#### Design and Construction of Integrated System of Bench-Scale High-Temperature Waste Melting/Vitrification Units at SIA Radon - 10141

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#### ABSTRACT

A new integrated bench-scale facility consisting of a plasma calciner (PC), a submerged plasma melter (SPM) and a cold crucible (CC) has been designed and constructed at SIA Radon. Combined PC/SPM and PC/CC units are perspective for melting/vitrification of liquid radioactive wastes (LRW) whereas the SPM is a promising unit for treatment of solid radioactive wastes (SRW). Plasma calciner is a shaft-type reactor for spray dehydration, denitration, and calcining of LRW with a salt concentration of up to 400 g/L in a single unit using impinging streams of high-temperature gases generated by two plasma torches and off-gas heat. Calculated LRW capacity of this unit is of 15 to 30 L/hr. Efficiency of the reactor is estimated to be  $\sim$ 70%. This reactor may be recommended as a preliminary stage of LRW vitrification in SPM or CC. The SPM process is based on bubbling of gas heated by plasma torches through melt (submerged combustion/melting). The "dog-house" melter has been designed and constructed. The melter consists of a case forming a melt bath, a batch feeder, a melt pouring unit, a melter cover, a melt collector with a sealing unit, a melter rake frame, and an off-gas umbrella. The melter is equipped with an off-gas system. Calculated melt productivity is up to 10 kg/hr. Calculated melter efficiency ranges between 30% and 47% depending on water content in the feed. Calculated energy expenses ranges between 0.85 and 3.5 kW per 1 kg of melt produced. The melter is equipped with an arc plasma torch having a thermocathode. This facility will be used for testing and trial of perspective waste treatment technologies to be implemented at industrial scale.

### **INTRODUCTION**

Development of new high-temperature processes for treatment of liquid (LRW) and solid radioactive wastes (SRW) producing waste forms with high chemical durability, radiation resistance and mechanical integrity is currently a key challenge of waste management problem. Application of high-frequency and plasma heating sources provides for high-efficient heat and mass transfer in reaction units opening the way to designing and construction of high-capable waste treatment facilities. Since late 1970s SIA Radon was being designed various high-temperature waste treatment processes using flame and plasma heating [1-4]. Plasma-activated incinerators and pyrolysers have been implemented for treatment of SRW. The combined PC/PM process for LRW vitrification was being developed at SIA Radon since early 1980s [1,2]. In late 1970s works on plasma treatment of SRW were started. By 1994 a pilot plant based on plasma shaft furnace has been constructed and tested [3,4]. The plasma shaft is multizone furnace. Temperature in the upper part of the shaft is 150-250 °C at an underpressure of as many as 200 Pa and increases towards to the bottom part achieving ~1400-1500 °C in the melting zone. SRW is fed into the shaft and undergone to drying and decomposition. Organic constituent is subject to gasification, pyrolysis, and burning. Inorganic constituent is melted and molten slag is homogenized and poured into canisters [3,4]. Later the shaft furnace was upgraded and modified. Up to date two plasma units with SRW capacity of 40-50 kg/hr and 200-250 kg/hr have been designed and constructed. The shaft 2.5 m in height is heated by two 150 kW plasmatrones and temperature in the melting (bottom) zone achieves 1500-1700 °C. Molten slag is poured into canisters to be disposed in a repository after product solidification. During active operation campaign SRW in amount of more than 32 metric tons (~180 m<sup>3</sup>) has been treated. Similar demonstration plant for treatment of non-radioactive municipal solid waste with a capacity of up to 500 kg/hr has been constructed and commissioned in Haifa, Israel.

For vitrification of high level LRW liquid-fed Joule heated ceramic melters are used in Russia [5] and USA [6,7]. Cold crucible inductive melter (CCIM) is considered as an alternative in Russia [8], France [9], USA [10], and some other countries. The full-scale CC-based plant is used at SIA Radon for low-level LRW vitrification [11]. Major advantages of the CC over other melters are higher specific productivity, longer lifetime, and smaller overall dimensions facilitating its dismantling and disposal in the case of failure. Some higher productivity of the CC may be achieved using pre-drying and calcining of LRW or/and feed. Therefore, application of small-sized dryers/calciners is required. Among their various versions plasma-, inductive- and microwave-heated units are mostly considered.

The Experimental Works and the Division of High-Frequency and Plasma Processes for Waste Treatment at SIA Radon deal with design, construction and testing of plasma calcining (PC), plasma melting (PM), and CCIM units as well as processes using microwave heating. This paper briefly summarizes major topics of activity of the Division of High-Frequency and Plasma Processes for Waste Treatment and current situation in implementation of the bench-scale units for SRW and LRW treatment at SIA Radon.

# PLASMA CALCINATION

Plasma reactor for spray drying, denitration and calcination of LRW with a salts concentration of up to 400 g/L using impinging streams of highly heated gases generated by two plasma torches (plasmatrones) and off-gas heat has been designed and constructed (Figure 1). Major advantage of this unit is much smaller off-gas volume as compared to fluidized-bed or fuel-injecting spray calciners.



Figure 1. Scheme (left and middle) and view (right) of the plasma spray calcination reactor.

The frame of the PC reactor consists of the lower rectangular part lined with refractory and heatinsulating materials and upper cylindrical channel reactor with outer spray cooling, a cover with spray injector, and a pneumatic mechanical vibrator. Off-gas is conducted outside tangentially. The frame of the reactor may be joined to optional equipment such as plasma torches and portable bin with a sealing unit for collection of calcine. The cover has water-cooled contour as a pipe around the edges. The lower part of the reactor is disposed after completion of service life.

The injector provides for high-efficient spraying of LRW. Outlet nozzle is equipped with a circular chamber to intensify aerodynamic jet spraying by means of compressed air. The injector may be moved along the axis of the reactor channel using lengthening pipes for feeding of LRW and compressed air from an input/output collector.

Calculated LRW capacity is 15-30 L/hr. Temperature in the upper part of the shaft ranges between 150 and 250 °C. Droplets size at the nozzle outlet ranges between 50  $\mu$ m and 150  $\mu$ m. Underpressure in the reactor is as many as 200 Pa. A power spent for calcination process in the reactor determined from the heat balance was estimated to be 65-70% demonstrating high efficiency of the PC process.

Application of the PC reactor allows recommending a calcination as a preliminary step before vitrification. The spray PC reactor may be installed above the CC thus heating surface of melt in the CC by plasma torch; at that pre-heated calcine (or frit/calcine mixture) is fed directly onto a melt surface at its highest temperature. The pre-calcination step eliminates aerosol exhausts being typical of liquid feeding at the current CCIM process.

## PLASMA MELTING

Plasma heated furnaces are applied in the metallurgy, power industry and waste treatment. As a rule, the furnace is a sealed chamber with a melting zone lined with refractory and energized from one or more plasma torches. The material treated is converted to a melt and poured in molds or canisters [12].

Analysis of the technologies used shows that an increase of specific process productivity is achieved by intensifying of external heat-transfer from a heat source to melt. Melting of low thermal conductive materials requires high temperature gaseous stream as a heating agent. Small-size bath may be heated by submerged plasma torch at formation of necessary hydrodynamic and thermal conditions in pre-combustion zones of high temperature gaseous stream. At that, melt productivity may be raised by increase of melt surface area, melt depth and input power without limitations.

The productivity of melting units is mainly determined by a time spent for completion of the slowest stage – dissolution of highly-fusible oxides in initial melt. Liquid slagging requires knowledge of molten slag viscosity and its dependence from melt chemical composition. It has been shown that addition of some fluxes in amount of 10-15 wt.% affects rheological properties of the molten slag according to the computational procedure proposed, reduces liquid slagging temperature and provides for required technological and physical-chemical properties of the final product. Forced intermixing of the melt causes violent acceleration of dissolution of highly-fusible feed constituents. Melt bubbling with high-temperature gaseous stream raises thermal efficiency of the melting process and intensifies all the heat and mass transfer processes in the melt.

To realize the technology proposed for melt production from highly-fusible oxide materials at melt bubbling with submerged plasma jet a new dog-house type melter has been designed and manufactured. The calculations of the melter design and geometry are described in details in our report [13].

The PM consists of a melting pot, a feeder, a pour unit, a cover, a melt collector with pour sealing unit, a melter inclination frame, and an off-gas pipe (Figure 2). Overall dimensions of the melter are  $580 \times 430 \times 600$  mm, melt bath volume is 30 L, calculated melt productivity is up to 10 kg/hr, cooling water flow rate is up to 5 m<sup>3</sup>/hr. Temperature in the melter may be widely varied (1250-2200 °C).



Figure 2. Scheme (left and middle) and view (right) of the dog-housing plasma melter.

The melter body is fabricated from demountable water-cooled dog-housing panels and a slab with a water-cooled port for a plasmatrone. Intensification of cooling of the melter wall and improvement of its design characteristics are achieved by the use of a Field's pipe as a furnishing structure. Resulting heat transfer coefficient in the Field's pipe is by 2 times higher in comparison with one-fold water flow through the pipe. The plasmatrone is fixed on either standard joint or by means of bayonet connection. The pouring unit is manufactured as a cavity with a pour spout in the side wall of the dog-house. The cavity is stud in the upper part, surrounded by mobile cylindrical shell with demountable cover, water-cooled pour stopper holder, and pouring channel. The pouring unit is connected and sealed to the melt collector and has looking window.

To reduce heat losses from the water-cooled wall and, therefore, increase of thermal efficiency for the melt bath heated by high-temperature torch a layer of corrosion resistant and heat-insulating material is required.

The melter body is equipped with a demountable cover with contact water-cooling pipe with a "snake" type profile. The melter has a channel above the melt surface for dilution of off-gas. An umbrella is used for removal of off-gas. Coating of the melt surface with feed layer decreases significantly loss of radionuclides and other volatile constituents. The off-gas pipe is equipped with a thermocouple. Batch feeding ports are located on the side panel of the dog-house.

Efficiency of the melter was estimated to be between 30% and 47% depending on water content in the feed. Calculated heat expenses for melt production (melting ratio) range between 0.85 kW×hr/kg and 3.5 kW×hr/kg. Lower value corresponds to melting ratio at production of borosilicate glass from the feed with a water content of 15 wt.%. Major heat losses are due to cooling of heat-stressed elements – plasmatrone electrodes (25-35% of total input electric power) and dog-house melter walls (9-13%).

## **MODIFICATION OF PLASMATRONES**

To implement PC and PM/bubbling technologies reliable heat sources with long lifetime are required. Design of the plasmatrone with a thermocathode is similar to that designed before by Institute of Heat and Mass Transfer of the Academy of Sciences of Belarus [14], but free of some disadvantages. So, water cooling of anode in the place of sealing gasket location was improved by means of water stream, sealing of water cooled chamber was improved due to additional cone-to-cone fitting. These measures were targeted to increase of plasmatrone lifetime. Moreover, joint of the plasmatrone to the melter body was

improved by application of bayonet connection. Longer anode provides for pass of the plasmatrone body through the width of lining and overrun of plasmatrone face into the operational space of the furnace. With the aim of cost reduction the cathode was manufactured on the basis of commercially available article – thermocathode for plasma cutting. Cathode and anode geometry was also changed for the conditions of the use of thermocathode with small commercially available power sources of stabilized direct current with a power of up to 30 kW.

## COLD CRUCIBLE MELTING

Another promising melter for RW melting/vitrification is CC. Full-scale CC-based facility is used for low-level LRW vitrification [11]. However some problems occurred at CC melting require their investigation in more details and a new bench-scale CC based unit is called to solve them. The unit is energized from a 1.76 MHz/60 kW generator. Electrical parameters of the CC by the full magnetic flux method have been calculated and a design procedure for demountable CC providing for melting runs at non-stationary batch melting has been developed. Moreover, from ~200 to ~350 mm inner diameter cold crucibles with improved pouring unit and cooling system have been designed and manufactured and their cold testing has been performed.

Numerous tests on production of glasses, glass-ceramics and ceramics potentially suitable as waste forms using a lab-scale unit equipped with a 56 mm inner diameter CC energized from a 5.28 MHz/10 kW generator (Figure 3a) were performed. Major process parameters (weight of the fed batch and produced material, feed capacity, material productivity, current, voltage, input power, and melting ratio) were determined. Various stages of the CCIM process are shown on Figure 3*b*. Photo *1* shows melt surface coated with fresh portion of batch, on photo *2* the batch is partly reacted, on photo *3* major batch is reacted, and photo *4* demonstrates that nearly all the batch portion is reacted and surface is almost clear.





Left: 1 – cold crucible with inductor, 2 – process box, 3 – high-frequency generator, 4 – off-gas umbrella and pipe, 5 – fan, 6 – HEPA filter. Right: 1 – just after feeding of batch portion, 2 – in 2-4 min., 3 – in 5-7 min., 4 – in 8-10 min.

Method of monitoring of melt depth in the cold crucible using an infrared scanner has been developed. Thermal profile shows zones with elevated temperature indicating non-uniformity of its cooling, which can be caused by clogging of cooling pipes due to salt deposits that is important to prevent failure.

Analysis of malfunctions during the CCIM process shows that most of them are due to destruction of cooling elements and sealing protective putty. All the malfunctions are divided on "external" due to formation of electric arc between the crucible and inductor and "internal" due to thermal destructions and electrochemical corrosion (Figure 4). Technical measures to eliminate reasons of failure have been proposed. One of them is application of separate cooling line for the most heat-stressed elements, obeying to specific conditions of CC start-up, melting run, and melt pouring. The gaps between the CC forming pipes should be filled with special sealing putty.



Figure 4. View of inductor and CC in the case of failure: a, b – due to electric arc between inductor and crucible wall; c – due to overheating, d – due to clogging of cooling pipes with salts.

## AUXILIARY EQUIPMENT

The facility has various auxiliary equipment such as water cooling supply, plasma-forming gas supply, LRW and SRW surrogates and batch feeding, off-gas and automated control systems. The scheme of partial water recycling in the cooling system of the most heat-stressed elements providing for decrease of cooling water consumption by 30-50% has been applied. The scheme of compressed air supply for various technological operations such as plasma-forming air feeding, reverse blow-out of filters, pneumatransport, LRW surrogate spray, etc. using a small rotational compressor was developed and applied. LRW surrogate is fed from tanks by pumps. Glass forming additives are fed by either screw feeder or pneumatransport with a compressed air.

The off-gas system serves for trapping of fine and ultra-fine particles, aerosols, toxic gases and vapors. The system consists of a cyclone, a two-step sleeve filter (apparatus for filtration of off-gas from aerosols with reverse blow-out), a scrubber with a line of spray solution recycling through a housing-pipe heat-exchanger, a channel-type off-gas heater, HEPA filters, an exhaust fan and sampling units.

A feature of the filter design is that the apparatus consists of two parts. Major aerosols with drop size of 4-8  $\mu$ m and more are deposited at the first stage of filtration (on stainless steel grid). Aerosol precipitation is completed at the second stage of filtration on glass fiber sleeve or cassette filter. Spent fiber may be added to feed and vitrified. The scrubber is meant for condensation of water vapor and purification of off-gas from toxic gases and aerosols.

The automated control system is a two-level information system operating in a real-time regime. The functions of the system are realized in two hierarchical levels: collection and treatment of the data from sensors monitoring the technological process and informational service of operational personnel at control desk, documenting and achieving of the data. A list of the parameters to be controlled and their variation ranges for melter, power source, water cooling system, and off-gas system have been defined.

Major measuring equipment with automated measuring system and software for process monitoring and analysis (VXI, NI LabVIEW integrated system) have been determined and applied. The first-level equipment includes temperature sensors (thermocouples, infrared thermocouples/pyrometers), flow rate meters, manometers as well as whirl rotameters for measuring of water and plasma-forming gas flow rate.

Computation of necessary parameters by specified dependences and data obtained to make timely decisions on process management in a real-time regime in a sub-system of data representation and registration is also provided.

## CONCLUSION

Integrated facility consisting of PC, PM and CC melters equipped with automated process control, batch feeding, off-gas, power and compressed air supply systems has been designed and constructed. The facility is under cold testing now. It will be used for demonstrations and development of operational conditions of new LRW and SRW treatment technologies with production of glassy, glass ceramic and ceramic waste forms. The given technologies may be also used for non-radioactive applications for production of glasses and ceramics. The PC is considered as an alternative to rotary calciner. The PM technology is perspective for melting of materials with low electric conductivity.

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