EVACUABLE SMALL-SIZED DIRECT JOULE-HEATED MELTER DESIGNED BY MAYAK PA TO IMMOBILIZE LRW FROM SNF REPROCESSING INTO BOROSILICATE GLASS DEVELOPED AND TESTED AS PART OF A NEW HLW VITRIFICATION COMPLEX

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The article describes the State of the Art in HLW vitrification at the PA Mayak's radiochemical production facility. It presents some conceptual and engineering designs that may further advance the vitrification technology at the enterprise considering various types of liquid HLW. The article discusses the key differences between the promising evacuable small-sized melter and the units previously operated at the enterprise. It overviews its evolvement stages and the results of tests run at PA Mayak in 2018—2020 on the prototypes of evacuable small-sized direct Joule-heated melters designed for borosilicate glass melting. The paper overviews a wide range of the implemented studies and the melters' operational experience. It focuses more specifically on the third improved prototype of an evacuable small-sized melter with its designs developed by PA Mayak in 2022 based on the previously tested prototypes.

Keywords: small-sized melter, evacuable melter, melter designed by PA Mayak, borosilicate glass, vitrification, high-level waste, RT-1 plant, federal target program, radioactive waste.

Introduction

Isolation of large radionuclide inventories generated due to uranium nuclei fission is seen as a most relevant and difficult challenge that should be addressed to promote further nuclear power development [1]. Spent nuclear fuel (SNF) from nuclear power plants is sent for reprocessing and, according to the total activity level, over 99% of uranium fission products resulting from such reprocessing get concentrated in a single waste stream, i.e., liquid high-level waste (HLW).

According to IAEA standards [2], such waste should be immobilized to reduce its volume to the maximum extent possible and conditioned, i.e., converted into a chemically and radiation-resistant form that would remain stable throughout the entire storage period.

Currently, the global HLW immobilization practice provides for HLW vitrification into two glass types: borosilicate and aluminophosphate glasses. The latter one has been applied at PA Mayak in Russia [3].

Development of a new vitrification complex at PA Mayak

At the RT-1 plant, the phosphate glass is produced by non-evacuable electric furnaces of the EP-500 type: heating is provided by industrial frequency current passing directly through the melt. Four similar furnaces were decommissioned at the enterprise after having completed their service life; in 2020, the fifth one, EP-500/5, was shut down

as well. Over two decades of operation (the effective operational lifetime), some 35,000 m^3 of liquid radioactive waste (LRW) was immobilized: by the end of the immobilization process, this waste inventory resulted in over 8,000 tons of highly active aluminophosphate glass with the total radionuclide activity amounting to over 800 million Ci (see Table 1) [4], [5]. According to their designs, these furnaces were not intended to be dismantled at the post-decommissioning stage, which is why this equipment still remains under supervision as part of the corresponding building. Advanced approaches to LRW vitrification should provide for spent melter dismantling, its fragmentation and final disposal of most highly contaminated melter's parts together with solidified HLW. Table 1 summarizes data on the operation of EP-500 type HLW vitrification furnaces at PA Mayak.

Commissioning of the sixth production electric EP-500-type furnace with a design service life of 6 years, glass production capacity of up to 450 t/year (60 kg/h and 250 l/h according to the initial solution) and fluxing additive introduction in a liquid form is expected in 2025. The EP-500/6 electric furnace is supposed to become the last non-evacuable melter of this type. It should be noted that large-sized high-performance electric furnaces designed for LRW vitrification have been operated only in Russia and the United States (at the Hanford site) [6].

Therefore, the following tasks contributing to further advancement of vitrification methods remain relevant for the radiochemical industry:

1) capacities enabling the solidification of all generated liquid HLW types should be provided;

2) methods providing maximum compaction of the solidified waste should be introduced, thus, in particular, vitrified HLW from the reprocessing of foreign SNF may be returned to the countries of origin;

3) small-sized evacuable units seen to replace non-evacuable melters should be introduced.

To address the above problems, a new multipurpose vitrification complex is going to be commissioned at the PA Mayak site in 2028: the facility will be able to treat all kinds of liquid HLW. Figure 1 shows its layout within the RT-1 flowchart [7].

Facility designs provide for two removable EP type melters producing aluminophosphate glass seen as a more advanced version of the EP-500 design. These units are required to process "challenging" HLW of complex composition accumulated

Table 1. Operational results considering HLW vitrification by EP-500 type furnaces at PA Mayak

Name	EP-500/2	EP-500/1r	EP-500/3	EP-500/4	EP-500/5
Operational period	1987–1988	1991–1997	2001–2006	2006–2010	2016–2020
Vitrified HLW inventory, thousand m^3	1	11.5	8.0	8.1	6.6
Produced glass, t	162	2,200	1,800	2,040	1,898
Immobilized radionuclides, million Ci	4	282	175	182	167

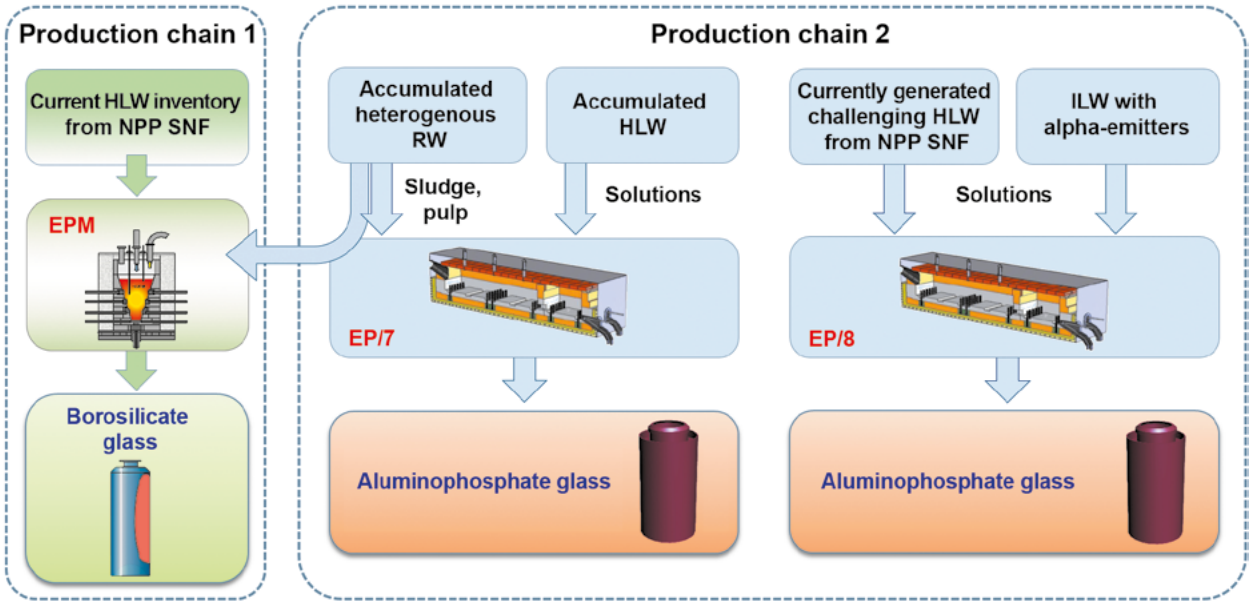

Figure 1. Layout of the new vitrification complex at the PA Mayak site

due to past operations at the plant, as well as some solutions from the ongoing reprocessing of SNF from power reactors (VVER-440, VVER-1000, BN-600, BN-800). A newly developed small-sized melter with direct electric heating on borosilicate glass was proposed to solidify most of these liquid HLW streams (mainly highly active raffinates from hydrometallurgical processes): its operation is expected to considerably increase the fraction of radionuclides immobilized into the waste form (compared to aluminophosphate glass).

Dismantling, disassembly and disposal procedures will be presented in the design documentation developed for all of these electric furnaces providing compliance with the environmental legislation and reducing the costs required to develop new installations, since considerable part of already available infrastructure is going to be reused [5], [7].

Thus, in the next decade, vitrification in direct electric heating furnaces is still going to remain the main HLW solidification method in Russia, nevertheless, providing much broader choice of applied glass types and immobilized waste characteristics.

Evacuable small-sized melter with bottom drain designed by PA Mayak

At present time, there are two basic glass melting methods implemented globally for LRW solidification purposes: with or without calcination at the pre-melting stage, which are conventionally called two- and one-stage processes. The former one is implemented in France and the UK, whereas the latter one — in the US, Russia, Germany, Japan, India and South Korea [6].

At present time, the two-stage process is not implemented in Russia due to relatively low reliability of the equipment interacting with heterogeneous media and inability to perform the required maintenance operations in the event of any failures.

In practice, all waste vitrification processes requiring high temperatures in the range of 1,000—1,250 °C have been running based on electrical heating, which is done either by passing a current through a glass melt (direct or Joule heating) or by inducing high-frequency currents from an external source. Directly heated melters are used in the US, Germany and Russia [8].

It should be noted that small-sized direct Joule-heated melter designs with a vertical layout and bottom drain are currently viewed as a relevant area of research for the global nuclear industry by almost all key SNF reprocessing enterprises. Thus, vertical direct Joule heating melters with a glass melt production capacity of 9 and 52 kg/h have been introduced in the designs of the Japanese Tokai and Rokkasho (RRP) Reprocessing Plants, respectively. In India, small-sized melters of the AVS type have been introduced at the Tarapur plants and WIP in Trombay [9]. A vertical melter has been operated at the Defense Waste Processing Facility (DWPF) in Savannah River (USA) treating the accumulated LRW inventory generated from defense programs implemented in the past [10]. The most advanced melter designs of this kind are the German VEK units developed by the Institute for Nuclear Waste Disposal of the Karlsruhe Institute of Technology (KIT-INE). These melters have immobilized up to $7.7 \cdot 10^{17}$ Bq of liquid HLW with a relatively high content of noble metals resulted from the operation of a pilot SNF reprocessing plant, namely, the Wiederaufarbeitungs-anlage Karlsruhe (WAK) facility [11]—[13].

In 2015, PA Mayak has tasked its staff, designers, researchers, computing and production engineers with the development of an evacuable small-sized melter with a bottom drain of a new design. The following requirements had to be accounted for the in the designs:

- small size (relative to 130-ton furnaces of the EP-500 type);
- the designs should provide for further dismantling, disassembling and removal operations;
- the designs should support the vitrification of HLW high in noble metals;
- high reliability (service life of at least 3 years).

From 2018 to 2019, the first prototype of the evacuable small-sized melter was tested at the PA Mayak's radiochemical plant, which resulted in a huge array of data on its electrical and thermal parameters. Figure 2 presents the general layout of the experimental bench designed for the first prototype testing.

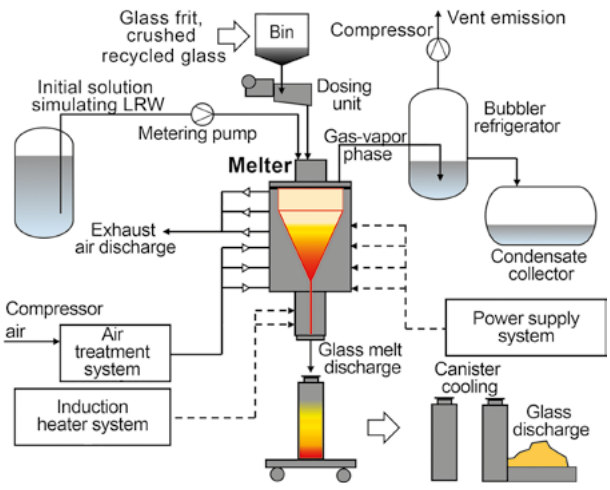

Figure 2. General layout of the experimental bench designed for the first melter prototype testing

The experimental bench includes:

- melter — a direct electric heating furnace with several pairs of main electrodes, a bottom electrode with two current leads, a drain die and an induction drain die heating unit;
- glass frit and model solution dosing systems;
- air cooling system;
- compressed air preparation system;
- gas exhaust system;
- starting heating system.

Figure 3 shows the first prototype of the evacuable small-sized melter, i. e., its schematic design layout.

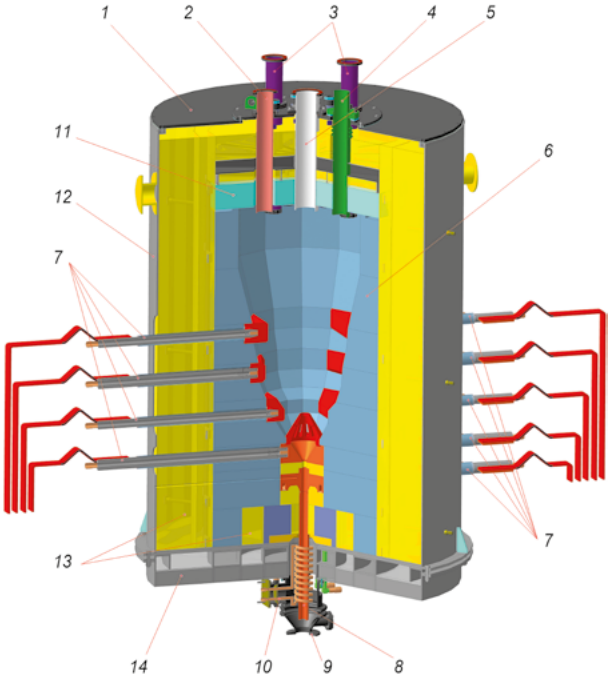

Figure 3. Schematic design layout representing the first prototype of a removable small-sized melter designed by PA Mayak: 1 — lid, 2 — thermocouple pipe, 3 — glass frit feeder, 4 — gas duct, 5 — solution feeder, 6 — melting zone made of ceramic refractory, 7 — current leads with electrodes, 8 — die, 9 — inductor body, 10 — inductor, 11 — melter roof, 12 — melter body, 13 — thermal insulation, 14 — melter footing

The first melter prototype was a small-sized direct Joule-heated vitrification furnace with a maximum power of 250 kW.

Its funnel-shaped fireproof melting zone consisted of ceramic blocks made of aluminum oxide and silicon with some additional double thermal insulation. The melting zone capacity accounted for some 0.5 m^3. The melt surface area and the melt volume accounted for 0.49 m^2 and 0.24 m^3 respectively. Three pairs of electrodes were fitted within the melting zone. A funnel with a protective device preventing the drain channel clogging and the accumulation of a solid phase insoluble in glass was fitted at its bottom. A bottom electrode passing onto the drain die was installed underneath it through a ceramic insert. All electrodes were produced from corrosion-resistant nickel alloy KhN70Yu; the outer melter body — from 12Kh18N10T steel. The electrodes and the furnace bottom were cooled with dried compressed air. Each of the three electrode pairs was powered via a separate transformer. Another transformer was reserved for powering the "bottom electrode — drain funnel" pair; it was also used when starting the melter to power the starting heater assembly installed in the roof penetrations. Then these heaters were removed and the penetrations were used to install necessary measuring tools and automation devices, a feeder supplying the initial solution and the glass frit.

Figure 4 presents the external appearance of the experimental bench.

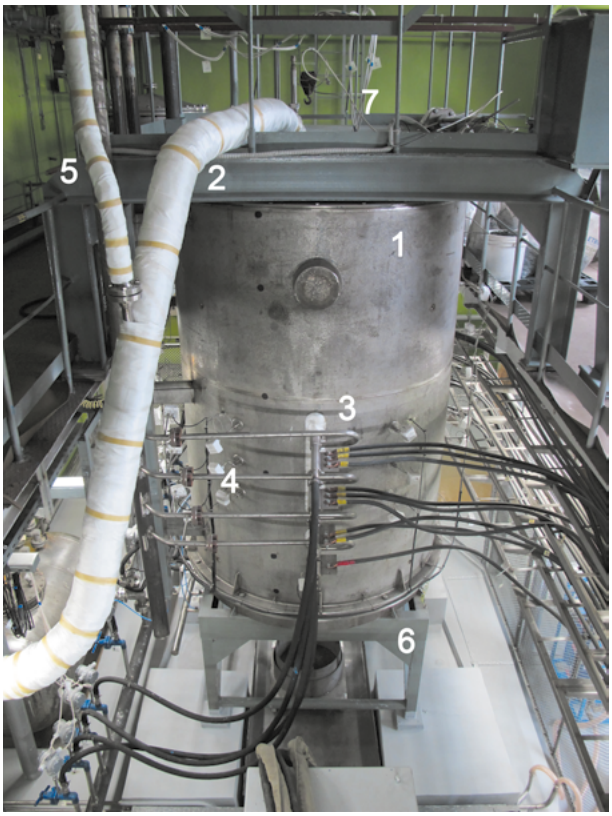

Figure 4. Experimental bench fitted to test a melter designed by PA Mayak: 1 — melter body; 2 — gas duct; 3 — current leads; 4 — cooling system for current leads; 5 — backup gas duct; 6 — melter footing; 7 — service area

To discharge the glass melt from the melter, it was necessary to provide controlled melting of the glass plug in the die of the draining device, which was done in two stages. First, the die was heated to temperatures from 1,000 to 1,100 °C using a water-cooled induction heater operating at a power of 70 kW. Next, voltage was applied to the die and the

bottom electrode. As the glass melt melted, conductivity emerged in the channel between it and the funnel. Thus, further on, the glass plug was heated via the Joule heating. Its melting was triggering the drain device to open and then the glass melt could be dispensed into a drum.

To discontinue the process, the die and the glass melt passing through it were cooled rapidly by supplying air into the bottom electrode cavity and blowing its outer surface from a pipe into the gap between the inductor turns.

Thus, the glass melt pre-draining operations were quite time-consuming which is viewed as a considerable disadvantage of the first prototype. The bottom electrode had to be heated for a long time (over 10 hours) to make the drain device ready for operation. After this, drainage could be implemented in a short time period (tens of minutes), nevertheless, requiring some precise control over the electric current in the drain channel and its parameters, since glass melt overheating was still possible, i. e., the recommended temperature of 1,100 °C could be exceeded in the metal elements of the drain channel.

In general, the first small-sized melter prototype and its operation provided data on electrical and timing parameters considered optimal for the electric furnace control system; preliminary (starting) heating of its working space with loaded glass frit was launched using starting silicon carbide heaters; the process involving borosilicate glass melting and fluxing was investigated. All electrodes were also tested to evaluate their performance under different switching options. The first prototype unit operated for a total of 200 days.

Based on these data, the second modified melter prototype was manufactured and tested in 2019—2020. Primarily, its bottom section has been upgraded: the inductor was lifted a few centimeters higher to provide uniform heating of the drain die along its entire length, and the cooling system of the bottom electrode was improved, the channel volume between the drain funnel and the bottom electrode was increased, and a new die blowing system was introduced, i. e., a hollow cylinder with nozzles surrounding the inductor.

Figure 5 shows the design layouts of the drain devices developed for the first and the second prototype of the evacuable small-sized melter.

An additional element was added to the melter design — an internal cylindrical shell made of heat-resistant steel fitted around the masonry, which was interlayered by a fireclay-quartz backfill. Thus, the service life of the sample could be extended and the glass melt locking along the masonry seams and the heated structural elements could be improved. No changes have been introduced to the designs of

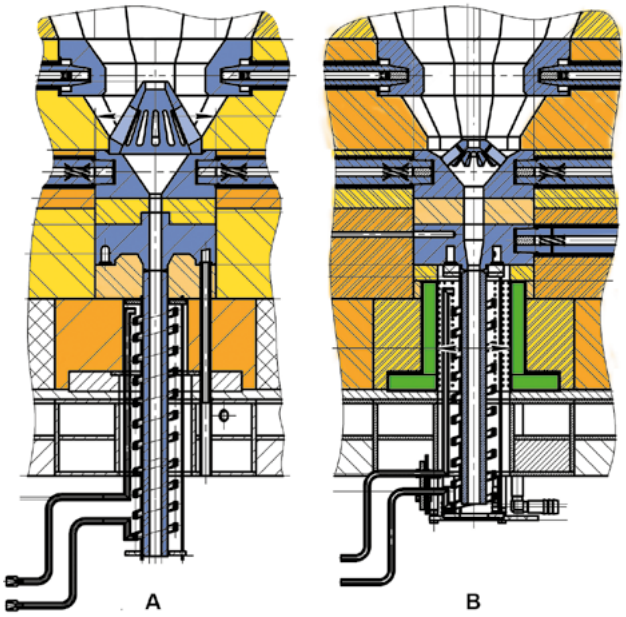

Figure 5. Schematic design layout of the drain devices fitted in the first and the second prototype of the evacuable small-sized melter designed by PA Mayak: A — the first prototype; B — the second prototype

the experimental bench used in the second prototype testing.

The maximum evaporation water capacity was measured for this small-sized melter, i. e., it accounted for 25 dm^3/h. Temperature parameters of the melter and the gas phase were monitored while metering the water volume supplied through a cooled feeder in the center of the roof with visual observations of the melt surface performed through a backup penetration in its lid. To reach stable indicators, each stage lasted for 12 hours. Evaporation efficiency was calculated based on the melt and gas phase temperatures, as well as the area of the cooled glass spot on the melt surface.

The estimated melter performance as regards the initial solution with fluxing additives (144 g/dm^3 of soda and 36 g/dm^3 of borax) amounted to 15 dm^3/h, and its performance according to frit production accounted for 10.5 kg/h assuming simultaneous dosing.

Optimal conditions for launching and discontinuing the glass melt drainage have been identified for the second melter prototype. Figure 6 presents its thermophysical parameters observed during these operations.

Before the glass melt was drained, the temperature in the gas phase of the melter accounted for less than 950 °C. To drain the second prototype, preliminary operations had to be launched approximately 10 hours in advance: the air-cooling system was removed from the bottom electrode and this element had to be heated gradually from 300 °C to 500—550 °C. Then, 3 hours before draining, the

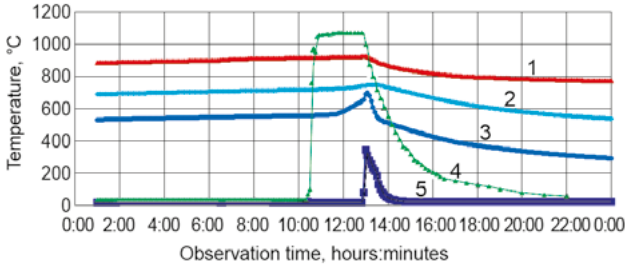

Figure 6. Thermophysical parameters of the melter along the glass melt draining process (temperature, °C): 1 – vapor-gas phase above the melt; 2 – drain funnel; 3 - bottom electrode; 4 – on the die; 5 – cooling air in the funnel cavity

inductor was turned on, the die was heated to 1,050—1,100 °C and this temperature level was maintained until the draining process started. Heat transfer from the die resulted in glass plug heating between the bottom electrode and the drain funnel.

Figure 7 presents the electrical parameters inherent in the second prototype at the draining stage. The first (top) pair of electrodes was powered by T-1 transformer, the second and third — by T-2 and T-3, respectively, T-4 was reserved for current supply to the "drain funnel — bottom electrode" pair. Along the operational stage, the voltage on the electrodes was adjusted. Weak current between the funnel and the bottom electrode in the second prototype started passing approximately two hours before the draining started: at the point when the glass plug resistance dropped to a sufficient level along its heating and gradual melting. By the time the draining started, the current in the gap between the bottom electrode and the funnel reached 70 A at a voltage of up to 36 V.

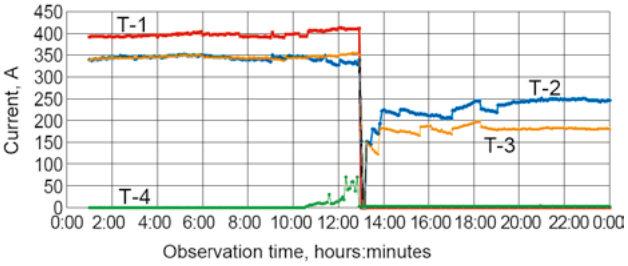

Figure 7. Current supplied to the melter electrodes along the glass melt draining process

In case of the selected glass melt composition, draining started exactly when the bottom electrode temperature reached 654 °C. Then it raised to 690 °C. The process was discontinued by supplying air to the annular cavity of the bottom electrode; the air temperature at the outlet reached the maximum level of 340 °C; after 30 minutes of blowing, it dropped gradually to 180 °C. However, reliable discontinuation of this process has required to turn the die blowing on. Typically, it could be cooled from 1070 °C to ~ 700 °C in 3—5 minutes.

At the beginning of the draining process, the temperature in the funnel accounted for 741 °C with the maximum temperature of 750 °C reached 20 minutes after turning on the air supply for die blowing and cooling. Then a gradual decrease to 530 °C could be observed by the end of the day.

In general, up to 10 minutes were required to discontinue the draining in the second prototype unit: gradual thinning of the glass stream was observed followed by a few threads and drops trickling down from the semi-hardened glass melt. The main factors affecting the discontinuation rate were the melt temperature, the glass viscosity and the rate of the cooling air flow supplied to the die.

No current flow was required between the drain funnel and the electrode constituting to an electrode pair enabling heating at the draining stage. Nevertheless, this option has been tested as well (for the upper and middle pairs of the electrodes). In the course of the process, transformer T-1 was turned off (see Figure 7), since the upper pair of electrodes powered by it tended to get exposed. T-4 was also turned off immediately after the draining process started. Then, the T-2 and T-3 voltage was increased since it was necessary to make up for the disconnection of the first electrode pair.

Figure 8 shows how the borosilicate glass was drained from the die in the second evacuable small-sized melter prototype.

A series of three drains was performed under the study providing for a stepwise increase in the viscosity and the glass melt melting points adjusted via varied metering of the glass frit with no fluxing

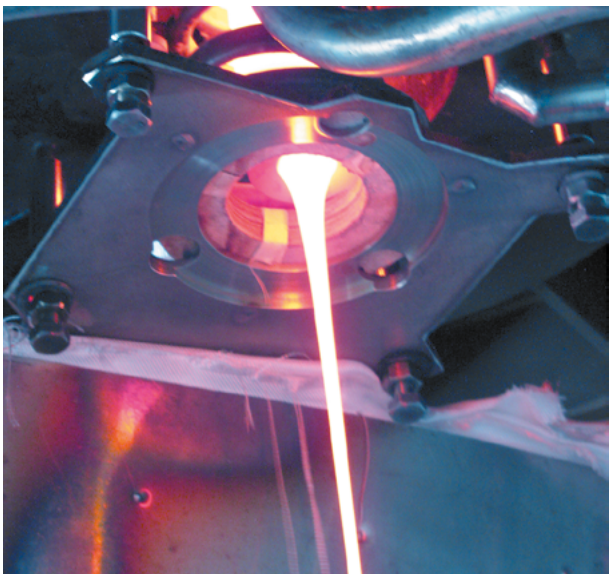

Figure 8. Borosilicate glass draining from the die constituting to the second prototype of the evacuable small-sized melter designed by PA Mayak

solution being involved. It was noted that it took more time for the drainage to start and the rate of the melt flow from the die has dropped considerably.

Two experiments have been implemented as well: metal-spherical powder made of stainless-steel grade 06hN28MDT with a particle size ranging from 0.40 to 0.63 mm was introduced into the glass mass simulating the dispersed phase of noble metals contained in HLW from SNF reprocessing. It has been established that at the draining stage, the fine powder appeared to be fairly evenly distributed in the glass melt; the sedimentation time of the powder particles in the melter accounted for less than 4 hours. The bottom drain device provided stable removal of metal particles from the melter along with the melt flow.

The study indicated the capacity of the vibration dosing system for dry reagents and the glass frit providing adequate dosage of the required weighed reagents within certain time intervals.

The second prototype unit operated continuously for 185 days. Based on the findings from the first and second prototype testing, as well as the data on their structural elements investigated after the melters were disassembled, designs of the third small-sized evacuable melter prototype were developed in 2022.

The designs of its masonry, body, lid, thermal insulation, electrodes and drainage device have been considerably upgraded.

Table 2 provides a comparison between the key performance indicators of small-sized melters of the first, second and advanced third generation.

Table 2. Comparison of the key performance indicators inherent in the first, second and third prototypes of the small-sized melters designed by PA Mayak

Parameter	First & second melter prototypes	Third melter prototype
Total capacity of the furnace bath, m^3	0.50	0.56
Working capacity of the furnace bath, m^3	0.24	0.40
Melt surface area, m^2	0.49	0.43
Estimated max. performance according to the initial HLW solution, dm^3/h	15	18–20
Melt capacity, not less than, kg/h	8.2	15
Volume per single drain, dm^3	35	75
Total electrical power, kW	250	165
Estimated melter weight, t	12	18*

* taking into account the removable external thermal insulation

Table 3 compares the borosilicate glasses used as a loading material in the melters. Based on the findings from the first prototype testing, the borosilicate glass composition was modified seeking to reduce the glass melting temperature and achieve well-controlled drainage. The resulting composition was used to test the second prototype melter. These efforts were followed by the development of a low-melting borosilicate glass [14] designed to meet the requirements set out in NP-019-2015 [15]. As a result, a new composition of borosilicate glass was proposed to test the third melter prototype designed by PA Mayak. Its composition is presented in Table 3.

Table 3. Composition of borosilicate glass used to test melter prototypes designed by PA Mayak

Prototype	Mass concentration of element oxide, wt. %							
	SiO_2	B_2O_3	Al_2O_3	CaO	Na_2O	MgO	Li_2O	TiO_2
1	49.1	21.5	4.1	4.9	19.9	0.6	–	–
2	44.5	21.4	3.9	4.5	25.1	0.6	–	–
3	48.2	22.1	4.2	4.9	16.0	0.6	2.0	2.0

For the third prototype, the glass melt volume that could be drained out from the improved melter till reaching the minimum operating level was increased to 75 dm^3: two drains were enough to fill the entire capacity of the new stainless container (canister), the dimensions of which corresponded to those of a widely applied CSD-V canister. Unlike the first and the second small-sized melter prototypes, in case of the third melter design, changes in the glass melt level contributed in no way to relevant changes in the melt surface area.

To provide reliable containment of the glass melt, the third prototype features a new multi-layer masonry made of chrome-corundum refractory. At the same time, the block geometry has been considerably upgraded with some simplifications introduced to provide better manufacturability. The melter designs are also expected to provide the feeder and the starting heaters with interchangeable block plugs.

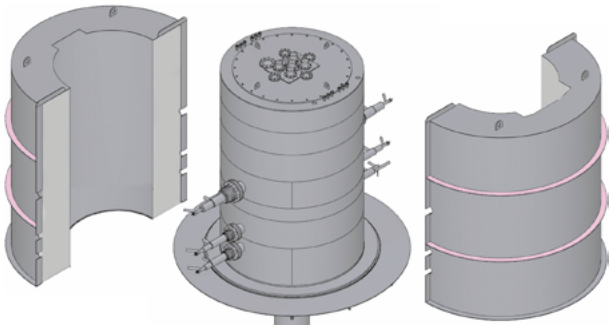

Figure 9. Schematic layout of the third small-sized melter prototype designed by PA Mayak fitted with removable heat protection

Its external thermal insulation should provide for two removable modules that can be reused in subsequent melters. Figure 9 shows the schematic layout of the melter internals and the removable thermal protection.

The main design solutions implemented in these three small-sized evacuable melter prototypes developed PA Mayak have been copyrighted [16].

Conclusion

Since 2015, PA Mayak has been engaging its engineers in the development of evacuable small-size melter designs. The new melter fitted with bottom melt drainage was designed to condition HLW resulting from the reprocessing of SNF from power reactors via its immobilization into borosilicate glass. In 2018 – 2020, long-term studies were launched to test the first and the second melter prototype units developed by PA Mayak.

Based on these findings, the third removable melter prototype was developed with upgraded designs and materials of the refractory furnace masonry, glass frit and solution supply units, individual elements of the cooling systems, the drainage device, melt level and temperature control system. The third small-sized melter prototype designed by PA Mayak is going to be tested in 2024 and is supposed to become part of a new vitrification complex at the PA Mayak site being established under the Federal Target Program Nuclear and Radiation Safety in 2016 – 2035 (FTP NRS-2) [17]. Its commissioning is scheduled for 2028.

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