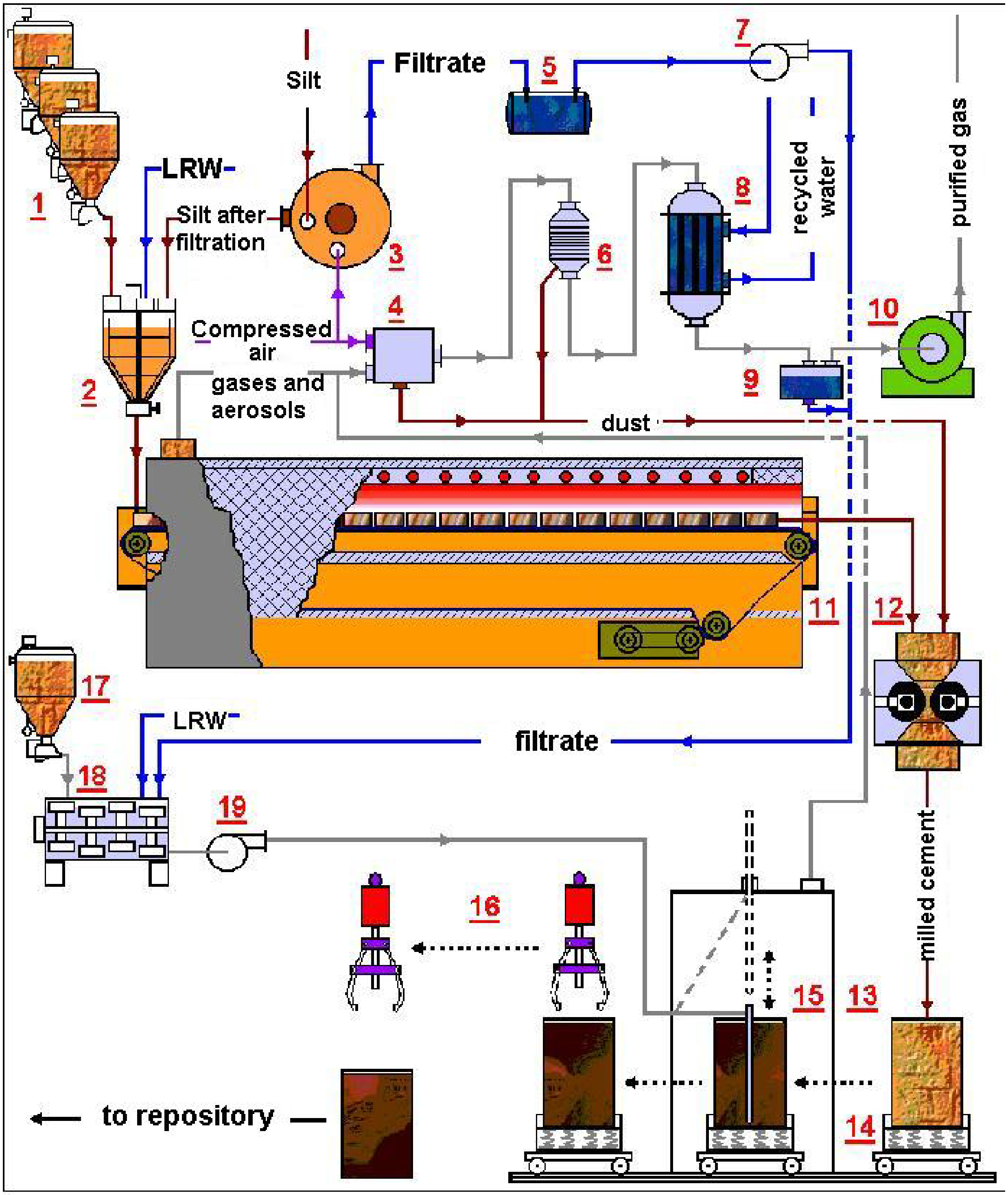


Figure 5. Radioactive silts thermal treatment process flowsheet.

1,17 – hoppers, 2 – mixer, 3 – separator, 4 – filter, 5,9 – collectors, 6 – HEPA-filter, 7,19 – pumps, 10 – ventilator, 8 – heat-exchanger, 9 – filter, 11 – electric furnace, 12 – crusher, 13 – container, 14 – truck, 15 – filling container with high-penetrating cement grout, 16 – tongs, 18 - mixer.

- Batch vitrification of incinerator ashes, calcines, spent inorganic ion-exchangers, contaminated soil, and other inorganic wastes;
- Treatment of $^{14}C$-containing carbon, nuclear reactor graphite, lubricants, etc.

The thermochemical decontamination process is based on thermal effect on surface layer of decontaminated material at burning of PMF layer spread over this surface. The thermochemical decontamination technology consists of several stages (Figure 6): determination of contaminated spot configuration, coating with thin layer (8-10 mm) of PMF; burning initiation, PMF burning, slag removal.

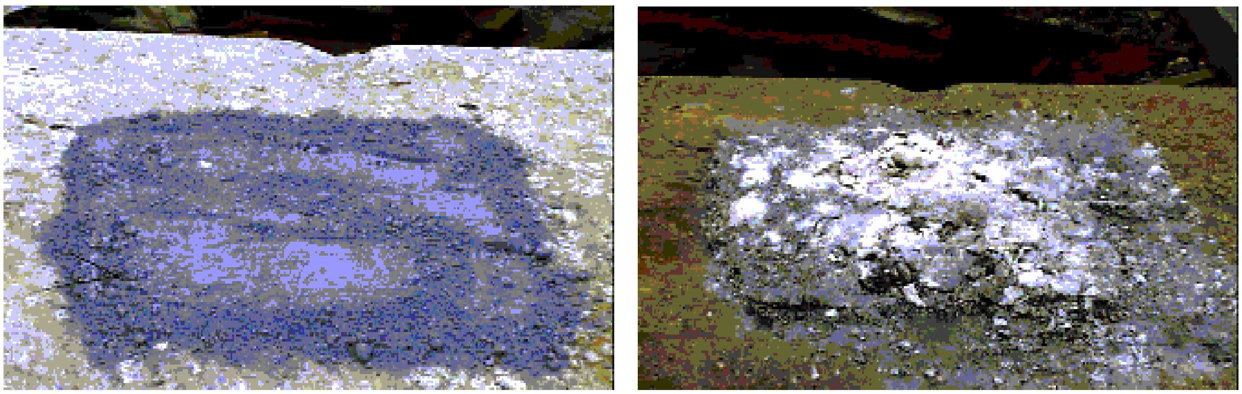

Figure 6. Concrete slab before (left) and after (right) thermochemical decontamination.

Decontamination of metallic surfaces takes place due to both radionuclide volatilization and immobilization in a slag layer produced at PMF burning. The method provides decontamination of metal at depth up to 50-100 μm with 90-95% efficiency.

Decontamination of concrete is based on the effect of concrete surface destruction due to thermal shock at PMF burning. Concrete fragments are chipped of with slag and removed after cooling. Concrete decontamination efficiency reaches 90-95% for single cycle. Decontamination depth is about 5-8 mm.

At asphalt decontamination contaminated layer is softened due to high temperature. Softened asphalt is easily removed mechanically at depth necessary to complete decontamination. In all the technologies decontamination cycle duration is 15-20 min.

Application of thermochemical methods provides processing of spent ion-exchange resins and organic wastes such as polymers, chlorine-containing waste (PVC), biological waste, etc. To process spent ion-exchange resin a mobile unit has been designed and constructed (Figure 7). Spent resin normally has high water content (>50 wt.%). Powdered metal (Al, Mg, Ca, Si and others) reacts with water with release of energy sufficient to keep resin decomposition process and interaction of slag with waste constituents yielding stable compounds.

Spent resin and PMF is pre-intermixed in the ratio required and charged into the reactor following by burning process initiation. Off-gas is purified in a mobile unit. The same method is used for thermochemical treatment of organic waste including polymers.

The process of batch vitrification of small portions of inorganic radioactive wastes such as incinerator ashes, contaminated soils, inorganic sorbents is also developed (Table II). The process is based on redox reactions in the mixture of PMF and radioactive waste with high heat release, which melts the mixture producing glass-like material without external energy source.

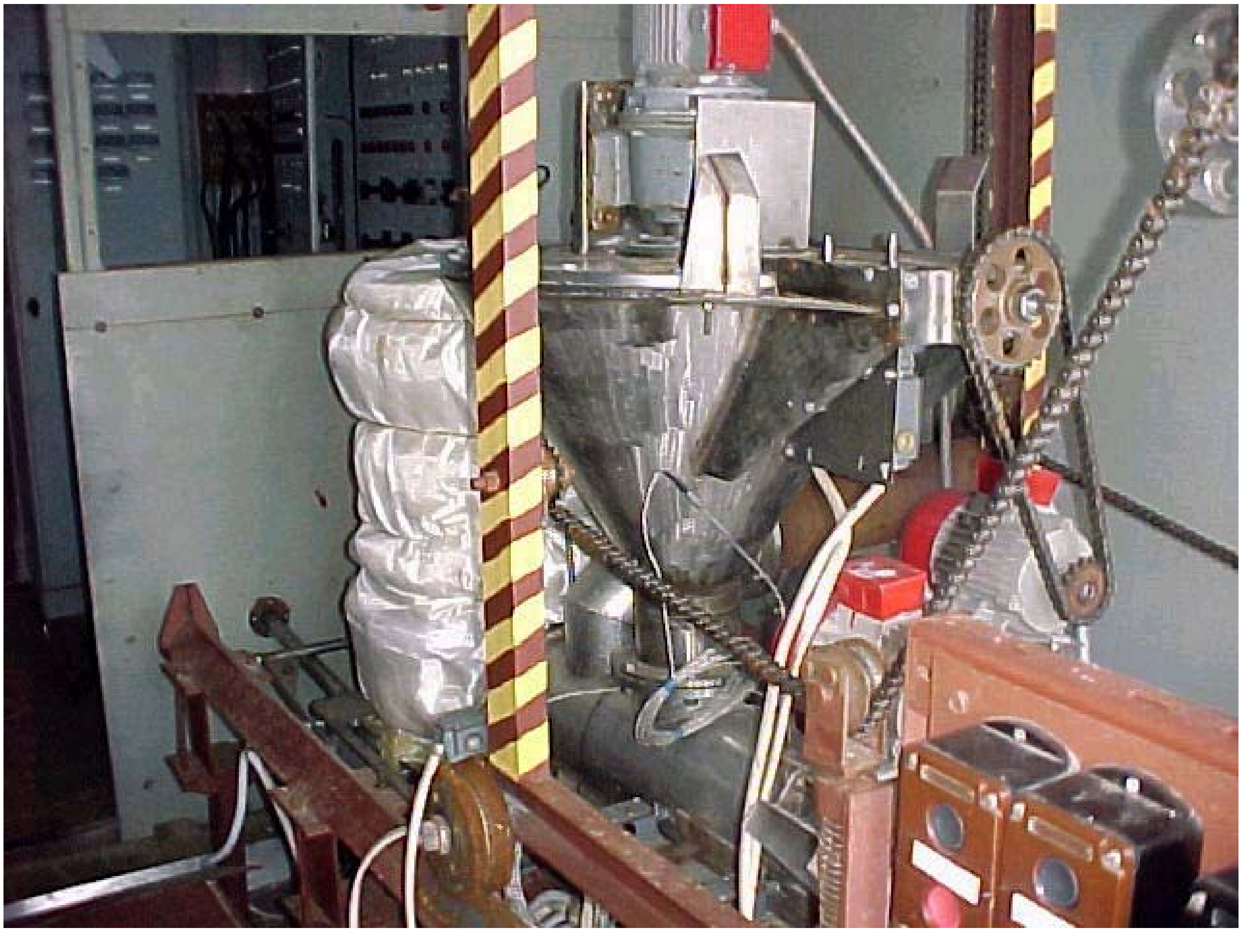

Figure 7. Spent ion-exchange resins treatment unit.

Table II. Batch vitrification process variables.

| Waste content, wt.% | Maximum process temperature, $^0C$ | Aerosols release, wt.% | $^{137}Cs$ loss, % | Leach rate (IAEA test [5]) from glassy product, $g/(cm^2 \cdot day)$ | |
|---|---|---|---|---|---|
| | | | | $^{137}Cs$ | $^{239}Pu$ |
| 50 (ash) | 1530 | 1.9 | 0.9 | $9.0 \cdot 10^{-6}$ | $5.4 \cdot 10^{-6}$ |
| 60 (ash) | 1200 | 1.0 | 0.3 | $7.9 \cdot 10^{-5}$ | $7.0 \cdot 10^{-5}$ |
| 45 (soil) | 1900 | 2.2 | 3.1 | $1.0 \cdot 10^{-5}$ | - |
| 56 (soil) | 1520 | 1.0 | 1.3 | $2.1 \cdot 10^{-6}$ | - |

The technology provides for intermixing of waste with PMF, charge of this batch in the crucible, initiating, and thermochemical process conduction. The final product is composed of PMF and waste oxides and has glassy structure, high density and low porosity.

Using this method contaminated graphite, for example graphite of uranium-graphite nuclear reactors, may be processed (Figure 8). Such graphite contains up to 1 wt.% of $^{14}C$ nuclide. Incineration of irradiated carbon is unacceptable due to high loss in atmosphere of biologically dangerous $^{14}C$ radionuclide in the forms of $^{14}CO_2$ and $^{14}CO$. Thermochemical processing of graphite is performed under inert atmosphere. The mixture of PMF and powdered

Figure 8. Thermochemical treatment of graphite: initiation (left), process development (middle), and final product (right).

graphite is placed in a crucible following by initiation of self-sustaining reaction. The result is chemically stable carbide-oxide assemblage safely fixing radioactive carbon and other radionuclides.

## CONCLUSIONS

Several high temperature processes and units deciding the problem of safe disposal of various radioactive wastes have been developed, designed, constructed and implemented at SIA Radon, among them are plasma treatment, vitrification, and thermochemical processes. Some of designs are applied in various fields of industries: nuclear industry, defence, science and medicine. A long time experience of SIA Radon in the field of radioactive waste conditioning and disposal is useful for not only Russia but for worldwide.

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