USE OF ELECTRIC-ARC PLASMA FOR RADIOACTIVE WASTE IMMOBILIZATION

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We analyze the basic trends in the use of electric-arc plasma for radioactive waste decontamination. We present a scheme and performance figures of a process for a shaft-type furnace with fuel-plasma heating. We present the results of numerical and experimental investigations for a 150-kW direct-flow plasma reactor.

A new trend in ecological application of thermal plasma is its use for decontamination of radioactive waste (RAW). Investigations are being carried out in several directions: intensification of the combustion of radioactive waste in shaft furnaces with vitrification of combustion products in the lower portion of the furnace, when air-operated plasmatrons are used along with fuel burners [1-3]; vitrification of radioactive waste in a melt in a plasma furnace with heating by an arc burning between a cathode and the melt surface [4-7]; treatment of a glass-forming charge in a plasma flow [8-10].

One of the variants of the use of plasma for immobilizing radioactive waste is thermal treatment of highly dispersed nitrate solutions for the purpose of synthesizing powders of a homogeneous mixture of oxides whose pressing and annealing yield chemically stable mineral-like compounds also involving radioactive elements from the original solution. For example, the powders that were synthetized by the authors of [11] held compositions of the oxides of zirconium, barium, aluminum, calcium, cerium, europium, gadolinium, thorium, and uranium.

A schematic diagram of a pilot plant for treating unsorted solid radioactive waste in a shaft furnace with fuel-plasma heating is presented in Fig. 1. The plant, having a waste output of up to 50 kg/h, was constructed at the Scientific Industrial Association "Radon" (Moscow) in cooperation with the Heat and Mass Transfer Institute of the Academy of Sciences of Belarus (Minsk) [2, 3]. The furnace has a height of 3 m, a hearth area of 0.12 m^2, and a volume of 8 m^3. Solid waste (wood, paper, polymers, ceramics, glasses, metals, building refuse, dead bodies of experimental animals, etc.) is charged through a sluice 1 into a shaft 2 heated by two electric-arc plasmatrons 3 with a power of 50 kW each and fuel prechamber 4. The air excess coefficient is 0.3−0.4. Sinking to the bottom under gravity, the waste passes through zones of drying, gasification, pyrolysis, combustion, and melting. An inorganic residue in the form of liquid slag drains down from the inclined hearth of the furnace at a temperature of about 1450°C into containers, where it is solidified, annealed in furnace 5, and is sent to be buried. The gases from the combustion zone pass through the upper layer of waste, enter a combustion chamber 6 with a fuel injector 7, pass through an evaporative heat exchanger 9, a metal-bag filter 10, a condenser 11, a preheater 13, a fine filter 14, and then by means of a fan 15 are released into tube 16. The volume of exhaust gas is 140−160 m^3/h, which is 3−4 times less than in flame furnaces of the same output, which consume excess oxygen. The condensate 12 is fed by a pump 17 through an injector 8 into a heat exchanger 9 as a coolant. Waste is added periodically as the height of the waste column in the shaft decreases.

The β- and γ-specific activities of the solid waste amounted to about 10^6 Bq/kg, whereas the α-activity was five- to tenfold lower. The main carrier of radioactivity was Cs^{137}. The volume of the wastes decreased by a factor of ten after having been treated in the furnace. Due to filtration of effluent gases by the bed of waste, the carryover of radionuclides for a bed of height 1.5−2 m was less than 2%. Radiometric analysis of samples of brick lining proved its slight contamination with radionuclides. The rate of leaching of radionuclides from the melted slags
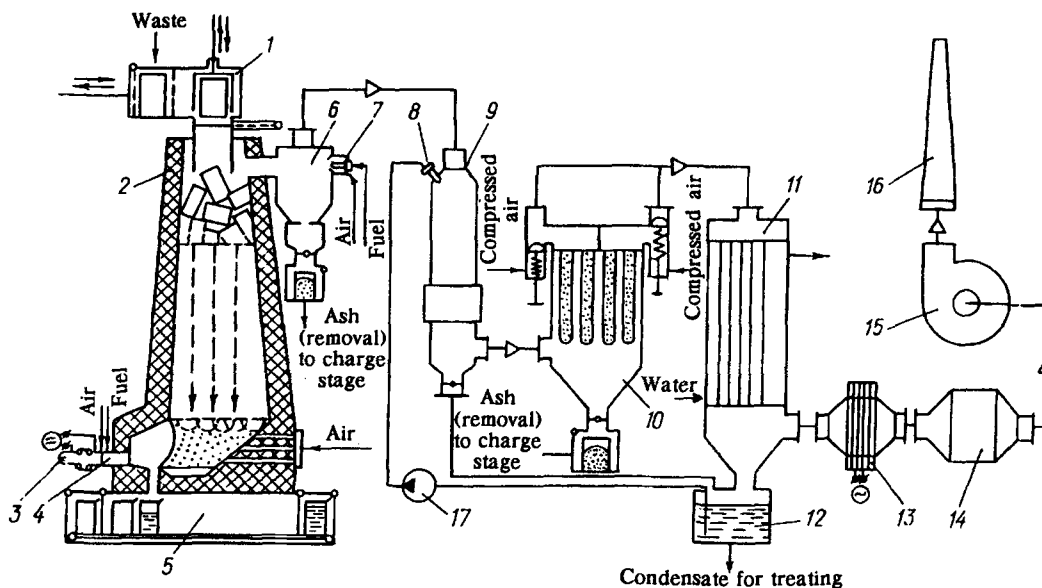
Fig. 1. Technological scheme of a pilot plant for treating unsorted solid radioactive waste in a shaft furnace with fuel-plasma heating.

satisfied the requirements imposed on solidified radioactive waste. The composition and structure of slags may be regulated by additional fluxing of waste with inorganic materials [3].

The possibility of vitrification of radioactive waste in 40-kW direct-arc plasma furnaces was investigated in Canada [5] and France [6, 7]. Waste in amounts of 2.5 kg containing 83% of cellulose and other organic materials as well as an admixture of radioactive salts Ce, Co, Cs, and Sr was fed for an hour into a furnace in which 15 kg of basalt was melted before hand [6, 7]. As a result, homogeneous chemically stable glasses were obtained that included 98.4% of the supplied quantity of Ce, 82.4% of Co, 96.9% of Sr, and 23.5% of Cs. The proportion of Cs that settled on the walls of the furnace was 52.4%, 11.5% along the route of its motion, and 12.6% on the filter. In [5] investigations on a 40-kW setup showed that transition from a direct arc to an indirect plasmatron for heating glass-forming materials containing radioactive substances makes it possible to considerably decrease the entrainment of sodium and cesium from the glass.

In the beginning of 1990 a plant was constructed in Switzerland for plasma vitrification of radioactive waste (PLASMARC-plant). The central part of the plant was a rotating plasma furnace heated by an indirect plasmatron of power 1.2 MW moving in the zone of the furnace [12]. The raw material can be fed into the furnace in dispersed form or in 200-liter barrels that are forced horizontally through a special apparatus to the furnace inlet, are cut into pieces, and fed into the furnace, where combustion of fuels, melting, and chemical interactions of the raw material components with additions occur. The melt is discharged through an opening at the furnace bottom when the motor that rotates the furnace is stopped. The exhausted gases are quenched by water. The admixtures that may occur in this case in water (for example, Cs^{137}) are removed in ion exchangers. After passing through scrubbers and a filter, the gases are freed of nitrogen oxides by means of catalysts and NH_3 additives at $300^\circ C$.

A continuous-flow plasma reactor can be used for heating a dispersed charge meant for vitrification of radioactive waste. Here, waste with a moderate level of activity, such as water slimes with the content of salts of $200-600 \, kg/m^3$, is suitable. The main component here is sodium nitrate. The content of calcium, magnesium, iron, phosphates, sulfates, and of other components can change from trace amounts up to 10–20%. Usually, the main carriers of activity are Cs^{137}, Cs^{134}, and Sr^{90}. As regards other β-, γ-, and α-emitters, the volumetric activity is by 3–4 orders of magnitude lower. In investigations of plasma vitrification of radioactive waste based on sodium salts and having a moderate level of activity that were carried out at the Heat and Mass Transfer Institute of the Academy of Sciences of Belarus in cooperation with the Scientific Industrial Association "Radon" (Moscow) the radioactive waste had the following composition (wt.%): $NaNO_3$ 68.5; Na_2SO_4 1.5; $CaCO_3$ 10.2; $Fe(NO_3)_2$ 16.8; $MgO+Al_2O_3+P_2O_5+Cl$ 3% and their simulators: $NaNO_3$ 90; $CaCO_3$ 5; $Fe(NO_3)_2$ 5% and $NaNO_3$ 90; Na_2SO_4

TABLE 1. Calculated Compositions and Certain Properties of Standard Glasses Obtained in Crucibles in Resistance Furnaces From Charges Used for Immobilization of Radioactive Waste

No. of a charge	Content of components in a glass, wt.%							Density, kg/m^3	Rate of the leaching of Na, $g \cdot cm^{-2}/24$ h	
	Na_2O	Na_2O_4	CaO	Al_2O_3	Fe_2O_3	B_2O_3	SiO_2		1st 24-h period	30-th 24-h period
1	19.3	–	17.5	3.3	3.7	7.5	48.7	2550	$4 \cdot 10^{-5}$	$5 \cdot 10^{-6}$
2	18.2	5.6	15.5	3.2	2.6	7.4	47.5	2520*	$5 \cdot 10^{-2}$	$8 \cdot 10^{-6}$
3	14.0	0.8	18.9	3.2	5.8	7.4	48.2	2510	$5 \cdot 10^{-5}$	$4 \cdot 10^{-6}$
4	27.7	–	3.7	10.0	6.7	–	51.9	2500	$1 \cdot 10^{-4}$	$1 \cdot 10^{-5}$

Note: Nos. 1-3 were obtained at 1300°C with holding for 3 hours, No. 4 — at 1650°C with holding for 1 hour.

*Density of the glass phase.

10%. The vitrifying additives were quartz sand (above 98% SiO_2), loam (70 ± 3% SiO_2, 12 ± 2% Al_2O_3; 6 ± 3% Fe_2O_3; 4 ± 2% Na_2O+K_2O; 3 ± 1% $CaO+MgO$ with admixtures) and datolite concentrate (CaO 37 ± 3%; B_2O_3 19 ± 3%; SiO_2 37 ± 4%, H_2O with admixtures). A special series of experiments showed that the use of boric acid in the charge for a plasma reactor is unacceptable because of the high level of boron oxide escape into the gas phase. The use of a datolite concentrate that contains the minerals datolite $Ca_2[B_2Si_2O_8(OH)_2]$, calcite $CaCO_3$, merwinite $CaMg(SiO_4)_2$, and α-quartz SiO_2 decreases the escape of boron multifold.

The compositions of the glasses vary depending on the proportions of the charge components, fluctuations of their compositions and on the temperature regime of the production of the glasses. Table 1 lists the compositions and properties of standard glasses produced by synthesizing in crucibles model charges Nos. 1, 2, 4 and charge No. 3 containing salts of radioactive waste, silica, datolite, loam in the weight proportion 4:1.5:3:1.5.

In order to meet the technological requirements for minimum escape of sodium compositions and production of glassses with a high stability against leaching, several variants of the introduction of a model nonradioactive charge into a reactor and of the organization of the motion of the charge, gas, and melt in the reactor channel were investigated on an experimental setup at the Heat and Mass Transfer Institute [8, 9]. It was found that tangential introduction of plasma jets or raw material into a mixing chamber did not exert a substantial influence on the quality of the glasses produced. The cyclone-induced motion of raw material with tangential introduction of the plasma-forming gas into a 5 cm-diameter reactor was observed at a distance not exceeding 7−8 cm from the reactor cross-section at the level of the nozzles of the plasmatrons. The countercurrent schemes were characterized by increased entrainment of the raw material. As a result of these investigations, the Scientific Industrial Association "Radon" selected as the main variant for its pilot plant a 150-kW direct-flow reactor with a cylindrical 5 cm-diameter mixing chamber and three electric-arc plasmatrons of 50 kW each and a water-cooled cylindrical reactor lined on the inside with electrocorundum. The inner diameter of the lined reactor was 5 cm. In the majority of experiments the overall flowrate of the plasma-forming air was 5−7 g/sec; the initial temperature of the plasma was 4800−5300 K; the consumption of the charge varied from 2.8 to 20 g/sec. The charge was delivered through an opening at the center of the top lid of the mixing chamber by means of an endless screw batcher and small amounts of a transporting air. In addition to a charge that was a mechanical composition of components (we will call it a dispersed charge), a compacted charge was also used in experiments, which was prepared by compressing a dispersed charge between rollers with the addition of liquid glass as a binder and a fritted charge obtained from the original one by preliminary treatment up to 700°C and subsequent grinding.

Numerical calculations of the heating of glass-forming particles containing CaO 11.6; B_2O_3 5.8; Fe_2O_3 3; SiO_2 60; Na_2O 19.6% in an air plasma flow [13, 14] give preliminary information on the regimes that ensure the needed temperature of the particles. The results of calculations (for particles of size from 25 μm and above) were
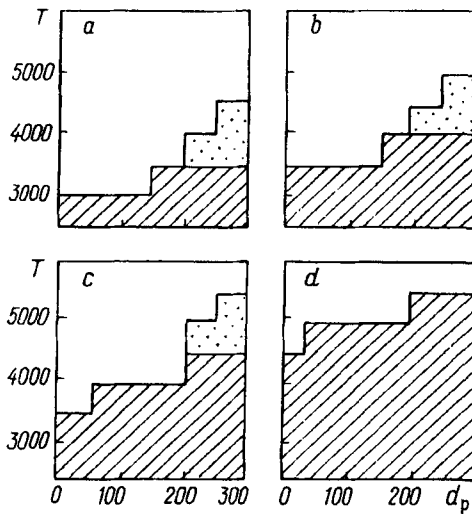
Fig. 2. Plasma temperature (T, K) — glass-forming particle size (d_p, μm) diagram. The hatched regions correspond to parameters at which there is no heating of particles up to 1400 K; unhatched regions, the heating of particles up to 1400 K occurs at a distance smaller than 1 m; regions with points correspond to distances above 1 m. The charge/air mass ratio: a) 1; b) 1.5; c) 2; d) 3) The diagram is constructed from the results of calculations for particles of size 25; 50; 100; 150; 200; 250; and 300 μm.
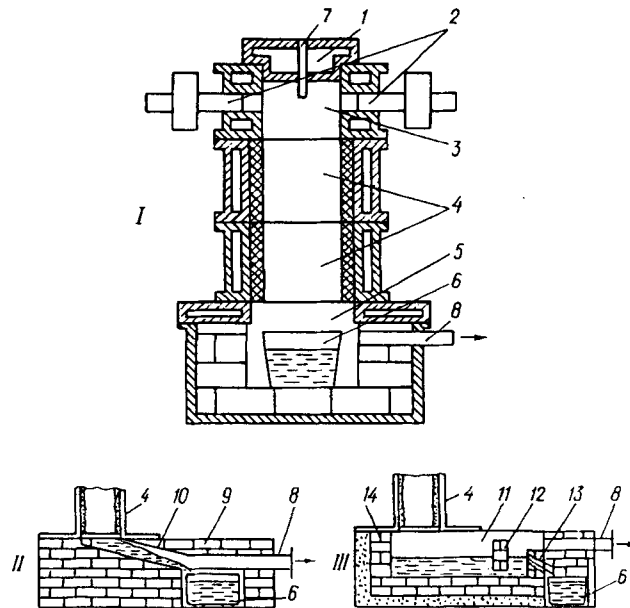

Fig. 3. Schematic of plasma reactor and three variants of a unit for output of the products of thermal treatment used for vitrification of radioactive waste and model charges: 1) lid of the reactor; 2) plasmatrons; 3) three-jet chamber of mixing; 4) channel of sectioned lined reactor; 5) separation bunker; 6) container with vitrification products; 7) supply of raw material; 8) exhaust gases; 9) refractory brickwork; 10) ceramic tube, 11) melt bath; 12) threshold; 13) drainage tube; 14) thermal insulation.

used to construct a diagram in which the hatched portions correspond to combinations of parameters that do not ensure heating up to the vitrification temperatures (>1400 K) for a reactor of any length, the portions with the points are for a reactor of length above 1 m, clear portions are for a 1 m-long reactor (Fig. 2).

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TABLE 2. Entrainment of Components in Plasma Vitrification of Radioactive Wasteand Model Charges

No. of glass in Table	No. of scheme in Fig. 3	Kind of a charge	Discharge of raw material, kg/h	Entrainment, %					
				Na_2O	B_2O_3	^{137}Cs	^{90}Sr	^{60}Co	α-emitters
1	I	Dispersed	20	26.0	24.4	–	–	–	–
1	I	Same	40	12.0	8.1	–	–	–	–
1	I	Same	60	6.5	4.2	–	–	–	–
1	I	Fritted	40	4.0	1.0	–	–	–	–
1	II	Same	40	5.5	8.5	–	–	–	–
1	III	Same	40	2.0	2.2	–	–	–	–
2	II	Same	40	6.3	2.2	–	–	–	–
3	I	Dispersed	40	12.0	7.9	18.8	7.5	5.6	0.6
3	III	Same	40	6.2	4.9	10.5	4.3	3.0	0.2
3	III	Fritted	40	2.7	2.8	4.0	0.7	0.5	0.1
4	III	Same	40	2.4	–	–	–	–	–

With a reactor 0.3 m long it was impossible to obtain a completely boiled and refined glass in a container under the reactor either by changes in the operating conditions or by variations of the charge composition (Fig. 3, scheme I). To increase the residence time of the raw material in the zone of high temperatures, provide homogenization of the melt and to improve the quality of the glass, the vertical channel of the 0.3 m-long reactor was connected in its lower part to an inclined ceramic tube through which the melt, while continuing to contact the high-temperature gas, drained into a container (Fig. 3, scheme II). In another variant, the melt was homogenized in a thermally insulated basin with a passage; the melt was discharged through a special tap hole (Fig. 3, scheme III). If needed, a partition in the basin makes it possible to separate the borosilicate glass melt from the upper layer of the sulfate phase, which has a higher rate of leaching. The heat of the high-temperature gas leaving the reactor provided the heating of the melt bath. The thickness of the melt layer in the bath can be brought up to tens of centimeters, the rate of diffusion of the components to the surface much smaller than that in the channel, and, therefore the melt bath is preferable from the view point of decreasing the escape of volatile components, as was confirmed experimentally (Table 2). As predicted [13, 14], the necessity of using the melt bath is also associated with the difference in the temperatures up to which the differently sized particles are heated on their interaction with the plasma flow, as well as with the possible appearance (under certain operating conditions) of a temperature gradient inside particles of size $\geq$ 200 μm, resulting in subcooling of the core of the particles.

In the presence of small fractions in the raw material, already over the first centimeters in the reactor separate particles may be heated to temperatures greatly exceeding that needed for the process of vitrification [13]. At temperatures of the particles $\geq$ 1500 K the degrees of the entrainment of sodium and its compounds into the gas phase increase greatly [15]. The conditions for the escape of sodium compounds are especially favorable in a dispersed charge, in which particles of sodium nitrate are present individually. By increasing the discharge of raw material, it is possible to decrease the rate of heating of particles and the probability of transition of volatile components into the gas phase. It was found experimentally that raw material loading in the reactor investigated can be brought up to raw material-plasma mass ratios of the order of 1.5–2.5 to attain a raw material-based capacity of up to 50 kg/h. An increase in raw material consumption reduces the escape of sodium and boron (Table 2). Of the radionuclides examined, those of cesium have the highest volatility. Just as for sodium, it is possible to decrease its escape to 4% by fritting or compacting the charge. The losses of cesium in plasma vitrification of RAW are larger than in shaft furnaces and an electric bath furnace, but the distribution of the activity in glass samples is more uniform than in the slags of shaft furnaces [16].

TABLE 3. Composition and Properties of the Products of Plasma Vitrification of Charges for Immobilization of Radioactive Waste

No. of glass in Table 1	Charge	No. of scheme in Fig. 3	Speed of charging, kg/h	Content of components, mass % (according to analysis)							Outer apperance of glass	Density, kg/m³	Rate of leaching, g·cm⁻²·24 h⁻¹						
				Na₂O	Na₂SO₄	CaO	Al₂O₃	Fe₂O₃	B₂O₃	SiO₂			1st 24-h period		30th 24-h period				
													Na⁺	¹³⁷Cs⁺	Na⁺	¹³⁷Cs⁺	⁹⁰Sr²⁺	⁶⁰Co²⁺	α-emitters
1	Dispersed	I	20	18.0	-	18.0	3.6	4.0	6.2	50.0	Inhomogeneous glass-like mass	1820	Not determined						-
1	Same	I	40	18.8	-	17.7	3.5	3.8	7.2	49.0	Same	2030	$2 \cdot 10^{-5}$	-	$2 \cdot 10^{-5}$	-	-	-	-
1	Compacted	I	40	19.1	-	17.6	3.3	3.7	7.2	49.1	Glass with batch stone and bubbles	2150	$1 \cdot 10^{-5}$	-	$1 \cdot 10^{-5}$	-	-	-	-
1	Fritted	I	40	19.2	-	17.5	3.3	3.7	7.4	48.9	Glass with traces of batch stone and bubbles	2200	$8 \cdot 10^{-6}$	-	$5 \cdot 10^{-5}$	-	-	-	-
1	Dispersed	II	40	18.3	-	17.7	3.5	4.0	6.2	50.3	Same	2300	$8 \cdot 10^{-6}$	-	$4 \cdot 10^{-5}$	-	-	-	-
1	Compacted	II	40	18.7	-	17.8	3.5	4.1	6.5	49.4	Glass with small amount of bubbles	2350	$7 \cdot 10^{-5}$	-	$2 \cdot 10^{-5}$	-	-	-	-
1	Fritted	II	40	18.9	-	17.8	3.5	4.0	6.7	49.1	Same	2350	$5 \cdot 10^{-6}$	-	$1 \cdot 10^{-5}$	-	-	-	-
1	Dispersed	III	40	18.9	-	17.7	3.6	3.9	7.0	48.9	Same	2360	$6 \cdot 10^{-6}$	-	$1 \cdot 10^{-5}$	-	-	-	-
1	Fritted	III	40	19.2	-	17.5	3.4	3.7	7.3	48.9	Glass with separate bubbles	2420	$5 \cdot 10^{-6}$	-	$1 \cdot 10^{-5}$	-	-	-	-
2	Dispersed	I	40	17.3	5.3	15.7	3.5	2.9	6.5	48.8	Inhomogeneous mass	2000	Not found						-
2	Same	III	40	15.0	1.6	15.8	3.4	3.0	7.0	54.2	Two phases: lye and glass*)	2370**)	$1 \cdot 10^{-5}$	-	$1 \cdot 10^{-2}$	-	-	-	-
3	Same	I	20	13.3	0.7	19.2	3.4	6.0	6.4	49.3	Inhomogeneous glass-like mass	2150	Not found						-
3	Same	I	40	13.6	0.7	19.1	3.4	5.8	6.9	48.5	Same	2200	$6 \cdot 10^{-6}$	$5 \cdot 10^{-6}$	$3 \cdot 10^{-4}$	$3 \cdot 10^{-4}$	$7 \cdot 10^{-7}$	$8 \cdot 10^{-7}$	$1 \cdot 10^{-7}$
3	Same	III	40	13.7	0.7	19.1	3.4	5.9	7.0	48.6	Homogeneous glas with bubbles	2320	$4 \cdot 10^{-6}$	$4 \cdot 10^{-6}$	$8 \cdot 10^{-5}$	$6 \cdot 10^{-5}$	$4 \cdot 10^{-7}$	$5 \cdot 10^{-7}$	$1 \cdot 10^{-7}$
3	Fritted	III	50	13.8	0.7	19.0	3.4	3.8	7.2	48.4	Homogeneous glass with separate bubbles	2480	$4 \cdot 10^{-6}$	$3 \cdot 10^{-6}$	$7 \cdot 10^{-5}$	$7 \cdot 10^{-5}$	$2 \cdot 10^{-7}$	$2 \cdot 10^{-7}$	$7 \cdot 10^{-8}$
4	Dispersed	III	40	27.0	-	3.9	10.3	6.7	-	52.1	Glass with separate bubbles	2450	$2 \cdot 10^{-5}$	-	$1 \cdot 10^{-4}$	-	-	-	-

*) Composition of glass phase is given.
**) Density of glass phase.

Investigations of the composition and quality of the materials obtained by plasma vitrification (Table 3) were carried out at "Radon" [10] using an S-115 atomic-absorption spectrophotometer and an installation for emissive spectral analysis on the basis of an ISP-30 spectrograph. The radionuclide composition was determined on a Nokia LP-4900 γ-analyzer and a ZnS-based α-detector with an FÉU-19. The rates of leaching of radionuclides from the glasses were determined by the IAEA technique [17]. The amounts of the components entrained (Table 2) were calculated by making up a material balance. Analysis of the results presented in Table 3 indicates that the use of compacted and fritted charges improves the quality of the glasses and decreases the rates of leaching of cesium and sodium from them. Probably, this is associated with a better balance of the components in the glasses produced because of their smaller escape, as well as with the favorable conditions for the occurrence of processes of silicate and glass formation in particules with a close contact of interacting components. The presence of small amounts of undissolved silica in the samples of glasses obtained did not lead to a noticeable decrease in their chemical stability. The requirements on the quality of glass for immobilization of radioactive waste are not as rigorous as for technical glasses, therefore, the available chemical stability is sufficient.

The immobilization of sulfate-containing RAW in glass has specific difficulties associated with the possibility of phase separation in glasses [18]. Experiments with a charge containing sodium sulfate (Tables 1 and 2) showed that despite high temperatures of the gas flow the plasma method in the regimes used does not guarantee the absence of a sulfate phase in the products formed. This is to be taken into account in selecting the means for the treatment of a specific kind of RAW. We note that the radioactive waste of atomic power stations usually contains sulfates in trace amounts and falls into the low-sulfate category.

The energy expenditures in a plsma reactor per 1 kg of glass at a speed of discharge of 30 kg/h was 3−4 kW-h, i.e., 1.5−2-fold larger than in electric bath furnaces [19]. The main advantage of a plasma reactor is the ease of assembly and small overall dimensions. The weight of the working part of the plasma installation does not exceed 1 ton, whereas the weight of a bath furnace of the same capacity attains 5−10 tons. This is essential in vitrification of radioactive waste, since on termination of use the equipment is to be dismantled, treated and buried. In further developments of the plasma technique it is necessary to take into account the possibilities for modification of both the chemical composition of charges meant for the immobilization of RAW and the structure of the material supplied, as for example, the use of the sol-gel process for preparing a homogeneous charge [10].

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