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Development of Plasma Techniques for Solid Radioactive Waste Processing

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Abstract—The analysis of development of plasma methods of processing of a waste is carried out. Technological, ecological and economic aspects of plasma processing on the basis of the Russian and foreign data are considered. The comparative analysis of efficiency plasma and traditional methods of processing of the radioactive waste is carried out and perceptivity of the further development of plasma technology of processing of a radioactive waste is shown.

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INTRODUCTION

Recycling of solid radioactive waste resulting from the operation and plant decommissioning and accumulating in stores at nuclear power plants in large quantities [1], is a technical challenge associated with the solution of the complex problems of technological and environmental issues. For radioactive waste disposal by dumping a special storage is needed, and therefore large investments are required in the construction of these stores (cost per 1 m^3 of radioactive waste storage facility is approximately 30000 rubles [2]). Therefore reducing the volume of radioactive waste becomes a crucial factor in the economy of their processing. Additionally, the final buring of radioactive waste requires its conditioning.

Depending on the specific activity, there are following categories of solid radioactive waste: very low active waste, low-active, intermediate active and highly-active. Waste of the last two categories that make up a small fraction of the volume of nuclear plant waste, are not recyclable, they are conditioned and sent for final burial.

Solid very low active and low-active waste are subjected to pre-processing, which involves extracting them from storage, fragmentation and sorting depending on the method of processing for combustible (wood, paper, clothing, rubber, footwear, personal protective equipment, rags, wiping, flammable fluids, etc.) and compressible (insulation, cables, glass, electronic and construction debris, filters, metal shavings, etc.).

Processing at the plant includes successive pressing and supercompacting, combustion in excess of air, cementing and bituminization. Metal and insulation melting is also used. Processed products in barrels (200 L) are placed into long-term storage in non-reusable protective containers, which also used to store some of the waste without processing.

Burning the combustible fraction of radioactive waste in kilns is one of the most common technologies to significantly reduce the amount of waste. The disadvantages of direct combustion of waste in excess of air include large volumes of exhaust gases and formation of secondary radioactive waste, ash requireing further disposal.

High-temperature plasma waste treatment methods have significant advantages: they provide high rates of waste reduction and the resulting decline in secondary waste volume, producing vitrified slag with high mechanical strength and chemical resistance, suitable for long-term storage and transportation.

Plasma Waste Processing Technologies

Development of plasma technology has a long history. Plasma heating sources were used mainly for melting and cutting of metals. From the 80ies of XX century

Table 1. Some of the commercial waste treatment plants[a]

Plant's site	Type of waste	Performance, t/day	Launch year	Notes
Bordeaux, France	MSW ash	10	1998	
Morcenx, France	Asbest	22	2001	
Bergen, Norway	Leather industry waste	15	2001	
Landskrona, Sweden	Fly ash	200	1983	
Jonquiere, Canada	Aluminium slag	50	1991	
Ottawa, Canada	Solid biological waste	85	2007	Demonstration plant, performance increased in 2010
Alpoca, Virginia, USA	Military equipment	10	2003	
Anniston, Alabama	Catalitic converters	24	1985	
Hawthorne, Nevada	Military equipment	10	2006	
Honolulu, Hawaii	Medical waste	1	2001	Closed
Madison, Pennsylvania	Biomass, Construction waste	18	2009	Pilot plant
Richland, Washington	Hazard waste	4	2002	Emissions occured several times: while treating hazardous, inluding radiactive waste
US Navy	Ships' waste	7	2004	
US Army	Chemical agents	10	2004	
Mihama-Mikata, Japan	Solid biological waste	28	2002	No energy production, slag used in road construction
Utashinai, Japan	Solid biological waste, auto scrap	300	2002	Pyrogas is used for energy and steam generation, slag is sent for burial
Kinura, Japan	MSW ash	50	1995	
Kakogawa, Japan	The same	30	2003	
Shimonoseki, Japan	″	41	2002	
Imizu, Japan	″	12	2002	
Maizuru, Japan	″	6	2003	
Iizuka, Japan	Industrial waste	10	2004	
Osaka, Japan	Chlorinated organic substances	4	2006	
Taipei, Taiwan	Batteries and medical waste	4	2005	

[a] According to Georgia Tech. Research Institute (2009).

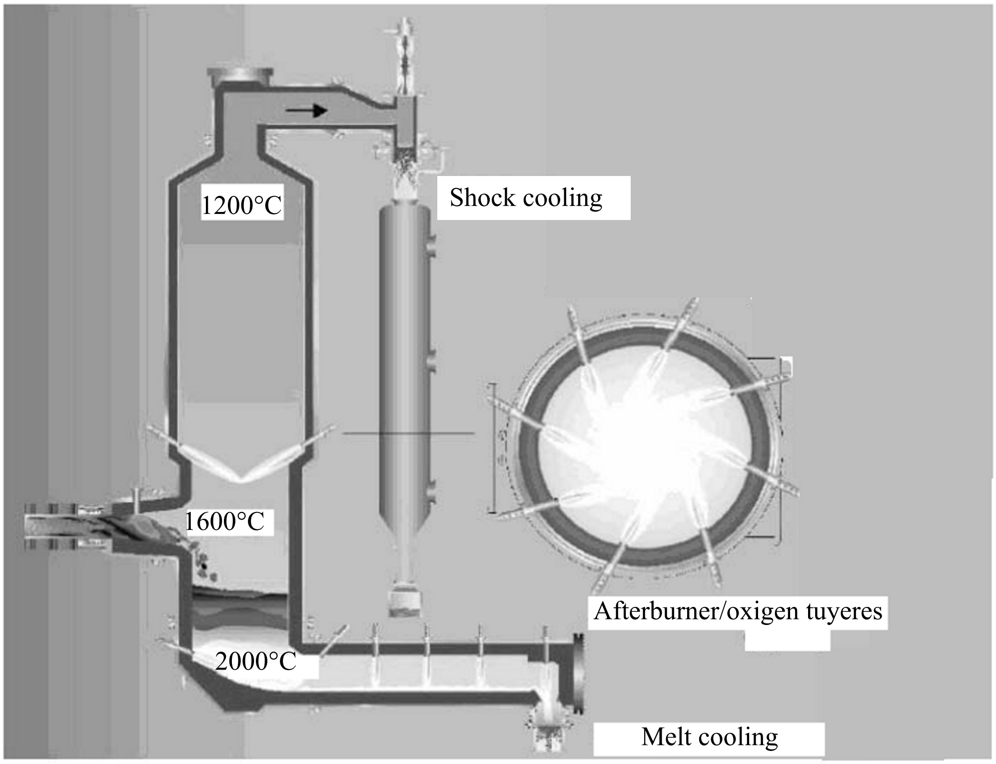

Fig. 1. Plasma gasifier "Thermoselect" flowsheet.

research and development to introduce high temperature plasma methods in industrial and consumption waste management. These methods provide almost a 100% recycling of waste to obtain inert vitrified slag and syngas.

Plasma methods are increasingly used in waste recycling, mainly hazardous industrial and medical, most of the technologies tested on the pilot/industrial scale (Table 1).

A realization of the waste gasification on an industrial scale using plasma heaters was a "Thermoselect" unit [3], built in 1992 (Fig. 1). This plant performs gasification of organic waste to produce synthesis gas (H_2, CO, CO_2) and melting of metals and mineral components.

Heating is provided by natural gas combustion with oxygen. Shaft furnace includes the following zones: melting, drying and gasification, organic waste incinerator. Pyrogas living the furnace is shock cooled from 1200 to 70°C with simultaneous cleaning by water. Rapid cooling minimizes supertoxicants formation.

Seven plants with performance of 95–555 t/day for recycling industrial and consumer waste, using "Thermoselect" technology were commissioned in 1999 in Japan.

Several commercial projects have been realized from 2002 to 2010: two plants in Japan (production and consumption waste, 24 and 220 tons/day) and two plants in India (hazardous waste, each 72 tons/day). As in the case of the technology from "Thermoselect," gasifying furnace is a shaft furnace [4] with melting, drying and gasification, organic waste afterburning zones. Heating is provided by plasma torches. Thermal power introduced by the plasma is relatively small: 2–5% of the calorific value of recyclables. The synthesis gas from the shaft furnace with a temperature of 890–1100°C is fed to the step of rapid cooling and water cleaning. Molten slag temperature is 1650°C.

Shaft furnace with countercurrent flows of raw materials and syngas is used in the pilot plant "Pluton" (250 kg/h), developed by SIA "Radon" for processing of solid radioactive waste [5]. Heating is provided by the plasma torches. Furnace contains feedstock drying, gasification, pyrolysis and melting zones. Pyrogas containing tar and volatile organic substances from the shaft furnace is fed to the combustion stage.

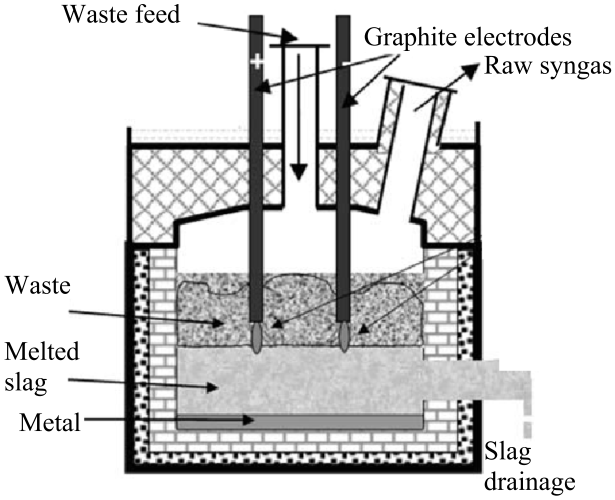

Fig. 2. Schematic diagram of the plasma furnace with waste feed on the liquid slag.

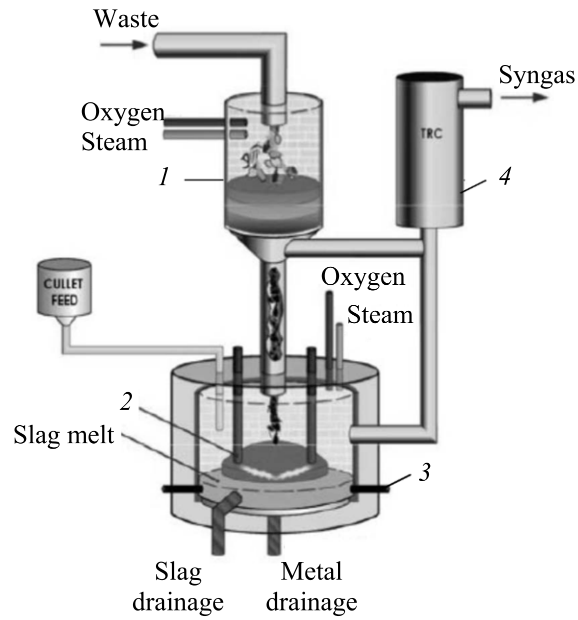

Fig. 3. PEM scheme: (*1*) pregasificator; (*2*) graphite electrode; (*3*) side electrode; (*4*) heat chamber.

In 2007, EER company from Israel commissioned a demonstration unit for solid waste treatment with the capacity of 500 kg/h, that uses technology of SIA "Radon" [6].

The project, developed at the Institute of Electrophysics and Electric Power of RAS (St. Petersburg), is based on a shaft furnace for waste gasification, plasma torches and the principle of co current flow of raw materials and pyrogas [7]. Technology is implemented at the pilot level (capacity up to 100 kg/h) and tested on different wastes. The main advantage of co-current gasification is the possibility of receiving at temperatures above 1200°C pyrogas without tar and volatile organic compounds, that eliminates afterburning step.

Besides plasma waste processes in shaft furnaces, a two-stage treatment of the waste is used: in the first stage low-temperature pyrolysis/gasification technologies are used, in the second stage pyrogas and solids formed in the first stage are subjected to high-temperature plasma treatment for afterburning of residual resins, volatile organic compounds and carbon gasification, as well as melting of solids. Two-step processes in commercial and demonstration scales were realized by "Plasco Energy" (Canada) [8], "APP" (England) [9], "BGT" (Germany) [10], "Europlasma" (France) [11].

Common technique, especially in the case of relatively small capacity plants, is waste feed on a layer of molten slag in the crucible melter heated by plasma torches or plasma arc (between the graphite electrode and the molten slag or two graphite electrodes) (Fig. 2).

From the melting furnace product gases enter the afterburning and cooling/scrubbing systems. Arc plasma heating process [12] was implemented by "Tetronics" (England) in commercial applications: 13 plants (performance up to 42000 tons/year) for the disposal of organic and inorganic hazardous industrial waste (ash incinerators contaminated with chlorinated organic compounds, oil sludges, spent catalysts, contaminated soil) were commissioned in 1997. For the disposal of hazardous waste "PEAT" (USA) since 1992 supplies similar plants with capacity of up to 24 tons/day [13]. "Pyrogenesys" developed a technology that combines gasification of waste in the melting crucible furnace with arc heating and organic substances afterburning stage with pyrogas feed to plasma torch [14].

"InEnTec" (USA) developed PEM unit with performance up to 25 tons/day for hazardous waste treatment[15] (Fig. 3).

PEM units use two-step process of pre gasification and gasification of waste in the electric arc heating with oxygen and superheated steam supply. Totally, 11 such units (including one transportable) were commissioned for the treatment of hazardous waste.

For disposal of hazardous and radioactive waste “Retech Systems” (USA) has developed a technology PACT. The technological process is carried out in a furnace including an inner centrifuge for uniform heating and mixing of the waste [16]. For heating to 1200–1500°C plasma torch operating in two modes is used: direct and indirect arc. The resulting gaseous products are fed to the stage of post-combustion, cooling and scrubbing. Five PACT units of different performance are currently in pilot and commercial operation. (A detailed description of a commercial plant Zwilag Plasma facility (ZPF), using PACT-8 technology for the disposal of radioactive waste is given further.)

In general, in the development of plasma waste disposal methods there are two main directions: the first, construction of large scale plants for plasma treatment of production and consumption waste with energy production, the second, small in-factory or transportable units for hazardous waste treatment. Computer modeling of physical and chemical processes and the movement of gas flows in the plasma furnace is widely used in the development and optimization of plasma technology.

Plasma Units for Radioactive Waste Processing

In 2006, the IAEA has recognized technology on the basis of plasma processes as manufacturing method, which allows to process all major types of low-level radioactive waste [17]: liquid organic, inorganic liquids, organic solids, inorganic solids, mixed organic-inorganic solids, mixed organic-inorganic liquids, used resins.

Several types of units for the processing of low and intermediate active waste by plasma technologies are in use. Generally, they are pilot or pilot-scale units, so there is a necessity of trial operation in test mode for several years after their launch. For example, in Japan there are three units [18], using the energy of the plasma arc for processing of intermediate level waste. Two of them are in Tokyo Research Center of Atomic Energy Research Institute of Japan. One unit with performance of 4 tons/day, uses direct heating of waste with DC arc. Another unit with the same performance consists of three induction plasma torches of 200 kW each and an induction furnace, of up to 800 kW. The third unit owned by “Japan Atomic Power Company” (JAPC), is based on a PACT-8 system, uses indirect and direct heating of waste with electric arc of 1200 kW plasma torch. Unit performance is 600 kg of waste/h.

A plant that uses PAM technology from “Retech” works in South Korea [19]. Operational principle is based on the direct contact of arc with the radioactive waste without centrifuge; plant performance is 200 kg/h of combustible waste, 300 kg/h of non-combustible waste.

British company “Tetronics” has an experimental unit for the processing of model wet sludge of intermediate level waste, including filter loading (sand, etc.) [20]. Two graphite electrodes are used for heating. Reduced emissions of the radionuclide Cs is achieved by the introduction of silicon into the melt.

Related high-temperature pyrolysis process developed in the company “Studsvik-THOR” (Thermal Organic Reduction) [17] for the processing of radioactive waste can be regarded as a High Tech process. It is two-step process, it allows to process different solid and liquid waste such as ion-exchange resins, charcoal, graphite, sludge, oils, solvents and cleaning solutions, with contact radiation level of up to 400 R/h. First stage, pyrolysis of the organic component at ~750°C, the second stage, afterburning of the volatile organic substances and tar at ~1200°C. Inorganic residue in the form of coke is fed to cementing. Temperature of pyrolysis zone is selected with a view of minimizing emission of volatile radionuclides (Cs and Ru).

In accordance with the technological solution, resulted from R&D in GupMosNPO “Radon,” launched in the late 70 of XX century [21] on the problem of disposal of low and intermediate level radioactive waste, units “Pyrolysis” and “Pluton” for the processing of solid waste by plasma method were built [22–24] (Table 2). Technology is based on the high temperature plasma pyrolysis of waste in a countercurrent shaft furnace with slag tap as the best way of getting conditioned waste. On the “Pyrolysis” (1998–2001) and on the “Pluto” (2007) units were optimized process conditions using mock-waste and real solid radioactive waste, similar in composition to the waste of nuclear power plants. Currently, “Pluto” is licensed to recycle solid waste with specific activity of 2.2×10^5 Bq/kg for α-emitting radionuclides and 3.7×10^6 Bq/kg for β- emitting radionuclides. Since 2011, this facility is processing MSW from the Kursk NPP [25].

Table 2. “Pyrolisis” and “Pluton” technical parameters

Parameter	“Pyrolisis”	“Pluton”
Solid waste performance, kg/h	40–50	200–250
Number of plasma torches	1	2
Plasma torches power, kW	70–120	90–150
Heating time, h	8–12	16–24
Specific energy expenses, kW h kg^{-1}	1–2	0.5–1
Dimentions (outer), m	8 × 8 × 10	12 × 18 × 12

Flow diagram and description of the “Pluton” are given in the paper “The Russian experience in solid waste processing: Achievements and Prospects” (authors: A.N. Bobrakov, A.A. Kudrinskiy, V.M. Kulygin etc.).

Since 1985, “Retech” (USA) conducted R&D on the plasma waste recycling. One result of this work was creation a series of units that use technology PACT (Plasma Arc Centrifugal Treatment) and PCF (Plasma Centrifugal Furnace) [26–28], that were designed with the view of processes producing highly pure metal melts and acetylene synthesis using plasma technology. Latest units of this series PACT-6 (pilot version) and PACT-8 were developed in 1991, and in 2000 technological upgrading of PACT-8 was done.

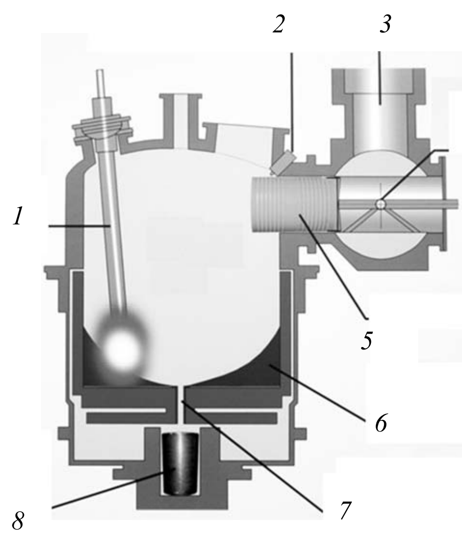

Fig. 4. PACT-8/Zwilag (ZPF) unit with radioactive waste feeder [26]: (*1*) plasma torch; (*2*) propane torch; (*3*) waste download chamber; (*4*) rotator; (*5*) barrel with waste; (*6*) molten slag; (*7*) drain valve; (*8*) vitrified slag.

The licenses have been granted for the processing of solid radioactive waste on two PACT-8 units owned by “ZWILAG” (Zwischenlager Würenlingen AG, Switzerland) and “Japan Atomic Power Company” (JAPC, Japan) [16]. Since 2004, Zwilag plasma facility (ZPF) unit is processing radioactive waste from Swiss nuclear power plants and medical facilities [29]. In 2009, the license was granted for handling radioactive waste ($\leq 1 \times 10^{12}$ Bq) for a period of five years.

Plasma chemical reactor ZPF (Fig. 4) is a rotary hearth furnace, with the molten slag (1200–1500°C) inside. Solid radioactive waste for treatment is fed in 200 liter barrels. Through the rotary device for shredding waste is fed on to a propane torch. Then cut pieces of barrels and waste are falling on a rotating melt layer. On the surface of the molten slag organic part of the waste is gasified, the inorganic fraction is melted and mixed with the melt. Rotating unmelted part of the waste is constantly falls under the plasma torch, with centrifugal force keeping the slag away from the drain hole. When stopping the centrifuge the slag is collected in a slag pan, and through the drain valve into the container.

The gas purification system of ZPF includes acidic scrubber and module for selective catalytic reduction of nitrogen oxides.

Secondary waste resulting from gas purification on ZPF, can be further processed by filtration and evaporation of the salt cake. This approach allows to transfer into the slag 90% of the radioactive volatile components [29].

PACT-8 is equipped with a linear arc plasma torch capable of operating in both direct and indirect heating mode. As the counter electrode in the plasma torch can be used directly melted slag and the electrode of the plasma torch. To initiate the arc inert helium is used, the working plasma gas-nitrogen. At a time when recyclable waste has not yet warmed up, the plasma torch operates in indirect heating mode, as the electrical conductivity of the waste is small. After melting the waste plasma torch switches to the direct heating mode.

The plasma torch in PACT-8 requires a sufficient electrical insulation of the melting chamber and the presence of conductive elements in the melt in sufficient proportion to maintain the electrical conductivity of the slag within the prescribed limits. Table 3 shows the comparative characteristics of the

Table 3. Comparison of "Pluton" and PACT-8

Parameter	"Pluton"	PACT-8
Plasmochemical reactor type	Countercurrent, shaft with melter	Rotating pan furnace
Dimentions (inner)	Shaft 0.8 × 0.8 × 7.2 m	Height 1.5, diameter 2.5 m
Performance	200–250 kg/h	200 kg/h—combustible waste, 300 kg/h—non combustible waste
Waste feed type	Cruft	Barrels (200 L)
Specific energy consumption[a]	0.4–0.6 kW h kg^{-1}	4–6 kW h kg^{-1}
Heating time	16–24 h	85 h
Plasma torches type	Electric arc, indirect	Electric arc, direct/indirect
Plasma torches power (el.)	90–150 kW	1200 kW
Number of plasma torches	2	1
Plasma forming gas air	Air	N_2
Flue gas temperature	200–300 °C	1200–1500 °C
Reactor temperature	200–1700 °C	1200–1500 °C
Slag temperature	1200–1500 °C	1200–1400 °C
Slag specific gravity	2.4–2.9 g/cm^3	2.4–2.6[b] g/cm^3
Afterburner type	Plasma torch	Gas torch

[a] Only power fed to plasma torches is accounted. [b] According to [30] for PACT-8 model waste.

"Pluton" and PACT-8 units and solid radioactive waste refining processes in them.

Environmental Aspects of Recycling Radioactive Waste

The possible impact of the plasma processing of radioactive waste on the environment and population is caused by radioactivity of the processed waste and the refining process itself(the formation of the flue gas, which contains harmful substances such as chlorine and fluorine-containing hydrocarbons, dioxins and furans, the formation of salt solutions in the gas cleaning system, etc.).

Heavy metals contained in the gas cleaning system and slag present a certain problem. For example, the content of heavy metals in the slag compound, produced by PAM-200 (it is assumed that all radioactivity "left" in the melter, goes to the slag) depending on the composition of the raw waste is: Cs 56–85.4%; Co 96–99, 96%; Hg 19–30%; Cd 57–91% [19]. Figure 5 shows the distribution of heavy metals, deposited at different stages of processing non combustible wastes on PAM-200.

Lowest content of cesium in the slag (or on the surface of the reactor) is observed in the case of combustible waste, maybe that is because the burning of these wastes emit extra warmth and thereby raises the temperature, and this can lead to increased

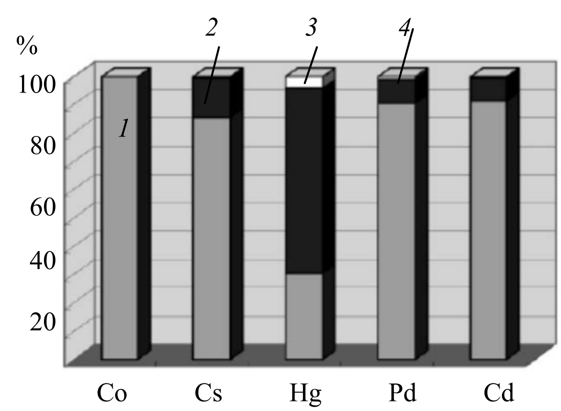

Fig. 5. PAM 200: Heavy metals distribution. (*1*) Reactor, (*2*) scrubber, (*3*) HEPA, (*4*) emission.

Table 4. Heavy metals distribution on “Pyrolis” unit

Isotope	Isotope content (mass%)		
	slag	pyro gas	furnace
^{137}Cs	27.8	11.8	~60.4
^{134}Cs	27.0	9.0	~64.0
^{60}Co	80.0	2.25	~17.7
Sa rel. ^{239}Pu	74.2	<1	~25

entrainment Cs as a result of evaporation and due to combustion reactions.

On the basis of measurements made in 2001, on the “Pyrolysis” unit, the following distribution of radioactive nuclides in radioactive waste was obtained (Table 4) [30].

Release of radionuclides is extremely sensitive to temperature and temperature distribution inside the shaft furnace.

From mid- 2004 to June 2005 on the Zwilag Plasma Facility (technology PACT-8) were processed mixed waste of total activity 9.4×10^{10} Bq, with 8.4×10^{10} Bq "left" in the slag [15]. The isotopic composition of the waste to be processed: ^{54}Mn, ^{60}Co, ^{65}Zn, ^{95}Zr, ^{134}Cs, ^{58}Co, ^{125}Sb, ^{137}Cs, ^{3}H, ^{14}C, ^{241}Am; average activity of solid waste 2.1×10^{10} Bq/barrel.

A broad range of wastes that can be processed by plasma method and a single stage process for obtaining a product suitable for long term storage, determine the advantages of this method compared with the method of combustion.

Thus, at the output of the gas cleaning system of “Pluton” unit total content of dioxins and furans in the flue gas is about five times lower than the European standards for waste incinerators. Concentration of heavy metals in gas emissions into the atmosphere is also below the standards for waste incinerators in Western Europe.

Economic Aspects of Plasma Processing of Radioactive Waste

Paper [1] gives an estimate of costs for processing in three 2,000 m^3 of solid radioactive waste for the russian conditions.

Option 1. Combustible waste is incinerated and then ash is cemented. Compactable waste is processed sequentially by compaction and supercompacting. Waste processing products in 200 l drums are placed in protective containers (container capacity 250 m^3). Costs of conversion and conditioning make ~45000 RUR/m^3 waste.

Option 2. Combustible waste is incinerated and then ash is cemented. Compactable waste are compacted. Waste processing products in 200 L drums are placed in protective containers (container capacity 515 m^3). Costs of conversion and conditioning make ~34000 RUR/m^3 waste.

Option 3. Plasma processing of combustible and compactable waste. Chilled receiving containers with slag placed in protective containers (capacity 60 m^3). Costs for plasma processing and conditioning make ~21000 RUR/m^3 waste.

Specific capital and operating costs for the plasma unit is higher than for the combustion plant, cementing, pressing and supercompacting facilities separately. But considering that variant 1 requires four units (incineration, ash cementing, pressing and supercompacting), while variant 2, three settings (incineration, ash cementing, compression), the capital and operating costs for the single-step plasma process are lower than in the case of traditional plants. Final costs for processing of 1m^3 of waste on plasma unit are 1.5–2 times less.

Table 5. Intermediate active sludge disposal requirements (UK). For 3000 m^3 of wet sludge

Parameter	Cementing	Plasma method
Waste volume change	+100%	–57%
Total storage room	60000 m^3	13000 m^3
Number of containers with raw waste (4 × 500 L barrels)	30000	6500
Number of containers with plasma processed waste per storage	6500	6500
Number of storage facilities	5	1

Table 6. Total expenses for processing 30000 m^3 of sludge (in USD) (UK)

Costs	Cementing	Plasma method
Conditioning	300000000	65000000
50-Year life cycle per one storage	200000000	–
Building/temporary storages	923000000	200000000
Transportation container	750000 (12 containers per storage)	–
Transportation containers(capital costs)	42000000	9000000
Transportation cost of one container	1000	–
Transportation to storage and final burial site	60000000	13000000
Total cost	1325000000	287000000

For economic evaluation the finite volume of products to be buried, and the associated capital and operating costs for storage of conditioned radioactive waste also should be taken into account.

Economic costs attributable to commercial installation Zwilag Plasma (Switzerland) (cost of processing, conditioning and disposal of radioactive waste) constitute 20.0 CHF/kg [29]. This amount includes the cost of transportation to the plant, temporary storage, processing, product packaging, disposal of conditioned product.

In [31] for the UK conditions, is given an estimate of costs for the entire life cycle of the processing of accumulated wet sludge with average activity by plasma treatment and cementation (Table 5). From Table 5 it is clear that the plasma processing provides a significant reduction in the conditioned product volume to be disposed of, compared with cemented.

The full costs for processing 30 000 m^3 of sludge plus costs at all stages of the life cycle (air conditioning, transportation, disposal, etc.) are shown in Table 6. These estimates show that the plasma processing provides ~4.6 times lower cost than cementing.

CONCLUSIONS

The introduction of plasma technology for the processing of solid radioactive waste plant enhances economic efficiency of handling combustible and noncombustible radioactive waste due to economies on waste storage equipment and conditioning operations.

Plasma technology not only solves the problem of the newly formed NPP operational waste, but also provides deep thermal processing of radioactive waste accumulated before or compacted in drums to make room for storage of solid radioactive waste.

Plasma methods and hardware design solve the environmental problems, allowing creation processing units both pilot to refine the process parameters and industrial for waste treatment.

Further modeling of physical and chemical processes, the transfer of cesium and other radionuclides is required for analysis of experimental data and process optimization of various plasma processing of radioactive waste.

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