Generalization of Experience in Radioactive Waste Melting with Application of Induction Melting Technology, Gained at Moscow Sia "Radon" During The Period from 1992 to 2006

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ABSTRACT.

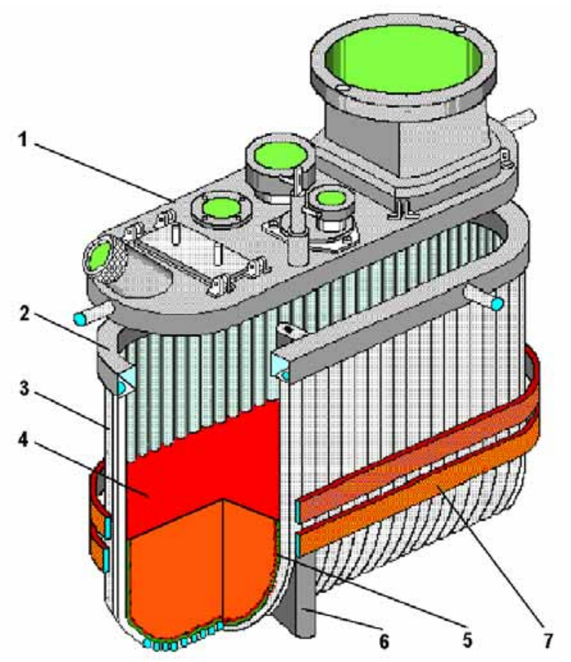
This report provides the results of experimental work in the field of RAW (radioactive waste) thermal treatment with application of the induction melting technology , results of engineering development of induction melting systems at Mos SIA Radon ,as well as aspects of designing and application of new designs of induction melters are considered. Technical problems and ways of their decision when developing and improving of induction melting technology at Mos SIA "Radon" and also experience in thermal treatment of various kinds of waste with application of this technology are reported in short . This technology at the certain completion is quite suitable for treatment of high- level waste (HLW) and intermediate- level waste (ILW). Technical decisions available at present for this technology can be a basis for development of new induction melting systems in which the new materials , new automated control systems, new approaches to designing, application, management should be applied.

INTRODUCTION

The induction melting technology is based on application of standard generators of high-frequency (HF) field and connected with it induction melters such as « Cold crucible » (IMCC). The melter is the original device. It is developed for specific targets of treatment of various kinds of waste. Except for generators and melters the IMCC technology is carried out on the whole complex of technical units - apparatus ,devices, , mechanisms, instruments, etc., - standard and non-standard units. The technology for RAW IMCC treatment, based on initial heating of melt by electromagnetic currents of high frequency, has been applied and improved at Mos SIA "Radon" already more than 15 years. Here liquid and solid waste of low- and intermediate level are treated . At present Mos SIA "Radon" has constructed a number of pilot plants using HF (high-frequency) field generators of different frequency (from 0,44 up to 5,28MHz) and various oscillatory capacity (from 20 up to 250 kw),and also the plant for liquid radioactive waste of low- and intermediate-level activity with generators capacity of 160кw (oscillatory capacity) and with operating frequency of 1,76MHz has been constructed and put into operation. Researches have been carried out and melting technologies for various wastes (LRW (liquid radioactive waste) of a various origin with obtaining of phosphatic and borosilicate glasses, ash residues, synthetic minerals (including Sinrock), waste products with the higher iron contents , ion exchange resins and catalysts, etc. have been developed. Thus products of various structure and properties have been obtained: various waste glasses, glass like materials, glass ceramics, crystal structure glasses. The project of plant where it is supposed to combine pyrolysis of combustible waste with the further melting of the ash residue is under development.

IMCC TECHNOLOGY SUMMARY

The basic technical feature of induction melter is lattice structure of its design. In other words, the melter represents the trellised vessel consisting of rectilinear or U-shaped elements (tubes of stainless steel), or the oval or cylindrical form. The distance between design elements makes 1,5-2,0 mm that provides penetration of an electromagnetic field into the melter. Other important technical feature of induction melter consists in application of water cooling of all elements contacting with melt. The system of water supply and water discharge from the melter was organized. From above the melter is tightly closed by a cover on which means of the control and auxiliary technological devices are placed. The cover is made of stainless steel. Melt is discharged through the special water-cooled discharge device which can be bottom or side. The melter bottom (if it is available) is also made of stainless steel and also cooled by water. Hermetic sealing of melter walls is made by specially developed coatings which in a wet kind are rendered on melter walls, are wound from above by fiber glass fabric and dried up. Thus, assemblage of the melter represents the tight vessel to which various technological communications and means of the control and management are connected. This vessel is located inside of inductor, which represents a ring (or coil of several coils), made of a copper pipe and having water cooling. One variant of induction melters is shown in Figure 1.



1 - cover, 2 -manifold , 3 - water- cooled sections, 4 - glass melt, 5 - scull, 6- discharge unit, 7 - inductor.

Figure 1. IMCC type melter .

PRINCIPLE OF OPERATION OF IMCC TECHNOLOGY

The high-frequency generator generates an alternating high frequency current which passes through inductor, thus the variable electromagnetic field of high frequency (in our case of 1,76 MHz) is created around of the inductor . This field through backlashes between tubes penetrates into working space of the melter where it interacts with glass melt (glass melt in molten state is electroconductive). The interaction promotes vortical currents in bulk of the melt , that results in heating a conductor (melt). As a result the glass melt starts to be warmed up and a surplus of heat acoomulated in it allows charging additional portions of the waste to be treated. Energy emitted at field-melt interaction is sufficiency to melt a new portion of this waste (charge). Thus, gradually there is an accumulation of the melt in working space of the melter. The melt is periodically discharged out of the meter through the discharge unit into the receiving steel container.

At the moment of start for formation of initial melt volume the special starting device is put into the melter and the starting charge structure is applied. Walls of the melter are cooled by circulating water all the time , at that in result of contact of the melt with a cold wall there is a formation of scull (mixture of glass material and unmelted charge) which serves as very good protection for a design material against aggressive melt influence. Gases formed at melting charge are removed out of the melter into gas purification system through a special branch pipe. To melt non-conducting in a solid state substances the frequencies in a range of 10E5-10E7 Hz are applied. According to the state requirements in Russia frequencies of 0,44 MHz, 1,76MHz, 5,28MHz of the given range are allowed.

PROBLEMS SOLVED IN DEVELOPMENT OF IMCC TECHNOLOGY

During work on research, application and development of induction melting technology for RAW treatment (in particular LRW (liquid radioactive waste)) the following problems have been successfully solved.

Problem of development of induction melter design . We should develop the melter suitable for RAW treatment allowing to provide acceptable productivity, safety and a resource of operation. Besides we should provide necessary practically manufacturing and application. Process of creation of new models of melters represents a package of complex calculation, design, and invention works. At a significant variety of the phase being processed in melters, it has been found, that design features of melters for treatment of these phases are practically identical. In all melter designs collectors with inlet and outlet ducts, to which hoses for cooling water supply are jointed (in our circuits we apply flexible joining the melter to water supply system) were stipulated . Since process of melting of any waste is accompanied by evolving of gases which to be cleaned and neutralized, the melter cover has to be equiped by branch pipe to remove off-gases into gas purification system . The melter cover should have a chaging device for initial material charging, a hole for service of internal space of the melter and for control of the starting device. The arrangement of this devices in melter cover is traditional at Mos SIA "Radon". Except for the above-mentioned devices there is a discharge unit lock drive on the melter cover and also a number of the devices required for the monitoring and control of the melter is mounted here. In some cases it is required to supply air under the cover when treating combustible waste. In this case the proper branch pipe is mounted on the cover. The melter cover has to be cooled also and for this purpose supply and remove of cooling water through special branch pipes is organized. Latest melter designs are equipped with a flat bottom where the discharging device is mounted. The melt is discharged through this device by means of drain lock. All these elements are also cooled by water.

Problem of equipment of the melter by monitoring and control devices. The problem of choice and application of automatic, driving and, in part, manual instruments for monitoring and control of thermal process with ensuring a safety of the stuff has been solved. The pair of pyrometers (work in a seen range) is applied for the control of temperature of a melt surface and for definition of the moment of loading a charge into the melter. Loading is conducted automatically under indications of the pyrometers on reaching goal in their indications. Also on some models of melters the digital device thermovision which can give a signal to loading the charge is applied for the control of a surface. Now at Mos SIA "Radon" the software and criteria for the organization of automatic loading the charge after the thermovision signal is developed. It provides new ways of full automation of process. The high-temperature thermocouple which is periodically immersed in the melt is applied to the periodic control of melt temperature in depth. All monitoring and control instruments, and also driving mechanisms (an inductor lift drive , drain lock drive, etc.) are operated with automated control system.

Problem of the coordination of melter geometry with physical properties of glass melt. This problem demanded the decision of a problem of optimization of distribution of capacity in the melter-generator system. Decision of such problems assumes preliminary mathematical modelling of processes of waste thermal treatment in induction melter. At Mos SIA "Radon" attempts to solve a problem of modelling of processes in the melter were undertaken. In particular, the problem of choice of optimum diameter of cylindrical melter for melting a glass material with set electroconductivity was solved by means of the generator with the set target frequency. An important point at the decision of this problem is definition of physical and chemical properties of a working medium (glass melt), and also a material to be treated(charge). Properties of the melt (electroconductivity and viscosity) determinatively influence on melter geometry or on choice of the generating equipment. Also the given properties are necessary for taking into account at development of the additional devices working in structure of the melter (mainly it is drain devices and receiving containers). As the basic parameter of the coordination of melter diameter and physical properties of the melt a capacity factor value Cos φ in the melter-generator system was choosen. Thus, the decision of a problem of optimization of distribution of capacities in the generator - melter system was reduced in definition of melter diameter which will provide at set melt electroconductivity and characteristics of the generator the maximal value Cos φ.

Problem of maintenance of melter tightness . The decision of this problem has crucial importance at RAW treatment. It has been developed a sealing coating for melter walls. The coating combines durability and tightness with plasticity and good adhesion to metal. For choice of components for this coating special researches were carried out . The coating in a wet kind was rendered on melter walls so that it filled backlashes between tubes of melter walls . Then a reinforcing glass fiber fabric layer was lined above the wet coating , whereupon the covering was dried up. Also, hermetic sealing elements for all sockets and connections of the melter as in assembly have been developed and effective germetics have been chosen. These development have allowed to increase tightness of the melter approximately three times in comparison with the first samples.

Problem of preparation of initial RAW for treatment and a problem of their loading into the melter. Physical properties of an initial material influence on choice of the equipment for preparation and transportation of this material to the melter: mixing devices, pumps - batchers, shut-regulating armature. Chemical composition of the initial material defines requirements to a material of the equipment and to its assembling (tightness). At use, for example, a liquid initial material (charge) there is a problem of keeping this material in a homogeneous state for a long time, a problem of loading the material with the set charge or under the set program, a problem of maintenance of smooth and uniform loading of material into the melter. At use of aggressive or chemically dangerous medium there is in addition a problem of a safety the problem of maintenance of rust protection of the equipment is arised. The given problem was solved by choice of units of the standard equipment most suitable to this purpose. At that, choice of the equipment was carried out on the basis of preliminary laboratory studies of properties of the material, choice of glass forming additives, hydraulic and technological calculations and on the basis of tests for pilot plants. For treatment of solid materials except for laboratory studies and calculations it was necessary to solve problems of mixture, crushing, transportation, suppression of dusting, etc. Now at Mos SIA "Radon" a system for initial LRW preparation by evaporation, LRW concentrate based material preparation for vitrification (charge), loading and batchings the charge into the melter has been created and it operates in structure of LRW vitrification plant. Thus it is constantly improved. Other variants of initial LRW preparation for treatment are investigated: radionuclides co-precipitation and treatment of resulting deposit or LRW preliminary calcination with the further calcinate treatment.

Problem of reducing the negative effects from interaction of the liquid phase of initial waste and heated melt. Result of such interaction is pulse increase of a stream of an evaporated moisture and drop carrying out from the melter at batchwise charge loading , that results in increase of radionuclides carrying over from the melter into gas purification system. The decision of this problem appeared to be possible only as a result of application of a complex of measures. So, for reducing pulse increases in the off- gases carrying over at the moment of loading the charge a free space volume above glass melt has been increased more than twice thereto the cover "cap" has been developed. Also, batchers of small productivity for maintenance of constant charge loading into the melter were chosen. Devices (feeders) for loading of paste into the melter were modernized to provide more uniform charge distribution on a melt surface. These measures have allowed to reduce essentially pulses of a stream of off-gases and stabilized operation of the melter and off-gases purification system.

Problem of recycling off- gases of a complex chemical composition. For neutralization of the gases formed at waste treatment , the step off-gas purification system was applied. At a stage of mechanical cleaning aerosols were separated by a method of filtering in special filters. Further an absorption of soluble gases with their transformation into salts in scrubbers irrigated by a special sorbent was carried out. At a final stage neutralization of nitrogen oxide by a method of ammonia catalytic reduction was carried out . Now work on increase of efficiency and reliability of the off-gas purification system is conducted. Researches on opportunity of application of wet methods of removing aerosols are carried out. For this purpose the unit of wet cleaning of off-gases from aerosols is developed. The experimental works is conducted to improve sorption and catalytic cleaning . The automation of off-gas purification system is of great importance , since it can raise accuracy of management, improve gathering and processing of the information and release the personnel from many routine operations.

The problem of obtaining of the qualitative product providing the greatest possible retention of radionuclides in glass matrix - choice of optimum structure of glass forming additives and the decision of a problem of annealing of glass blocks to prevent their cracking. One of the main differences of the technology accepted at Mos SIA "Radon" is application of borosilicate matrixes developed here for LRW vitrification. For obtaining of borosilicate glasses no special reagents are required owing to all components of charge are of a natural origin (sand, bentonite clay) or industrially made mixes (datolite concentrate). It essentially reduces operation cost. Also, process of charge preparation for such glass matrix is not connected to use of aggressive medium, as, for example, in a case of phosphatic glass. Besides ,it is necessary to note, that application of borosilicate glass matrix appeared to be more preferable because of high chemical stability under influence of water that has crucial importance at RAW treatment. Thus Mos SIA "Radon" has experience in obtaining of phosphatic glass matrix too. The borosilicate matrix has radionuclides leaching rates about 10-6-10-7g / (cм2*day). For optimization of glass blocks annealing (anniling is process of smooth away internal stress in glass matrix) researches on application so-called receiving containers in a shirt were carried out. Rather encouraging results have been already received. Application of such containers promotes economy of the electric power comsumption (for maintenance of a mode of glass blocks cooling the electric annealing furnace is used) up to 30 % , and, the main thing, does not demand the large material inputs.

Problem of the melt discharge into the receiving container with maintenance of comprehensible tightness, reliability and a resource of work of devices for a glass melt discharge. For the decision of this problem discharge devices with the allocated cooling line have been designed. The problem of a supply and removal of cooling water has been solved for this devices. It has allowed to arrange the discharge device in bottom of the melter, and it became possible to displace the device closer to the melter center. As a result the formation of melt overheating zones about the discharge device have been terminated. At the earlier melter designs the discharge device was mounted at a melter wall and it resulted in formation of such zones due to increase of concentration of internal currents in the melt near prominent elements on walls . In this discharge device a two-chamber cooling scheme was applied. The external chamber, contacting with melt is cooled by a turbulent flow of water. Then by tangential transition water comes from the external chamber into internal chamber from which it is poured outside. Also, in this device a polishing of the cooling channel is applied for decrease in hydraulic resistance and reduction of cavitation risk . The discharge device is opened by means of drain lock - the water-cooled device with a drive. Such design of the drain device provides convenient melt discharge, increase of a resource of work of the device, and also an opportunity of arrangement of this device in any point radially from the center up to a melter wall.

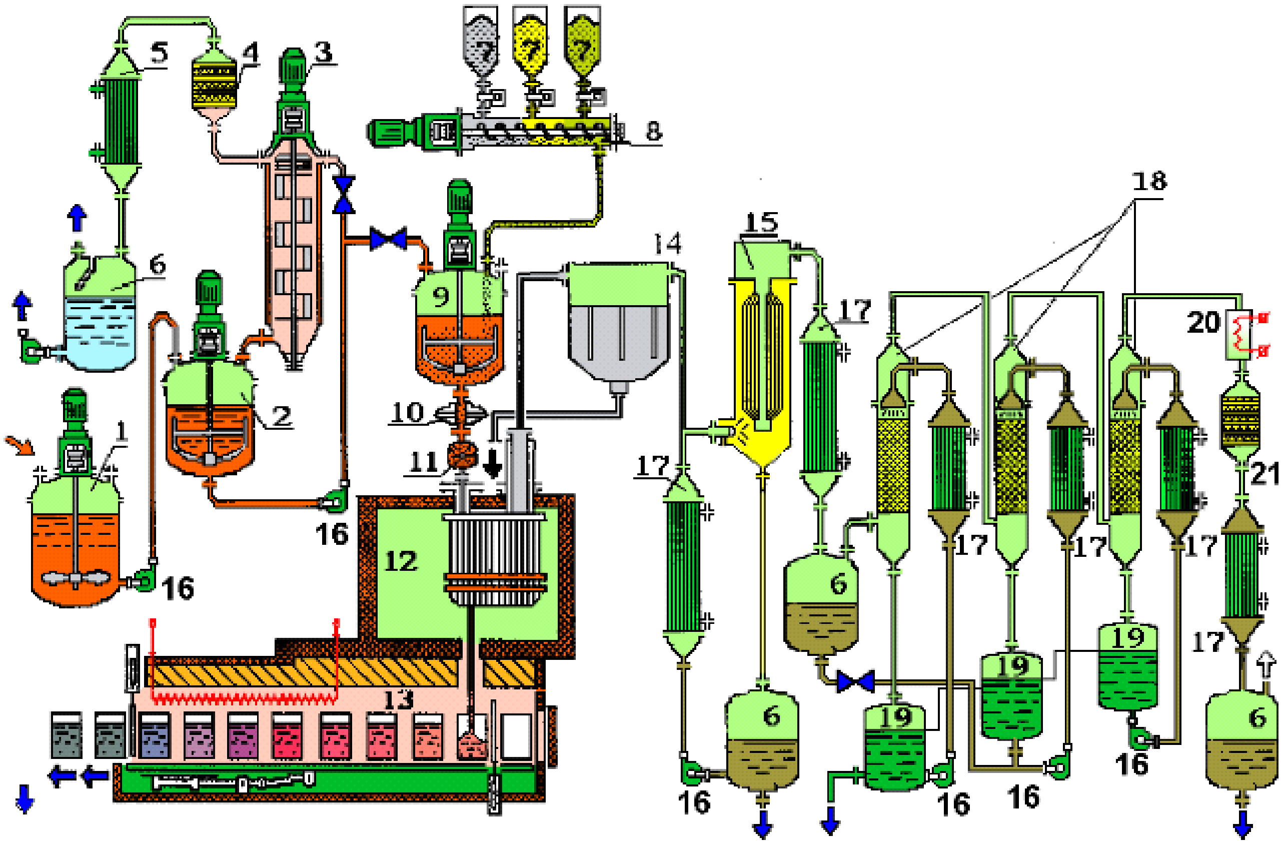
ADVANTAGES OF THE IMCC TECHNOLOGY

The melter design developed at Mos SIA "Radon" is applied now on the LRW vitrification plant. The melter has the following characteristics: diameter-418мм (internal), height 700мм; working temperature +1250˚C; oscillatory capacity of the generator 160kw (frequency nominal); working frequency 1,76MHz; cooling water consumption up to 12м3/hr.; productivity on glass about 18кg/hr. Application of the IMCC technology for LRW treatment gives a number of technical advantages. Such melter can be operated at melt temperatures up to +3000˚C because of a protective layer of scull and water cooling perfectly protect a melter design material. The melter does not require application of a bulky heat-shielding because of water cooling - temperature of an external melter surface is about +20˚C. Small dimensions and weight of the melter allow to dismantle and replace it quickly in case of fault; For the same reason the melter cools down completely after shut down approximately for 4 hours. Input of energy by contactless way and small dimensions of the melter provide its quick-response operation (the system quickly response to managing influences), so this advantage anables automation of LRW treatment process to be improved to high level subsequently . IMCC melter does not apply immersible electrodes which should be changed periodically. Simplicity of the melter design enables its easy repair - to have the welding equipment is enough.

The minimal interaction of melt and melter material with an environment allows to obtain glass product of high purity.

EXPERIENCE IN APPLICATION OF IMCC TECHNOLOGY

As a result of the decision of the above-mentioned problems the completed concept of RAW treatment with application of induction melting technology has been developed as a whole. This concept has been realized in 1999. During application of this technology at Mos SIA "Radon" about 3000м3 of LRW of total activity about 1011-1012 Bq has been treated on LRW vitrification plant (Fig. 2).



1 - LRW storage; 2 - concentrator vessel; 3 - rotory film evaporator; 4 - filter; 5,17 - heat exchanger; 6 - vessel; 7 - glass-forming additives; 8 - screw; 9 - mixer; 10 - mechanoactivation device; 11 - feeder; 12- melter; 13 - annealing furnace; 14- coarse filter; 15- fine filter; 16- pump; 18- absorption column; 19 - acid collector; 20- preheater; 21 - catalytic reactor.

Fig. 2. LRW vitrification plant

This plant has been created for LRW industrial treatment. The average composition of the glass waste product is given in Table I.

Table I. The average composition of the glass waste product obtained from of LRW treatment. (The concentration of components in glass waste product is given in weight percent - %wt.)

Na_2O	K_2O	CaO	Fe_2O_3	Al_2O_3	SiO_2	B_2O_3	Cl^-	SO_3	P_2O_5
18-24	0,3-1,9	6,2-12,1	1,8-2,7	2,2-4,3	46,8-56,9	6,9-9,0	0,1-0,5	0,5-0,7	0,1

The scheme of plant allows to carry out complete automation of technological process, that considerably reduces probability of an irradiation of the staff, creates an opportunity of operation of plant, both in continuous, and in a continuous - periodic (week) mode; anables high reliability of plant operation due to exception of synchronization of flows of material mediums between main units of plant which can work independently , that finally results in some cost reduction in waste treatment. Except for at Mos SIA "Radon" plants for induction melting of solid waste were developed : plant for ash residue vitrification , and also a number of pilot plants for trial treatment of other materials were created. On these plants experiences were carried out on application of induction melting process for treatment of such wastes as ash residue from SRW (solid radioactive waste) incineration , spent catalysts, fibrous materials, natural and artificial minerals, and in each case work has been aimed at obtaining a chemically steady material retaining radionuclides - glasses, ceramics, crystal structure . Works were carried out within the framework of the international cooperation. The induction melting technology was adapted for treatment of modelling wastes from firms of France, Germany, Czechia, the USA, etc. (Foster Wheeler, Nukem, BNFL, Battelle, Nuclear Research Institute). In cooperation with French firm SGN researches were carryied out whenever possible application of technology « cold crucible » for treatment of radioactive waste from this firm. Positive results were received. In cooperation with this firm researches on ion exchange resins recycling in « cold crucible » were carried out. Products of decomposition of resin were included in structure of glass waste product (borosilicate basis). Some researches on inclusion of simulators of radioactive waste from the Novovoronezsh NPP, the Kalinin NPP into glass structure were carried out In cooperation with firm Nukem process was developed for treatment of the ash residue from SRW insineration. On the base of positive results obtained a plant for ash treatment has been designed . Glass like materials of high chemical stability were obtained. Researches on RAW treatment and their inclusion in structure of various glasses on the basis of phosphatic compositions, including glasses with the high contents of lead, iron were carryied out . Except for ion exchange resins from firm SGN the sorbents (NZHA, NZHS), fibrous heat-insulation materials (glass wool), catalysts (AVK 10M), ion exchange resins of a domestic production (KU, AV, and also Dowex) were processed in «cold crucible » and included in structure of various glasses. Now researches on treatment of high iron content LRW under the contract with the U.S. Department of Energy are carried out. The results satisfied the American partners have been already obtained. The main conclusion from the above-stated report is the following: Mos SIA "Radon" has wide experience in the complex decision of a problem of RAW induction melting as against many other organizations. Using the technologies developed or being developed at Mos SIA "Radon", it is possible not only to treat any type of radioactive waste, but thus also to solve a problem of recycling of all secondary waste products formation of which is inevitable, and also to obtain final waste product allowing reliably to immobilize radionuclides and to provide safe and reliable long-term storage of this product.

FURTHER DEVELOPMENT OF IMCC TECHNOLOGY AT MOS SIA "RADON"

The main directions of the further development of IMCC technology at Mos SIA "Radon" are improvement of its technological level and development of a process control system with use of modern means and ways of management.

In the field of development of a technological level of IMCC technology we intend the further perfection of melter designs, development of melters for processing new materials, and also application of modern devices and means in auxiliary systems : gas purification, LRW preparation, engineering maintenance, etc. At absence of standard units of the equipment with the required characteristics a specific equipment will be developed . Also, application of new materials, application of new glass forming additives, etc. is perspective. The special seaches are focused on ways of LRW preliminary preparation before treatment. A key condition of safety of IMCC technology as well as other technologies dealing with LRW treatment (in particular high - and intermediate level) is automation of process. And the level of automation thus should aspire to full automation of 100%. At Mos SIA "Radon" melter control algorithms which then have been realized as the software for an automated control system (ACS) of LRW vitrification plant have already been developed. Besides under this software the complex of the measuring equipment and control devices was installed. The melter automated control system is capable to gather the information on electric parameters of generators, on a condition of flows of cooling water and off gases. The primary goal of development in the field of control of technology today is full equipment of technological plants by the modern unified means of measurement, control and management, development of algorithms of automated control by indidual devices, units and systems. Further these algorithms should become a basis for creation of local contours of management and then full automated control system for IMCC technology. Creation of full automated control system will allow to raise quality of operation of plantss and finally quality of a glass waste product that will provide increase of safety of RAW treatment and storage of waste products from this treatment . Thus it is necessary to provide an opportunity of the further development and improvement of IMCC technology in a direction of a technique as well as in a direction of development of control systems.