Cold Crucible Induction Melting Technology for Vitrification of High Level Waste: Development and Status in India

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ABSTRACT

Cold crucible induction melting is globally emerging as an alternative technology for the vitrification of high level radioactive waste. The new technology offers several advantages such as high temperature availability with long melter life, high waste loading, high specific capacity etc. Based on the laboratory and bench scale studies, an engineering scale cold crucible induction melter was locally developed in India. The melter was operated continuously to assess its performance. The electrical and thermal efficiencies were found to be in the range of 70-80 % and 10-20 % respectively. Glass melting capacities up to 200 kg m^{-2} hr^{-1} were accomplished using the ESCCIM. Industrially adaptable melter operating procedures for start-up, melting and pouring operations were established

INTRODUCTION

Immobilization in suitable glass matrix is presently being practised in many countries for the conditioning of high level radioactive liquid waste (HLW) originating from fuel reprocessing plants. In India, induction-heated metallic melter was developed initially to carry out vitrification process. Waste immobilization plants at Tarapur and Trombay employ metallic melters for the vitrification of HLW [1]. In the vitrification process employing the metallic melter, the progressive steps of evaporation, drying, calcination and melting of waste and glass additive slurry are achieved in a metallic (inconel – 690) process pot heated by a multi-zone induction furnace. The molten product after homogenization is drained out from the process pot through a bottom freeze valve to a stainless steel canister which is seal-welded and over-packed subsequently, allowing reuse of process pot. Though induction-heated metallic melter is a compact and simple system, it has low throughput on account of limited diameter and short melter life due to high temperature glass corrosion.

In order to circumvent these limitations Joule-heated ceramic melter was developed locally. An advanced vitrification system based on the ceramic melter is under operation at Tarapur. In the Joule-heated ceramic melter, thermal energy required for the vitrification is generated using multiple pairs of metallic electrodes immersed in a pool of electrically conducting glass. Availability of unrestrained heat transfer area and amenability to continuous mode of operation facilitate larger processing capacity and presence of glass corrosion resistant refractory wall enhances the life of the ceramic melter. Natural convection currents prevailing in the electrically conducting molten glass pool improves the product quality. By virtue of the large thermal inertia of the glass pool, the Joule melter can accommodate variations in the feed streams to a greater extent. However, the major operating constraint for the ceramic melter is that the metallic (inconel 690) electrodes are not to be exposed to temperatures higher than 1150^{o}C in order to ensure melter life. In addition, decommissioning of the ceramic melter at the end of its service is quite involved. Globally emerging vitrification technology based on the cold crucible induction melter (CCIM) offers several advantages such as long melter life, high temperature availability, high waste loading, high specific capacity, etc. The CCIM is also more tolerant to presence of noble metals than the traditional Joule heated ceramic melters because of the heat release in the melt by direct induction [2]. The CCIM technology can effectively be used to condition hard-to-process waste streams [3]. It is also a promising technology for adopting glass-ceramic and synroc-based matrices for high level waste immobilization [4]. In view of the management of high level liquid waste that will arise from the reprocessing of Fast Breeder Reactors and Advanced Heavy Water Reactors, cold crucible induction melting technology is being developed in India. Developmental works ranging from laboratory scale experiments to engineering scale demonstration have been carried out to establish the CCIM technology. Design and development aspects of the engineering scale cold crucible induction melter are presented in this paper.

COLD CRUCIBLE INDUCTION MELTING TECHNOLOGY

In cold crucible induction glass melting, molten glass is directly heated by electromagnetic induction. The direct heating is accomplished in a segmented crucible which is manufactured from contiguous segments forming a cylindrical volume, but separated by a thin layer of electrically insulating material [5]. The segmented crucible facilitates penetration of electromagnetic field into interior of the crucible resulting in eddy currents and associated Joule heat in the electrically conducting glass.

Direct heating of glass facilitates heating the process material to high temperatures. In order to avoid high temperature glass corrosion of the metallic crucible, internal cooling of the segments is provided. This cooling produces a solidified glass layer, which acts as a protection against glass corrosion along the melter inner wall. High temperature availability without substantial corrosion makes the CCIM a promising technology for immobilization of high level nuclear waste.

In 1980, a direct induction process using a water-cooled metal crucible was developed in France to melt metal waste from nuclear fuel reprocessing. This experience led to the development of cold crucible melter for inductively heating glass in a water-cooled crucible [6]. France have more than 20 years of research and development experience in the field of cold crucible induction melting related to immobilization of radioactive wastes [7]. A full scale plant using cold crucible induction melter is operational in Russia since 1999 for vitrification of low and intermediate level liquid waste [8]. In USA, Idaho National Engineering and Environmental Laboratory, in collaboration with Russian scientists, has developed CCIM prototype for waste vitrification [9]. Present paper describes certain design and developmental aspects of CCIM in detail based on the work carried out in India.

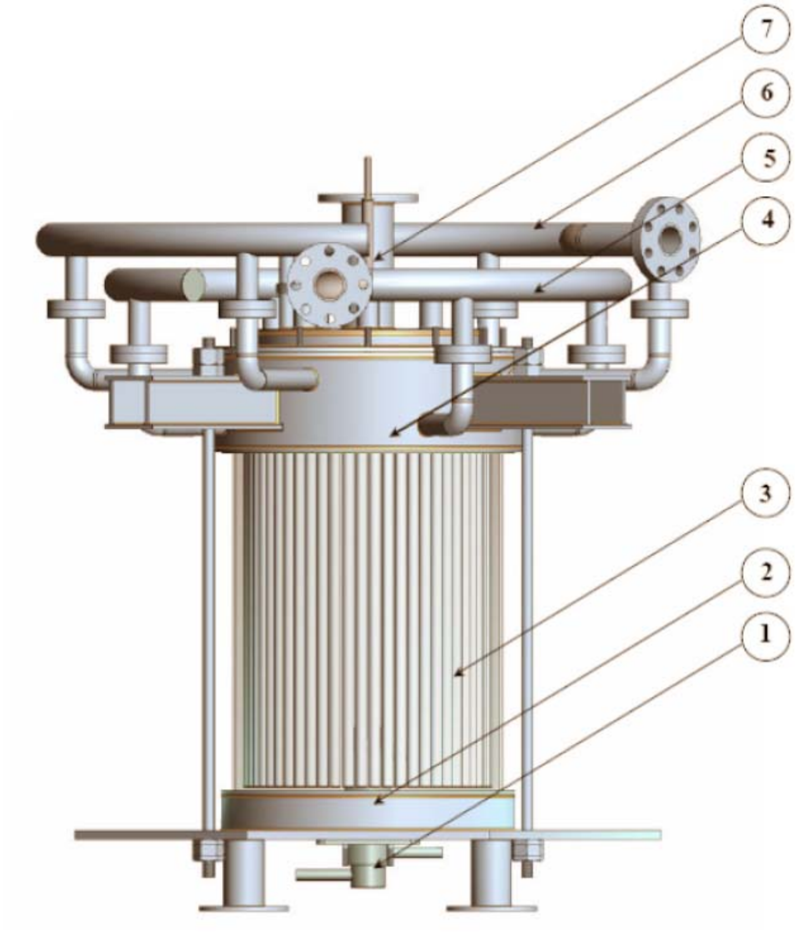
ENGINEERING SCALE COLD CRUCIBLE INDUCTION MELTER

In a continuous metallic crucible, the magnetic field generated by the coil current is opposed by the magnetic field caused by the induced current in the crucible. In such a scenario, the required magnetic flux to heat and melt an electrically conductive charge inside the crucible does not exist. In a segmented crucible, induced currents flowing in each segment create an electromagnetic field inside the crucible. Such an arrangement is equivalent to two air-cored transformers - one formed by the induction coil and the crucible segments and the second formed between the segments and the charge. Thus, the segmented crucible acts as a field concentrator, indirectly reducing the gap between the induction coil and the charge.

The number of segments, shape of the segment and spacing between the segments strongly influence the electromagnetic behaviour of the system [10]. The Joule heat generation due to eddy currents in the segmented crucible is governed by the above factors [11]. The arcing risk between the segments reduces as the number of segments increases. Another important requirement of segmentation is to establish an efficient cooling to generate a uniform protective layer between the molten glass and the metal parts.

Based on the laboratory and bench scale experimental studies carried out at Bhabha Atomic Research Centre, an engineering scale cold crucible induction melter (ESCCIM) was developed for technology demonstration [12]. Fig. 1 shows major components of the ESCCIM. The melter essentially has a water-cooled base to support the molten mass, a circular array of electrically isolated segments to facilitate electromagnetic field penetration into the melter and an inlet-outlet chamber to provide cooling water to the vertical segments, which constitute the melter wall. The ESCCIM comprises of 56 segments with a tube-in-tube configuration. Each of these segments consists of a one inch outer tube (BWG 12) connected to the cooling water inlet chamber and a half inch tube (BWG 12) connected to the cooling water outlet chamber. These stainless steel cooling tubes are arranged in a circular array to hold a molten glass pool of 500 mm diameter. The base and the tubular segments are electrically isolated with a 3 mm thick Teflon gasket in order to avoid electrical short-circuiting of the segments at the bottom. An inlet ring header is employed to distribute cooling water to the lower inlet chamber and an outlet ring header connected to upper outlet chamber is used to discharge hot water from the melter. The base is provided with an independent cooling water circuit.

A water-cooled mechanical plug assembly is used to drain molten glass from the melter. The mechanical plug assembly consists of a water-cooled plug which can be pneumatically actuated to open or close a 15 mm opening of water-cooled plug seat fitted to the melter base. The plug seat protrudes 200 mm into the melter in order to maintain a minimum amount of molten glass always inside the melter. During the melter operation, glass level is allowed to reach a maximum of 400 mm. The exterior of the vertical segments is provided with a 25 mm thick casting of high temperature refractory cement. All metallic parts of the crucible were fabricated out of nitric acid grade stainless steel (SS 304 L) considering the acidic nature of the high level liquid waste.



1 water-cooled plug seat
2 water-cooled base
3 water-cooled segments
4 cooling water inlet-outlet chamber
5 cooling water inlet header
6 cooling water outlet header
7 water-cooled plug

Fig. 1 Major components of the Engineering Scale Cold Crucible Induction Melter (ESCCIM)

INDUCTION HEATING POWER SUPPLY SYSTEM

On account of relatively poor electrical conductivity, induction melting of glass requires high frequency for efficient heating. The heating efficiency is governed by the diameter to skin-depth ratio of the load to be heated. Ratios ranging from 2 [5] to 7 [13] have been reported in the literature for cold crucible induction melting of glass.

In Russia and USA, valve based generators are being used for induction heating power supply for the CCIM [13] while in France, semiconductor based induction heating power supply is being used [5]. Valve type generators are less efficient compared to transistorised units. In addition, valve based generators are prone to frequent breakdowns because of high voltage related problems. Globally valve type RF (radio frequencies – above 250 kHz) generators are now being replaced by transistorized generators. Therefore, transistorized generator was selected as a better choice for cold crucible induction melting of glass.

On the basis of locally available, transistor based induction heating power supply systems, a frequency of 200 kHz was chosen for the present ESCCIM application. This power supply, procured from M/s. G. H. Induction India Pvt. Ltd., provides diameter to skin-depth ratios ranging from 2 to 4.5 for different glasses with electrical conductivities ranging from 5 Ohm cm to 1 Ohm cm. A current-fed, parallel resonant load circuit was selected for the inverter circuit topology. The inverter was rated for 350 kW with a maximum power delivery of 200 kW in the load.

The basic elements of the induction heating power supply are:

a) an air-cooled, three-phase, double wound step down transformer (415 V to 320 V) with two secondary windings, one in star and the other in delta formation to reduce harmonics and ripples,
b) two controlled rectifiers with a common dc bus through two separate chokes to feed constant current,
c) an inverter, with MOSFETs as switching device, configured as a bridge with four arms arranged in the conventional H-format to generate an operating frequency of 200 kHz and
d) a capacitive voltage multiplier to provide a maximum coil voltage of 1600 V from a nominal inverter output of 375 V, 200 kHz.

The power transistors of the current-fed inverter are switched always in resonant mode, when the zero-cross-over of the voltage takes place. Thus, switching losses as well as the voltage spikes are kept low. As per the frequency control scheme, the frequency is tracked automatically on a cycle-to-cycle basis and constant correction is applied to keep the system latched on resonance. The power is controlled by varying the output dc voltage of the controlled rectifier.

INDUCTION COIL

The induction coil was designed to maximise the electromagnetic coupling with the ESCCIM for an efficient heating and to minimise the coil impedance for an efficient current pumping. A one-turn, 3 mm thick, 220 mm high, water-cooled copper inductor was used to limit the maximum coil voltage requirement to 1600 V. A coil inside diameter of 600 mm was chosen to provide a 25 mm thick casting of high temperature refractory cement at the exterior of the crucible segments. The coil was located 150 mm above from the water cooled base of the ESCCIM to reduce the Joule heat generation in the top metallic plate of the base. A 5 m long water-cooled, copper busbar was used to connect the inductor and the capacitor bank. The impedance of the coil is a function of the level of the glass in the crucible, resistivity of the charge and physical coupling available. The glass level varies from a minimum level of 200 mm to a maximum level of 400 mm during the melter operation. The 220 mm high inductor was chosen to ensure proper coupling even under the minimum glass level condition.

HEAT REMOVAL SYSTEM

The heat removal system is comprised of primary cooling water (PCW) circuit which cools various components of the segmented ESCCIM and secondary cooling water (SCW) circuit which removes the heat gained by the PCW from the melter. The PCW loop consists of a plate type heat exchanger (heat duty: 200 kW) and a stainless steel centrifugal pump (capacity: 750 LPM, head: 35 m) and a DM water storage tank. The SCW circuit consists of a cooling tower (heat duty: 250 kW) and a circulation pump (capacity: 1000 LPM, head: 35 m) in addition to the heat exchanger connected to the PCW loop. Both PCW and SCW circulation pumps were provided with 100 % standby to ensure cooling water supply which is essential for the uninterrupted operation of the ESCCIM. A once-through emergency cooling water line from an overhead tank was connected to the PCW circuit through an air-to-close isolation valve to ensure sufficient cooling in the eventuality of a power blackout. Appropriate pressure relief valves were provided in the cooling circuit to vent out steam generated during loss-of-cooling simulation studies as a part of the safety related analysis. In addition, various safety-related alarms, trips and interlocks were incorporated to ensure the safe operation of the melter. Adequate instrumentations such as flow rate and temperature measurements were provided to generate sufficient engineering data for design verification.

OPERATION OF ENGINEERING SCALE COLD CRUCIBLE INDUCTION MELTER

The ESCCIM along with associated auxiliary systems are shown in Fig. 2. A number of experimental runs were carried out using different sodium borosilicate glass compositions to establish melter start-up, glass melting and glass pouring operations. The power control mode was selected for all these operations. Based on the experimental results, power control strategies for various stages of melter operation were arrived at. Fig. 3 shows various stages of a typical operation comprising of start-up heating, melting, soaking and pouring using sodium borosilicate glass with chemical composition given in Table I and electrical resistivity given in Table II.

Table I Composition of sodium borosilicate glass

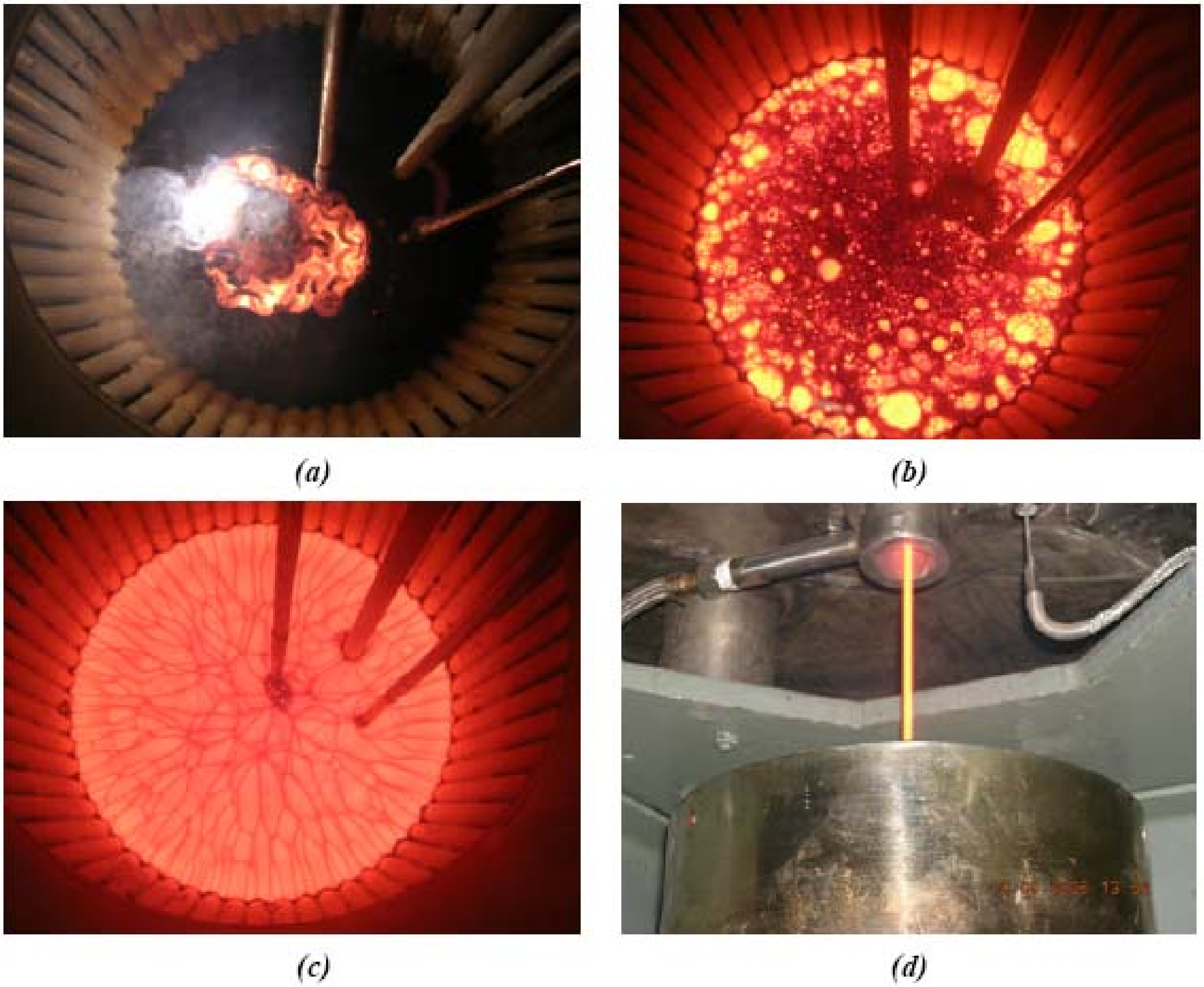
Chemical	Weight %
SiO_2	32
B_2O_3	25
Na_2O	28
Fe_2O_3	13
TiO_2	2

Table II Electrical resistivity of sodium borosilicate glass

Temperature (^{o}C)	Resistivity (Ohm cm)
900	3.3
1000	1.9
1100	1.2

Fig. 2 Engineering scale facility for cold crucible induction melting

Glass being an electrical insulator at room temperature, it must be preheated enough for substantial electromagnetic coupling and sustained induction heating. Since most of the operations including start-up heating need to be carried out remotely in a high level waste immobilization plant, a safe and robust start-up procedure is essential for adapting the CCIM technology for waste vitrification. Among the various possible options, use of a sacrificial conducting material appeared to be promising. Major considerations while evolving the melter start-up procedure were remote feeding of the sacrificial conductor into the melter, coil voltage and power requirements for start-up heating, sustainability and consistency of induction heating, time required for establishing the minimum glass level (200 mm), impact of residual material on product quality and possibility or extent of damage during any arcing during the start-up. Based on several experiments carried out, graphite rings (of 50 mm outside diameter, 30 mm inside diameter and 10 mm thickness) were found to meet all the requirements of a robust and safe start-up. A chain formed from 30 graphite rings can be fed remotely to the centre of the crucible filled with initial glass charge, as seen in Fig. 3(a). The chain form enhances glass melting by means of heating and arcing. The rectifier output voltage was set to 350 V (dc) to produce 80 kW in the load. The graphite rings were continuously covered with thin layer of glass frit to reduce radiation losses. As seen in Fig. 4, the rectifier output voltage decreases and the feeder line current increases as the molten glass pool builds up substantially indicating direct coupling with the glass melt. As per this procedure, the glass melt grows gradually from the centre to the periphery. Thus, the present procedure eliminates arcing between the crucible segments and the graphite rings as the arcing potential reduces significantly when the molten glass starts electrically conducting substantially. The arcing between the crucible and graphite rings is highly undesirable as severe arcing can puncture the water-cooled segments.



(a) start-up heating
(b) glass melting
(c) glass soaking
(d) glass pouring

Fig. 3 Different stages of ESCCIM operation

In order to establish the glass melting kinetics, prefabricated glass frits of 5-10 mm size was fed onto the surface of the molten glass pool, as shown in Fig. 3(b). The melting rate was observed to vary strongly with the melter operating power as expected. The melting rates varied from 20 kg/hr at an input power level of 120 kW to 40 kg/hr at 150 kW. The melting rate was enhanced by better mixing on account of the stronger natural convection currents in the molten glass pool. The surface conditions and the convective flow patterns during glass melting are shown in Fig. 3(c). Skin effect caused by the high frequency magnetic field results in a relatively hotter glass at the periphery of the molten glass pool. As a result, lighter glass rises at the periphery and flows towards the centre of pool surface. As it flows along the pool surface, it looses heat to the surrounding leaving the glass cooler near the centre where the denser glass sinks. Enhanced operating power improves net gravitational body forces; weakens the resistive viscous forces; and thereby results in better mixing. The electromagnetic coupling improves as the glass level builds up resulting in an improved heat generation in the glass melt for a given coil voltage. However, this is offset by the additional heat loss through the added solidified protective layer.

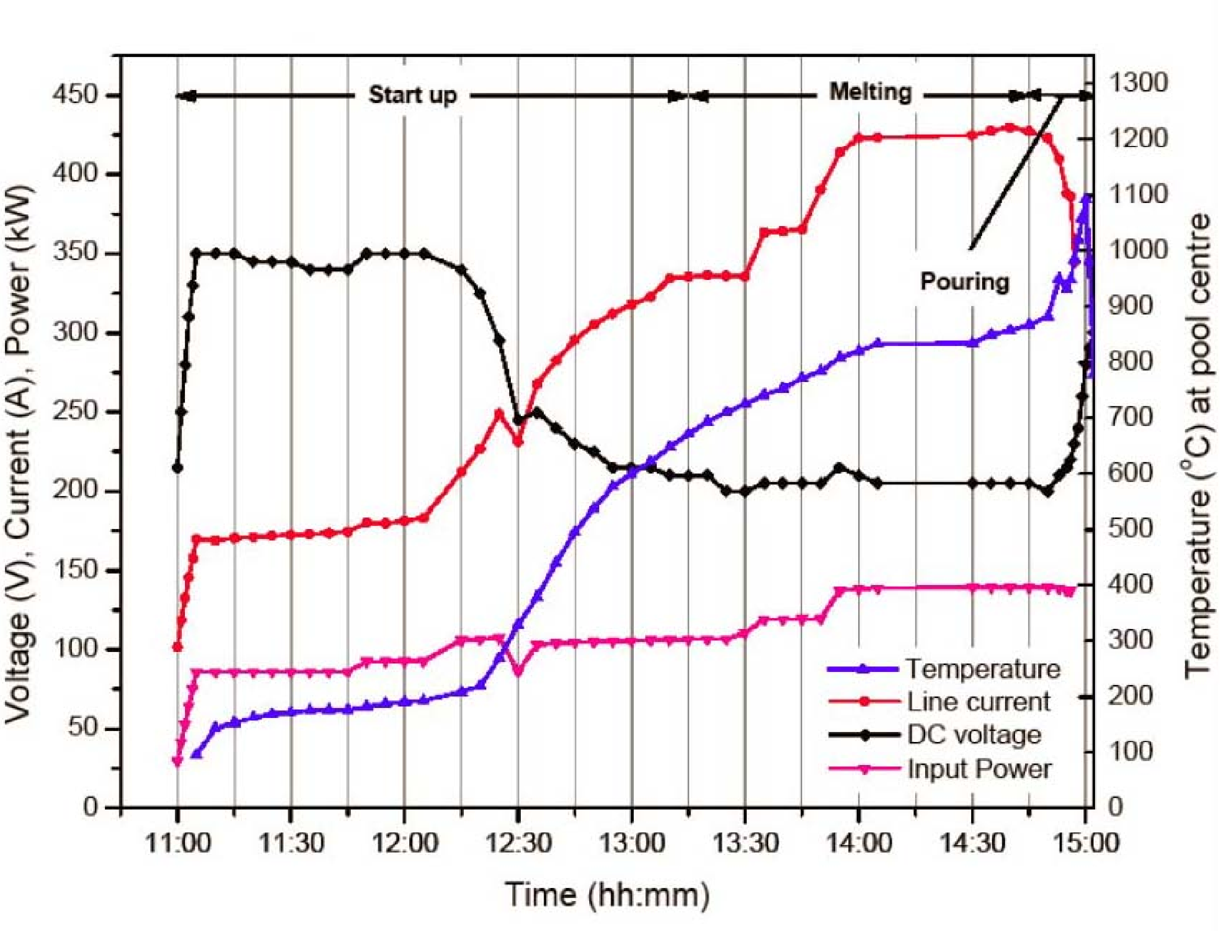


Fig. 4 Melter parameters during a typical operation of the ESCCIM

Once the glass level reaches the maximum level (400 mm), as indicated by a thermocouple located at the maximum level, feeding can be discontinued. A batch of 100 kg glass can then be drained after a soaking period to ensure the product homogeneity. This is accomplished by pneumatically lifting the water-cooled mechanical plug to a height of 100 mm above the plug seat. Glass pouring rate of 10 kg/min was obtained with a 15 mm opening in the plug seat. Fig.3(d) shows the glass pouring in progress. During the pouring, glass level decreases resulting in poor electromagnetic coupling as indicated by an increase in rectifier output voltage in Fig. 4. The temperature at the pool centre, which is in the plane of the plug seat (at 200 mm level), increases as the high temperature glass above this plane approaches it. The plug is lowered back to the plug seat at the end of pouring to keep the melter ready for next cycle of operation.

Based on the experimental results obtained during several operations of the ESCCIM, energy balance calculations were carried out to estimate the electrical and thermal efficiencies of the system. These results are tabulated in Table III in which energy losses are expressed as percentage of input energy to the respective components. Analysis of the experimental data confirm that the overall efficiency of the ECCIM for a chosen glass composition is influenced by the glass level, operating power and associated thermal field in the glass melt.

Table III Efficiencies for the ESCCIM

Component	Energy losses (%)	Efficiency (%)
Power supply system	14-21	79-86
Coil and busbar	6-12	88-94
Melter - thermal	73-86	14-27
Electrical	20-30	70-80
Thermal	80-90	10-20

The purpose of the present experiments was to locally demonstrate the cold crucible induction melting technology, establish industrially adaptable operating procedures and validate the melter design for vitrification of high level liquid waste. At present, experiments are in progress to establish optimum melter throughput with liquid feeding. Further, experiments will be carried out to study the characteristics of the product glass such as leach resistance, crystal content, homogeneity etc. in order to arrive at an acceptable product.

CONCLUSIONS

In India, an engineering scale cold crucible induction melter was locally developed for the inactive demonstration of the technology. The melter was operated continuously to assess its performance. Industrially adaptable melter operating procedures for start-up, melting and pouring operations were established. Glass melting capacities up to 200 kg m^{-2} hr^{-1} were accomplished using the ESCCIM. The electrical and thermal efficiencies were found to be in the range of 70-80 % and 10-20 % respectively.

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