

ANALYSIS OF ELEMENTARY PROCESS STEPS IN INDUSTRIAL GLASS MELTING TANKS - SOME IDEAS ON INNOVATIONS IN INDUSTRIAL GLASS MELTING

RUUD BEERKENS

TNO Science & Industry, Glass Group, De Rondom 1, P.O. Box 6235, 5600 HE Eindhoven, The Netherlands

E-mail: ruud.beerkens@tno.nl

Submitted May 6, accepted October 9, 2008

Keywords: Fast fining, Glass furnace design, Compact melter, Controlled melting

Conventional industrial glass furnaces show broad glass melt residence time distributions in the melting tanks and average residence times may be up to more than two days for high quality glass products, such as float glass or TV glass, despite the minimum residence times of 8-10 hours (or even less than 4 hours for container glass furnaces). Long residence times are associated with large melt tank volume/pull rate ratios and high structural heat losses. The recirculation flows, necessary to supply the batch blanket with sufficient energy for the fusion processes of the batch materials, are often poorly controlled and cause these shown residence time differences. For each elementary process step the most relevant process parameters in terms of desired flow patterns, mixing behavior, temperature, chemistry and required time for completion of the process step are analyzed. From this analysis, it is concluded that the different steps need very different conditions comparing the processes with each other. Therefore glass furnace design modifications should be focused on the development of furnaces with segments, each of them dedicated and optimized for the a certain stage of the melting process: melting-in of batch, sand grain dissolution, removal of gases (bubbles and dissolved gases), re-absorption of residual bubbles and glass melt (chemical and thermal) homogenization. Some developments in fast fining and rapid melting-in of batch will be shown and shortly discussed.

INTRODUCTION

Residence time distributions, determined for batch and the glass, after batch fusion, in glass melting furnaces show that the flow of material in the tank partly follows plug flow regime, but that also sections with strong mixing and re-circulation flows exist. In the glass melting tank, different zones should preferably correspond to different elementary process steps. But, in most industrial glass melting tank furnaces, these zones are not physically separated from each other, and the different processes in the tank may interfere. These basic processes are: a). heating of batch up to "melting or fusion" temperatures, b). fusion reactions to form a silicate melt and c). dissolution of sand grains in this primary silicate melt and final molten glass composition, d). removal of gas bubbles and dissolved gases and e). chemical homogenization plus thermal homogenization of the viscous material.

Re-circulation of the melt in these tanks is relatively intense: in most cases the glass melt passes a zone 5 up to 8 times before the melt leaves the tank. Most of the well-molten en completely degassed melt from the hot spot area, returns to the batch area and is mixed with freshly molten non-homogeneous glass, still containing

very large numbers of bubbles and seeds. The high quality hot glass melt from the hottest spot of the tank just brings part of the required melting energy to the batch tip. The mixing with the freshly molten glass, that contains many un-molten grains and seeds, spoils this high quality melt.

Glass with a melting history representing 4 hours of melting and with 48 hours of melting can be in the same glass container or glass sheet product. This means that if the glass quality is still acceptable, the parts with a residence time of 48 hours has been in the tank up to 12 times longer than essentially necessary. Němec and Jebavá [1], showed that dead water zones and the so called space utilization factor (depends on the ratios between minimum and average residence times) have a large influence on the energy consumption of a glass furnace for given pull or the maximum pull obtainable from a melting tank. They also showed, that very high temperatures may not be the optimum solution for high pull and low energy consumption and that certain optimum conditions in terms of optimum temperature, stirring, enhanced bubble growth rate can be derived for a certain glass melting tank to get high pull and relatively low specific energy consumption and high flexibility. They propose developments in the direction of

Paper presented at the seminar "Advanced Glass Materials and Innovative Glass Melting Technology in the Year 2020", Brig, Switzerland, March 26-29, 2008.

better space utilization (avoid to many different trajectories in the melting tank and dead water zones) and techniques to speed-up sand grain dissolution and fining without increasing temperatures.

Conclusion: most industrial glass melting tanks operate far from optimal and furnace designs today allow very strong recirculation flows, keeping the glass melt longer in the tank than would be necessary. Less recirculation and improved heat transfer to the batch in order to minimize the need for hot spot glass re-circulation offers a potential for considerable energy savings and space reduction of the glass tank volume.

The paper first addresses the features of industrial melting tank furnaces operated today by more than 90 % of worlds' glass production and identifies the weak point of this concept existing since about 1865-1870.

Then the essential process steps will be described and the optimum conditions for each of the sections in the furnaces accommodating the process steps will be presented.

Finally some suggestions for improving glass furnace tank designs will be discussed and some dilemmas and problems still to be solved will be shown.

Essential elements seem to be:

- Improved heat transfer to and into batch blanket: most of the batch material melts (fuses) in the direct vicinity of the batch tip;
- Control of flow patterns and optimization of the hot spot plus spring zone position in the tank (for glass quality it is important that spring zone and hot spot locations in the melt are close together and that flow patterns are stable);
- Faster fining (gas removal from melt) processes;
- The availability of refractory materials allowing higher temperatures that enables faster fining (see Figures 3).

RESIDENCE TIME DISTRIBUTION IN CONVENTIONAL GLASS MELTING TANKS

In general, the molten glass obtained from the batch after fusion of these raw materials, does not follow a distinct trajectory in the melting tank [2]. Many different paths will be travelled by glass melt volumes in the same tank. In the worst case, the freshly molten glass moves from the batch blanket into the direction of the tank bottom and flows directly (short cut flow) along this relatively cold bottom of fossil fuel fired melting tanks, to the neck or throat to leave the melting-end. The temperature history of this melt shows generally that the glass has hardly been exposed to temperatures that allow all seeds or bubbles to be removed from this highly viscous melt. On the other hand, in case of a well-developed spring zone in the tank, located between the

batch tip and the exit of the tank, the molten glass flow along the bottom is interrupted by an upward flow at the spring location. The glass melt may reach the surface of the melt. Ideally, the spring zone position covers the hot spot area of the tank or is very close to the location of the highest temperatures in the melting tank. The upward flowing glass enters the highest temperature section of the melt and viscosity decreases, fining agent becomes active, fining gases are formed and growing bubbles can reach the glass melt surface.

Mathematical modeling shows that the largest part of the melt reaching the surface of the spring zone area, flows back to the tip of the batch blanket.

Figure 1 shows residence time distributions of three different industrial glass melting tanks. One can observe that the minimum residence time is typically 15 to 20 % of the average residence time.

Figure 2 shows as an example the projection of the flow pattern of a piece of glass melt in a typical float glass furnace with a residence time close to the average residence time. The figure shows that this 'particle'

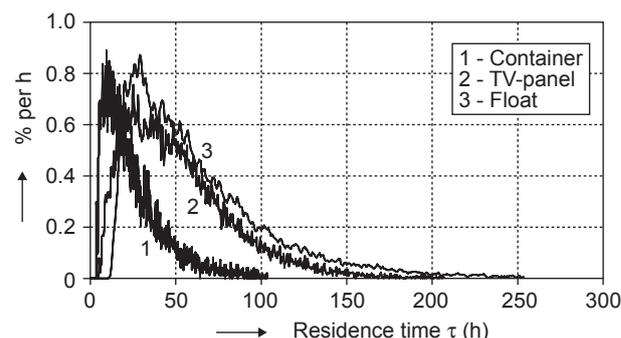


Figure 1. Residence time distribution determined by mathematical modeling [2] for 3 different melting tanks, producing: TV panel glass, clear float glass and container glass.

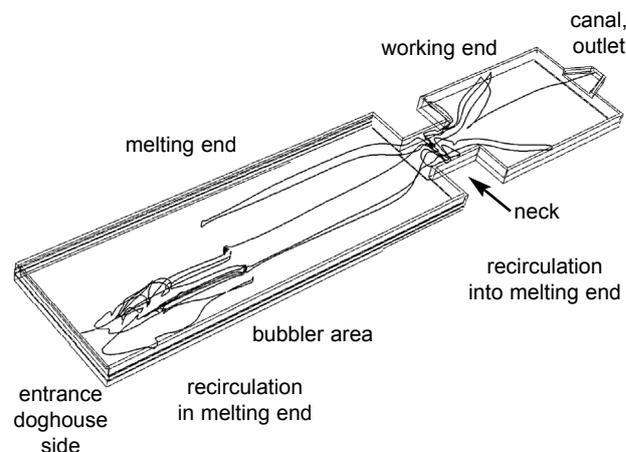


Figure 2. Flow pattern of glass melt volume with an average residence time. Note the strong re-circulation patterns in the float glass melt tank with a bubbler row.

(in modeling we call a small piece of melt or volume of melt a 'particle') re-circulates a few times between the batch zone and the spring zone of the tank.

Apart from the residence time, the melting behavior depends strongly on the temperature level at which the glass has been exposed: fining is characterized by a fining-onset temperature [3]. At this temperature, the fining agent dissolved in the molten glass starts to decompose and forms a large volume of gases (total internal gas pressure by gases in melt exceeds the 1 bar level) that diffuse in existing bubbles and seeds to be removed from the melt in order to achieve a homogeneous, bubble-free glass. In a static liquid, the ascension of the bubbles depends on the density difference between melt and bubble, glass melt viscosity and the radius of the bubble [4]:

$$V_{\text{ascension}} = 2/9 \cdot \Delta\rho \cdot g \cdot R^2 / \mu \quad (\text{Stokes law describes ascension of small seeds [3]}) \quad (1)$$

with: $\Delta\rho$ - density different between melt and bubble in kg/m^3 , g - acceleration of gravity (9.81 m/s^2), R - radius of bubble in m, μ - viscosity of melt in Pas.

A glass melt volume, that follows a trajectory that will not reach the fining onset temperature, will contain seeds that will hardly grow and the R-value (Equation (1)) may be too small, so that within the available time, the bubble can not reach the surface of the melt. Such a situation will lead to a glass product with small bubbles.

In order to characterize the ability of a glass melting tank, at prevalent operating conditions, to deliver a melt that has been well refined (bubbles removed), a fining index can be calculated for each possible trajectory in the tank [2]. In case that all trajectories show a value of their fining index larger than a certain threshold value, fining is probably sufficient.

Thus residence time distribution and fining index distribution modeling approaches [2,5] can be used to characterize the fining behavior of a glass furnace. Additional indices can be introduced for sand grain dissolution capability (dissolution index) [2] and homogenization (mixing index).

In Figure 2, the fining process (the bubble removal from the melt) mainly takes place in the spring zone area, at least when this area and the hot spot zone are close to each other. Thus, the melt that has been passed the top sections of the spring zone has also been exposed to ideal fining conditions: low viscosity, strong bubble growth (large bubble radius) and short distance to the surface to escape from the melt. However, in almost all industrial melting tanks, most of this high-quality glass melt, will flow downstream to the batch to be mixed at the batch tip with freshly molten glass. This freshly molten glass contains in the order of 10^5 - 10^6 seeds per litre [6] and probably some not completely

dissolved sand grains or alumina containing raw materials, and the high-quality glass is spoiled by this mixing process. On the other hand, this re-circulation flow from hot spot down to the batch tip is essential to maintain a short batch blanket length and for carrying sensible heat from the hot spot into the batch, to supply the required heat for the fusion reactions and heating of the batch. This re-circulation flow is the main cause of the very wide glass melt residence time distribution shown for most industrial glass melters.

Thus, in order to achieve an acceptable glass quality, a well developed spring zone in the vicinity of the hot spot area of the melting tank is required and a short cut flow along the bottom should be prevented, a return flow from the hot spot to the batch blanket provides part of the energy necessary for melting-in of the batch. Although, the batch is directly heated by the radiation from the flames or combustion space above the batch, this energy supply accounts generally for less than 50 % of the required energy for batch melting-in. Thus, extra energy is needed and supplied by the hot recirculating melt. This process of strong re-circulation determines the volume of the tank, and the flow pattern with the minimum fining index characterizes the effective glass quality that can be achieved, but only when the melt is not polluted somewhere downstream in the process (e.g. by dissolving refractory material or refractory stones).

In a well operating furnace (good fining), the glass melt with the minimum fining index passes the fining zone at least once, but on average the melt typically passes the hot spot/spring zone area 6 to 8 times.

This behavior presents the dilemma of the current glass melting process used in almost all glass industrial sectors, leading to wide residence time distributions for the glass melt and the relatively large glass melt tank volumes required.

TRANSITION FROM SINGLE-TANK FURNACE TO DESIGNS OF ADVANCED GLASS MELTING SYSTEMS

The wide residence time distribution in most glass furnaces using a melting-end consisting of a single tank, suggests that most of the material in the tank stays for a longer time than principally necessary. But, due to the recirculation flows in the tank, also required to bring heat from the hot spot area of the tank to the batch, the tank volume has to be rather large. Thus, glass melt has to be kept during a long time on a high temperature and the volume of the furnace is relatively large compared to the pull. The heat losses through the furnace walls and superstructure, therefore per ton molten glass are high: typically 0.6-1 GJ/ton container glass furnaces

and 1.75 to 2.25 GJ/ton float glass and even higher values for small furnaces, 2 to more than 2.5 GJ/ton molten glass. This energy loss has to be compensated by extra fuel consumption [7], which leads to extra volume flows of flue gases, resulting in extra flue gas heat losses. Thus, efficiently the contribution of the furnace structure heat losses to the energy consumption of a furnace is higher than the above presented specific heat losses.

The transition to innovative glass furnaces includes the following criteria to meet future demands:

- Significant decrease of the specific energy demand for melting;
- Using cheap energy to preserve economic feasibility of melting processes;
- Decrease of the flue gas volume flow to reduce the specific emissions of glass furnaces and reduce costs for Air Pollution Control (scrubbers, filters, DeNOx);
- Reduction of the volume of the glass furnace and a decrease of the surface area of the furnace to limit structural heat losses;
- Limit the re-circulation flows that cause the wide residence time distribution and that limit the flexibility of the melter: thus avoid relatively long transition times after changing glass composition or glass color;
- Control of each individual process step: a) melting-in of batch, b) dissolution of sand and alumina raw materials in silicate melt, c) removal of bubbles and dissolved gases [3] and d) homogenization and e) secondary fining (dissolution of remaining bubbles, containing chemical dissolvable gases, in gas lean melt);
- A uniform glass melt quality throughout the product: melting history being more uniform.

The dilemma, that in the current glass melting technology commonly used in the glass industry, the strong re-circulation flow is needed to provide sufficient heat to the batch should be overcome by modifications in the melting-in zone such as:

- Improve the heat transfer from the combustion space to the batch blanket;
- Apply other heating sources, able to transfer heat rapidly into the batch;
- Change the effective heat conductivity of the batch (e.g. using pelletized batch);
- Decrease the batch blanket thickness;
- Apply melting fluxes;
- Improve the stirring of the batch blanket or mixing of batch with the heating medium (submerged combustion melter [8]).
- Creating an increasing area of the batch tip, where melting rates for the batch seem to be the highest (higher temperature, high mass and heat transfer coefficients are met at the batch tip location).

Experimental studies show that special melting devices can be applied to melt 15 to 20 tons of batch per day per square meter applying intense heat transfer between the combustion process and batch [8]. The melt still requires a fining step to remove dissolved gases and gas bubbles after this batch melting stage. For example, for a container glass furnace melting 300 tons of glass per day, the melting-in zone could probably be limited to a tank surface area of about 15 m². Strong mixing, intense convection will speed up heat transfer and the dissolution process of the remaining raw materials (sand, alumina, feldspars, clay, etcetera) in the formed silicate melt.

A transition from the single melting tank furnace to an advanced glass furnace system, meeting the desired criteria also requires a controlled fining process. After melting-in and complete sand grain dissolution, removal of dissolved gases and gas bubbles by stripping by fining gases or by physical fining processes has to be completed. Contrary to the desire for strong mixing in the melting zone and the sand grain dissolution process, the fining process should be performed by a laminar flowing melt, preferably with a shallow tank depth to reduce the vertical trajectories that the bubbles have to travel in order to escape eventually through the glass melt surface. Here, in the fining section, the glass melt should reach a temperature above the fining onset temperature or the bubbles in the melt should start to grow within a very short time to bubbles $R > 0.5$ mm by bubble agglomeration or exposure to low pressure or fast diffusing gases (helium, water vapor). In a typical sulfate refined float glass melt (soda-lime-silica glass for clear sheet glass applications), the fining onset temperature is about 1420-1440°C in air-gas or air-fuel oil fired glass furnaces [9]. The bubble ascension should not be hindered by recirculation flows, pulling the melt (with some bubbles) also in downward direction.

Figure 3a shows the bubble ascension rate depending on glass melt viscosity and bubble size. Figure 3b presents the time required for gas bubbles to reach the glass melt surface, depending on the height of the melt in the fining zone. Very important, however, is that during the primary fining processes, all existing bubbles will grow and that generation of new seeds or bubbles is restricted. Thus, bubble formation at the glass melt - refractory interfaces in the fining section and downstream the fining zone should be prevented by the choice of suitable refractory materials and controlling the interface temperature to values below a certain critical level. The glass melt entering the fining zone should be free of grains (sand, alumina, aluminum-silicates) that still have to be dissolved, since these grains may generate (nucleation, or a decrease of CO₂ solubility in melt surrounding dissolving grains) new CO₂ seeds interfering with the fining process.

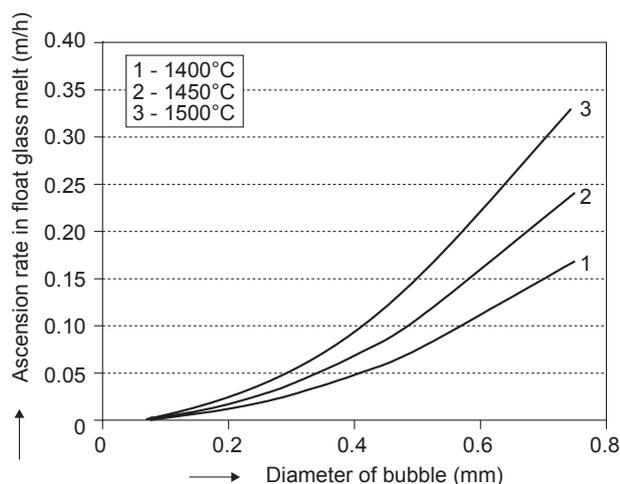


Figure 3a. Bubble ascension rates in viscous float glass melts.

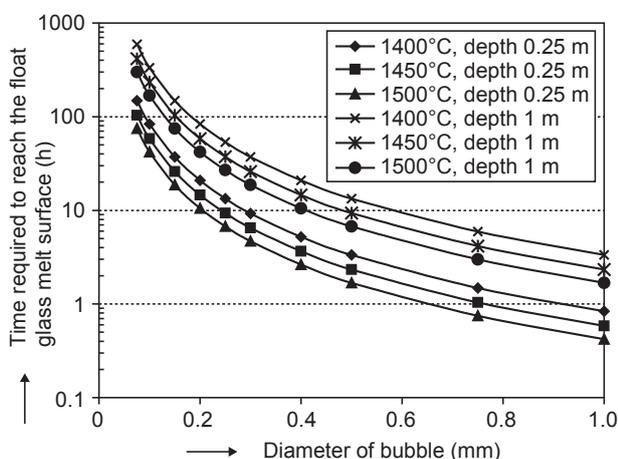


Figure 3b. Required fining time, depending on temperature, bubble radius and depth of fining tank for soda-lime-silica glass.

ENERGY CONSUMPTION OF FUTURE FURNACES

CFD modeling calculations have been performed to design a glass furnace with different sections for batch melting plus sand grain dissolution, primary fining and refining and conditioning. The CFD modeling enables the identification of the critical trajectories and can be used to change design, dimensions and location of heating devices to obtain relatively small tank volumes for melting and fining and for creating optimum conditions for melting and for fining.

The sections for melting and sand grain dissolution are/should be preferably connected to the fining zone by a channel that prevents re-circulation of glass melt. The residence time and temperatures in the compact fining zone with adapted glass melt level should be sufficient to complete the primary fining process, using sulfate fining or using helium as an aid of the fining process.

Helium atmospheres above the melt [9, 10], accelerate the fining process by fast diffusion of helium in the molten glass (helium shows a high diffusion coefficient and relatively high solubility). The high helium permeability will cause a fast pick-up of helium of the melt and the helium diffuses subsequently into the existing seeds. These seeds will grow by the helium gas take-up and the helium will dilute the other gas species in these seeds/bubbles. The decrease of the partial pressures of these gases in these bubbles will stimulate the diffusion of these gas species from the melt (melt contains dissolved CO_2 , N_2 , Ar, H_2O and oxygen) into the bubbles. This process will aid the bubble growth rate and will result in gas stripping from the melt.

For an example of a fossil-fuel fired float glass, furnace, the energy costs can be decreased by about 20 % according to energy balance [7] and CFD modeling [2], after transition of the conventional tank furnace to a segmented furnace with a smaller total glass melt volume compared to the conventional tank furnace.

REQUIRED CONDITIONS FOR ELEMENTARY PROCESS STEPS IN GLASS MELTING PROCESSES

In this paragraph, the required conditions of each step of the most essential processes in glass melting are summarized:

Melting-in of batch materials

For soda-lime-silica glasses, the melting-in of the batch [11] requires about 0.5 MJ energy per kg glass for endothermic reactions, about 1.5 MJ/kg for heating of the batch until almost melting reactions have been completed and 0.3 MJ for heating of the released batch gases, further heating of the melt is often required for fining and bringing the melt to the required temperature and viscosity level. These figures refer to normal dry batch without cullet.

This minimum required energy (excluding heat losses) of 1.8 to 2.5 MJ/kg glass, depending on the cullet fraction in the batch has to be supplied in the first zone of the melting process. In order to limit heating times and to avoid the necessity to use intense re-circulating hot glass melt for heating of the batch, a change from a thick batch blanket situation to a shallow batch, or pelletized batch blanket or a stirred batch may be considered. Extra heating by boosting electrodes just underneath the batch blanket may replace part of the heat supply by the re-circulating hot melt.

Mathematical descriptions of the heat transfer to the batch from the hot glass melt and mass transfer relations, show that the batch blanket thickness decreases

rapidly at the very end of the batch blanket. Close to the batch tip (position at glass melt surface where batch meets the blank melt), the melting of batch is much stronger than upstream the batch tip. Therefore, melting of the batch blanket can be speeded up by creating a long batch tip length or by breaking up the batch.

Mixing of batch supports heat transfer and melting-in of up to 20 tons per m² per day might be achieved [8, 12]. However, violent motion of powder batch, with different raw material particle sizes may lead to intense carry-over, de-mixing or depletion of one or more raw materials prone to carry-over. Therefore pre-pelletizing of raw materials using flue gas heat in the pelletizing process can be recommended. Flue gases can be used to dry and preheated the raw material spherical pellets.

Preheated pellets (about 300°C) need about 15-20 % less energy than normal batch (energy saving potential depends also on glass furnace construction), and show better heating rates than batch blankets.

Sand grain dissolution

After melting-in of most of the batch materials, a silica rich melt is formed and often not all sand grains in the raw material batch are completely dissolved in the silicate melt flowing downstream the batch blanket. The batch free time or sand grain removal time depends on the SiO₂ solubility (depends on the glass composition and temperature), the SiO₂ content of the freshly formed melt, diffusion coefficient of dissolved SiO₂ in the silicate melt, the sand grain size and convection of the melt. The applied sand should not contain too much fine silica grains, because this will lead to very fast increase

of the SiO₂ content of the melt (approaches silica saturation), slowing down further dissolution of the coarse sand grain fraction. On the other hand sand grain sizes should not be too large, to avoid long dissolution times (dissolution time is over-proportional to sand grain diameter). Sand grain size distribution should be narrow, and typically in the range of about 100-150 micrometer diameter for most glass types. For E-glass fiber production, finer sand is preferred, but this may lead to extra carry-over or silica scum (agglomeration of floating sand grains on top of the molten glass) formation.

Strong mixing (velocity gradients in the melt), such as accomplished in the submerged combustion melter [8, 12] will aid the dissolution of sand grains, but the shortest residence time may be too short to ensure complete dissolution of all sand grains after the 'mixed melter' section. It is recommended to optimize conditions or to apply an extra zone to complete sand grain dissolution before the melt enters the fining zone. The dissolution of sand grains depending on the convection patterns in the melt, characterized by the glass melt velocity gradient of the melt or the glass melt velocity relative to the sand grain can be calculated by a model with input: velocity gradient of the melt, diffusion coefficient of SiO₂ in the melt, initial sand grain radius, viscosity of the melt and maximum SiO₂ solubility. Several of these parameters depend on the glass composition and temperature [2, 13].

Each glass composition (depending on glass melt oxidation-reduction state, glass composition, content of fining agent, furnace atmosphere composition) shows a fining-onset temperature that can be determined by well-designed laboratory experiments [9].

The glass should not reach the fining onset temperature in the melting-in zone or sand grain dissolution sections, because this can lead to foaming in these areas and loss of active fining agent from the melt, before entering the fining segment.

Thus, temperature control in the melting-in zones is essential for the quality of the following process steps.

Primary fining

The fast removal of gases and gas bubbles from the melt, during the primary fining process, requires a melt that is heated at temperatures above the fining onset-temperature (this might be more than 1400-1440°C for typical soda-lime-silica float glass melts). Ideally the melt is heated above this temperature just before entering the dedicated fining section of the furnace. Bottom temperatures of the fining tank should exceed this temperature level, therefore the tank depth in that area should be limited and the bottom refractory material should not react or release gases at these temperature levels. The steady ascension of growing bubbles

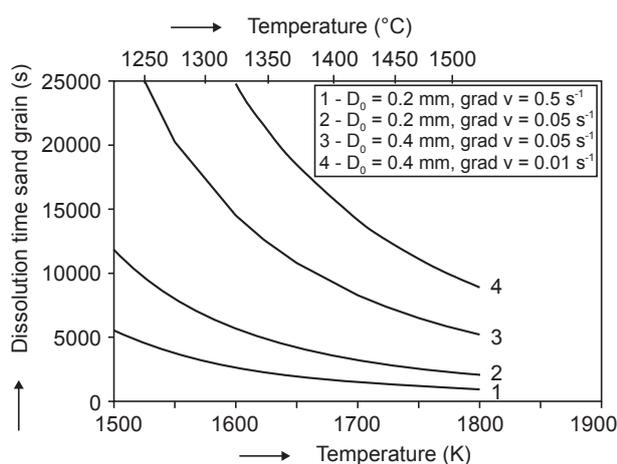


Figure 4. Modeling results for dissolution times of sand grains in molten float glass melt (soda-lime-silica) for different velocity gradients ($grad\ v$) and initial sand grain diameters (D_0) as a function of temperature. Calculated with sand grain - glass melt dissolution models [2, 13]. $grad\ v$ is velocity gradient in melt.

in the melt should also not be interrupted or hindered by downward directed flow. Almost plug flow/laminar flow conditions are necessary in the fining section. The dimensions of the fining section can be derived from the bubble size, ascension times (depending on size and glass melt temperature) see Figure 3 and pull: all bubbles much have reached the glass melt surface by the end of the fining section. In shallow fining zones, the temperature difference between the glass melt surface and bottom can be limited and avoid the need for excessive glass melt surface temperatures to reach sufficiently high bottom temperatures, because such high surface temperatures may lead the strong evaporation of volatile species such as alkali or boron components [14]. It is important to identify the most critical path of the melt in a melting tank, that may ever not exceed the required fining onset temperature, see Figure 5.

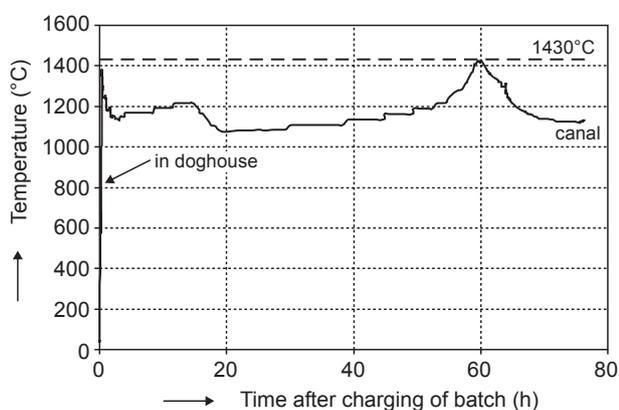


Figure 5. Temperature history of a critical path of glass melt in a float glass melting tank. In this case, the fining onset temperature should be below 1430°C to ensure removal of gas bubbles in all the glass melt by primary fining.

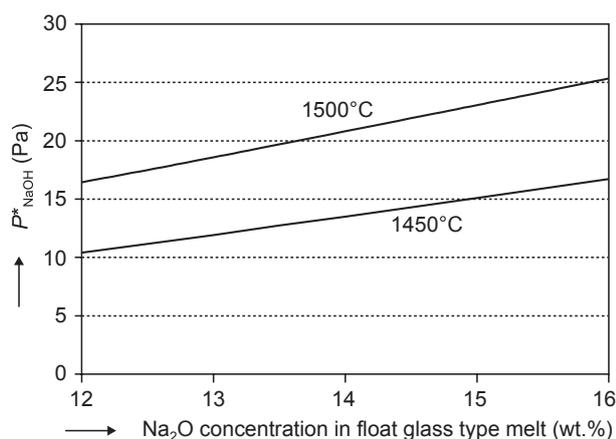


Figure 6. Sodium hydroxide vapor pressure above molten soda-lime-silica glass melt. Concentration ratios between other oxides than sodium oxide remain constant. Calculated by thermodynamic model for silicate melts: TNO modified Associated Species Model [15].

It is recommended to develop fining processes that prevent the need of surface temperatures above 1475-1500°C. A decrease of glass melt surface temperature of a float glass melt from 1500 down to 1450°C, reduces sodium evaporation by about 35% and will decrease dust formation (from sodium condensation reactions e.g. sodium sulfate condensation) and will decrease silica corrosion in the superstructure by sodium species. Figure 6 shows the saturation pressure of NaOH in a soda-lime-silica glass furnace fired with air and natural gas for different temperatures. Sodium hydroxide evaporation is the main cause of fine dust emissions of container, float and soda-lime-silica tableware glass furnaces.

The fining process may be speeded up by: a) enhancing agglomeration of bubbles or bubble growth under the influence of acoustic waves transferred by waveguides into the melt or b) by changing the furnace atmosphere to a sub-atmospheric pressure space above the melt [4] or c) using a special gas atmosphere or d) pre-conditioning the melt by bubbling the melt with a gas showing high solubility and diffusion in the melt [9, 10].

After primary fining, not only most of the fine seeds and bubbles have to be removed from the melt, but also the content of dissolved gases (e.g. CO₂ and N₂) should have been reduced to allow complete dissolution of still remaining fine seeds during the controlled cooling process after primary fining.

Conditioning of melt

Just, before the next stage in the glass production process: the forming or shaping of viscous glass, the molten glass should be chemically homogenized and conditioned to a required (often uniform) temperature level. The cooling process should not be too fast to avoid strong temperature non-uniformities or to freeze-in the remaining seeds, without having the chance to be completely dissolved.

Today, working-ends, refiners, feeders or forehearths are used to bring the glass melt temperatures down to the desired level, prior to forming. The importance of a uniform or controlled temperature gradient in the spout of the feeders or canal of the working end on the quality of the forming process is still not completely understood. Due to the re-heating of the melt at the sidewalls of the working-end and feeders by small burners or heating electrodes and the cooling of the hotter melt flowing in the axis of the feeders or forehearths, extra energy is required for the process of conditioning and controlled cooling of the melt to the required temperature level. A certain temperature level is required (depending on type of product and kind of forming process) to achieve the proper viscosity for forming and shaping of the glass products from the delivered melt.

The energy efficiency (balance between cooling and heating) might be improved by using CFD models for feeder designs or by advanced temperature control in the different feeders sections.

Stirrers in the feeders and special stirring compartments in feeders can be applied to improve mixing especially for glass products requiring high optical homogeneity (display glasses, float glasses, tableware glasses or color-feeders with coloring fritte addition to the melt at the entrance of the feeder).

ALTERNATIVE GLASS MELTER CONCEPTS

In the past, many attempts have been made to separate the process steps involved in the conversion of a mixed batch of raw materials into a uniform glass melt. Barton [16] showed an overview of developments until about 1990. Such melters are often indicated or referred to as segmented melters, examples are:

- P10 melter, with rotary kilns, one for burning of limestone and dolomite in a sand mixture and one for pre-reacting part of the sand with soda. The materials from both kilns are mixed and melted in a rotary melter and heated up to about 1250°C. Then, the molten mass is flowing into a channel to increase temperature by electrodes and submerged combustion burners (oxygen). In the last stage the melt flows into a shaft furnace that is kept at sub-atmospheric pressure. In this stage bubbles grow rapidly and gases are stripped from the melt. The bubbles are removed from the melt that can now be poured on the molten tin for the float glass forming process.
- FAR melter from Saint Gobain, with batch compaction and batch preheating by flue gases exhausted by a recuperator connected to segmented type glass melter. After batch preheating, a thin layer of batch (compacted) is heated by burners before it enters a foaming compartment (sulfate foaming), that is heated by glass melt electrodes. After foaming, the melt enters the clearing zone with a shallow glass melt depth to remove residual bubbles and then it flows through a zone with extra stirring (homogenization).
- FARE melter from Saint Gobain, similar to the FAR process, but without fuel burners in the melting zone, the melting zone is stirred and there the melt is heated by electricity.
- RAMAR-Rapid Melting and Refining from OI (Owens Illinois): based on different compartments, starting with electric melting in 2 stirred basins and then the degassing takes place in a centrifugal (re)finer. The centrifugal 'forces' remove the bubbles from the melt.
- Pilkington, developed a melting process in which the batch is charged into the furnace creating a sloped batch surface. The batch is heated from above or at the sides by burners. The obtained melt layer flows by the slope downwards in a compartment for complete melting and fining.
- Advanced glass melter [17]. Raw materials (without cullet or carbonates) are injected in a flame (natural gas), the residence time in this flame is much less than 1 second. Carbonates are added separately, but only at the end of the combustion process to avoid destabilization of the flame by the gas decomposition product CO₂. The suspended particles (already molten) and gases are blown into a melting chamber through a small opening. The melt is collected on a ceramic target (cone-shaped). The obtained melt flows as a thin layer downwards but needs additional fining for most glass types, however this technology is not further pursued.
- SEG melter: This is a combination of a cold-top electric melter equipped with glass melt electrodes for melting normal batch, and a fossil fuel fired melter. The pre-molten batch is mixed with cullet in the fossil-fuel fired tank furnace segment. The cullet can be preheated by flue gases. The mixtures of batch melt and cullet melt is further heated by oxygen burners to finalize fining and homogenization. Return flow from the fining zone to the melting-in zone is prevented. The mixing in the mixing zone is enhanced by bubblers. Advantages are: the low emissions, since the melting-in zone is a cold top melter, low NO_x emissions, because of oxygen-fired fining compartment, relatively low volumes of the tanks and narrow residence time distribution, because of the limited return flows. The author reports some other aspects [18].
- British Glass plasma melter [19]. Three plasma electrodes (120° angle between electrodes projected on horizontal plane) are immersed in a batch pile contained in a ceramic container. Nozzles in the electrodes supply ionized argon gas. Due to an alternating current between the electrodes, the plasma travels between these electrodes. The plasma transfers within the batch pile the heat to the material, forming a silicate melt that becomes electrically conductive. The electric current will partly pass through the melt and will heat the melt by the Joule principle. The melt flows from an orifice in the bottom of the ceramic container to a next process step. The quality of the melt depends on the residence time. The molten glass is covered by cold batch preventing volatilization losses. A special starting-up procedure is required. Often extra fining after this kind of melting processes is required to produce glass products, such as tableware or container glass. Total energy (mainly electricity) costs (plus argon) can be rather high but the melter is very compact and flexible.

Segmented melters, still applied in the glass industry are the so-called LoNOx[®] melter [20] and FLEX[®] melters of SORG [21].

The LoNOx[®] melter applies a recuperator for combustion air preheating, but the flue gas from the recuperator is used to preheat cullet, therefore these types of furnaces are especially applicable for glass production from cullet rich batches.

The fining zone, exists of a shallow fining bank heated with fossil fuel - preheated air burners. The hot gases from this zone flow into the space above the melting-in zone, with a deeper glass melt tank that is equipped with electrodes. The raw materials batch extracts heat from these combustion gases before these gases enter the recuperator at reduced temperatures. These LoNOx[®] melters show similar energy efficiencies, as energy benchmark studies (TNO studies on benchmarking energy efficiency of glass furnaces [22]) show, compared to end-port fired regenerative furnace, but also very low NOx emissions due to the relatively low air preheat temperatures.

Fining processes can be supported by physical fining methods [3], for instance by applying a sub-atmospheric pressure above the melt as demonstrated by Asahi [23].

After the year 2000, rapid glass melting has become increasingly interest in the USA and systems using high temperature, intense plasmas have been further developed to heat batch within a few minutes up to melting temperatures [24]. Oxygen-fuel burners

mounted in the bottom of a water cooled melting tank are applied in the so called submerged combustion glass melter (SCM). The batch and melt are very intensively heated by oxygen-gas flames ascending (as a column) through the batch and melt in the about 1 meter deep tank [8, 12]. The very intensive and rapid radiative and convective heat transfer from the flames channeling through the melting batch, will enable fast melting, and the batch can be heated and melted within about 1 to 2 hours at very high specific melting loads of 15 to 20 metric tons per m² per day. The melt delivered by this system contains many seeds and bubbles and requires in most cases a subsequent fining step to remove gases from this melt to be suitable for fiber, tube, container or tableware glass production. The energy efficiency for a relatively small melter (footprint: 1-2 m²) is reported on about 34-40 % (energy for fusion reactions and heating of batch and melt and batch gases relative to total combustion energy supplied). This energy efficiency for melting will further improve for larger furnace sizes, with a larger volume-surface ratio. The walls of the submerged combustion melter are refractory lined, but water-cooled. Figure 7 shows a schematic drawing of the Surmerged Combustion Melter.

The application of high intensity plasma's for heating of batch and glass melt was investigated by British Glass [19] and Plasmelt [24], showing very fast melting rates and short residence times (down to 5-15 minutes). Even glass compositions or refractory materials that require temperature of up to 1900°C can be processed. In the Plasmelt technology, the plasma is generated between a DC anode and cathode torch. Argon gas is used for the plasma. The torch positions are generally fixed and the plasma touches the raw material batch and melt contained in a rotating skull. The torches do not contact the melt. Especially, this plasma technology seems to be suitable for glasses that require high-temperature melting temperatures or for special glass types with low production volumes. This technology also offers a high level of flexibility by the small hold-up volumes and presents therefore possibility of glass type changes within a few hours. Energy consumption (based on primary energy, taking into account the energy efficiency of electric power generation), however is much higher compared to fossil fuel fired glass furnaces for large volume glass production.

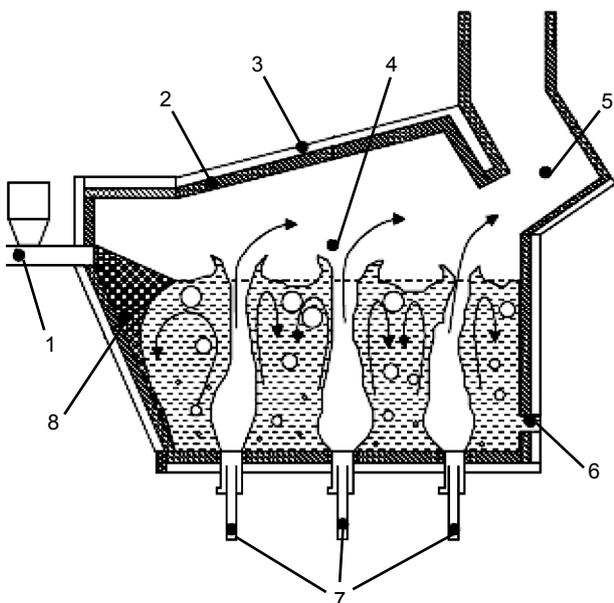


Figure 7. Submerged Combustion Melter (SCM) [8]. 1 - Batch charger; 2 - Refractory lining; 3 - Water cooled panels; 4 - Melting zone; 5 - Exhaust; 6 - Melt outlet; 7 - Submerged oxygen-gas burners; 8 - Batch material (sloped pocket of batch).

Most of the furnace designs developed before 1990 were based on empirical approaches or using physical models that were limited in predicting glass melt quality and energy consumption.

Today, CFD based mathematical models can be applied as has been demonstrated for the submerged combustion melter and plasma melter and developments at TNO enable the design of new furnaces and to opti-

mize not only furnace design (combustion chamber and melting tank segments), but also process settings and to predict glass melt quality based on melting and fining index distributions, residence time distribution calculations and determining evaporation from the melt [14, 15] and refractory corrosion rate behavior [2, 25].

The recognition of the different basic requirements for each process step: batch melting, sand grain dissolution and removal of gases and homogenization, leads to proposals for segmented melters, consisting of compact compartments that are optimized by the use of CFD modeling and advanced sensor and control systems with specific functionalities. Because of: a) the developments in glass melt sensors (electrochemical redox sensors or polyvalent element sensors [26-29]), b) reliable models to optimize design and to predict the effect of furnace design and process settings on energy consumption and glass melt quality plus c) the simulation model-based process control systems, new glass melting concepts can be designed. The aim of such innovative melters is and should be: improved energy efficiency, increased stability and control of glass quality, flexibility by fast glass type changes, low emissions and increased furnace lifetime.

CONCLUSIONS

Most glass melting tanks today show very wide glass melt residence time distributions. The average residence time can be 6 or 8 times the minimum residence time for most industrial melting processes. A ratio of minimum versus average residence time of 0.4 would allow a reduction in the glass melting tank volume of more than 50 %. This means less capital costs for the construction of the furnace by smaller quantities of refractory materials and less heat losses through the walls of a compacter furnace. A change of the melting tank volume by about 50 % may decrease total energy consumption by about 15 %.

But, in order to enable this transition from a conventional melting tank design to an "advanced" melter with a smaller volume and better energy performance, the batch melting-in process has to be changed to realize this. Without strong recirculation glass melt flows (this re-circulation dominates the wide residence time distribution), the direct heat input from the combustion space into the batch blanket has to be intensified by special technologies (an example is the submerged combustion melter).

Moreover, each essential process step requires its own conditions, characterized by its temperature level, degree of mixing, chemistry (redox state, fining agent, melting fluxes, viscosity dependent on composition) and time required to complete the process step at the given conditions. Optimum conditions for fining (gas

removal from melt) are very different compared to the preferred conditions of melting-in and sand grain dissolution or for homogenization.

For instance for soda-lime-silica glass melting, the following conditions seem to be adequate [1, 30, 31]:

Batch melting

- 1 hour residence time for fusion of most batch materials into a silicate melts, with preheated batch, this time can be reduced;
- Thin batch layer (< 0.05 m) or a very strong convection around the batch, this violent convection disrupts the batch blanket, creating a large area of batch tip;
- Compacted batch (intense grain contact and fast heat conduction).

The heating process from room temperature to the temperature levels where most batch reactions take place consumes most of the time, due to the poor conductivity of the low temperature areas of the batch. Batch preheating is very efficient to overcome part of the initial stages of the batch heating process in the glass furnace.

Sand grain dissolution

- Depending on maximum sand grain size, the average residence time would be about 1-1.5 hours ($D < 0.2$ mm);
- Strong convection by stirring or bubbling promotes sand grain dissolution strongly;
- Sand grain dissolution preferably takes place (soda-lime-silica glass melts) at $1200 < T < 1350^{\circ}\text{C}$; at higher temperatures the fining agent may already start to decompose and that will jeopardize the subsequent fining processes;
- A relatively narrow sand grain size distribution, (especially avoiding large sand particles): 80-250 microns is recommended to avoid carry-over, to limit fast silica saturation of the melt by the rapid dissolution of the fine grains, too prevent long dissolution times for too large grains and to avoid silica scum formation by sintering silica fines at the glass melt surface.

Fining (primary) process

- For deep tanks: at least average residence time 3 or more hours for good fining;
- But for glass layers < 0.3 - 0.4 meter and temperatures well above the fining onset temperature: 1.5-2.5 hours for complete bubble removal;
- The tank bottom temperatures at the fining shelf should be above the fining onset temperature to avoid a current of glass melt with remaining seeds along the bottom;

- The surface temperature of the melt is recommended to keep below 1480–1500°C, to limit evaporation of sodium (NaOH, Na, NaCl) or boron (in borosilicate furnaces), that may cause refractory corrosion in combustion chamber or regenerators and which governs dust formation (sodium vapor will form fine sodium sulfate dust or boron vapors create borates or boric acid in the flue gases).
- Residence times about 2-3 hours in a shallow fining zone requires only about 60-80 m² fining area for 500 metric tons float glass melt per day.
- Preconditioning of the melt by helium bubbling (< 1350°C to avoid too much decomposition of fining agent during bubbling) will support fining in the fining zone or a shallow atmosphere of helium (or < 1 bar) above the melt in fining zone will support the degassing of the melt.
- In the fining zone, plug flow conditions should prevail and mixing or stirring should be avoided;
- In the primary fining process, a large quantity of dissolved gases (CO₂, N₂) should be stripped to obtain a melt lean of gases.

Refining/Conditioning

- After a primary fining process, the melt should have a low concentration of dissolved gases (stripping of gases during primary fining);
- Fast cooling will freeze in remaining seeds and should be avoided;
- By slowly decreasing temperature of the melt, the small bubbles in the glass melt, that contain gases with increasing solubility in the silicate melt upon cooling, may be re-absorbed, this typically takes place at: 1325-1275°C for float glass melts. A residence time of about 1 hour is recommended at this temperature range. Below these temperatures, diffusion of gases in the melt becomes too slow.

The total residence time without re-circulation between the different process steps can be limited to about 6-8 hours for high quality soda–lime–silica glass melts.

The fining process requires the longest time and may be accelerated by heating technologies providing higher glass melt temperatures in the fining zone or by physical techniques such as sub-atmospheric pressure fining as applied in the P10 [16] process.

Acknowledgement

The author addresses his appreciation for the contributions of Ing. Andries Habraken and Dr. Hans van Limpt, both from TNO.

References

1. Němec L., Jebavá M.: *Glass Technol.: Eur. J. Glass Sci. Technol. A* 47, 68 (2006).
2. Beerrens R. G. C. in: *Contribution in Mathematical Simulation in Glass Technology*, pp. 17-73, Ed. D. Krause, H. Loch, Schott Series on Glass and Ceramics, Science, Technology and Applications, Springer Verlag 2002.
3. Beerrens R. G. C.: *Ceramic Transactions* 141, 3 (2004).
4. Rongen M., Beerrens R. G. C., Faber A., Breeuwer R.: *Physical Fining Techniques of Glass Melts*, Proc. 8th European Society of Glass Science & Technology Conference session B2-Refining and melting II, paper B2-3, Sunderland, September 10-14, 2006.
5. Muschick W.: *The Validation of Glass Furnace Simulations in a Multilevel Process*, Proceedings XXth A.T.I.V. Conference on Modern Technologies and Techniques for Glass Manufacturing, pp. 53-55, Parma, September 14-16, 2005.
6. Mulfinger H-O.: *Glastechn. Ber.* 49, 232 (1976).
7. Beerrens R. G. C.: *Analysis of energy consumption and energy saving measures for glass furnaces*, 80th Deutsche Glastechnische Tagung der DGG (Deutsche Glastechnische Gesellschaft), Dresden Germany, 12.-14. June 12-14, 2006.
8. Rue D., Kunc W., Aronchik G.: *Operation of a Pilot-Scale Submerged Combustion Melter*, 68th Conference on Glass Problems, Columbus, Ohio, USA, October 16.-17. 2007.
9. Rongen M., Beerrens R., Faber A. J.: *Observation of Chemical and Physical Fining Processes in Glass Melts*, 81th Glastechnische Tagung, Aachen, June 4-6, 2007.
10. Kawaguchi M., Aiuchi K., Nishimura Y., Aoki S.: *Helium gas transfer into an E-glass melt with bubbling technique*. Proceedings of the XXIst International Congress on Glass, Strasbourg, July 1-6, 2007.
11. Verheijen O.: *Thermal and chemical behavior of glass forming batches*. PhD thesis Eindhoven University of Technology, June 25, 2003. ISBN 90-386-2555-3.
12. Purnode B., Aronchik G., Collins L., Venuturumilli R., Jaydeep J.: *Numerical Simulation of the Submerged Combustion Melting Process*, 68th Conference on Glass Problems, Fawcett Center for Tomorrow, The Ohio State University, Columbus Ohio, USA, October 16-17, 2007.
13. Beerrens R. G. C., Muijsenberg H. P. H.; van der Heijden T.: *Glastech. Ber. Glass Sci. Technol.* 67, 179 (1994).
14. Beerrens R. G. C., Limpt van J.: *Glastech. Ber. Glass Sci. Technol.* 74, 245 (2001).
15. Limpt Van, J. A. C.: *Modeling of evaporation processes in glass melting Furnaces*, PhD thesis Eindhoven University of Technology, November 26, 2007.
16. Barton J. L.: *Glass Technol.* 34, 170 (1993).
17. Bender D. J., Hnat J. G., Litka A. F., Donaldson Jr. L. W., Ridderbusch G. L., Tessari D. J., Sacks J. R.: *Glass Industry*, March 1991, pp. 10-37.
18. Argent R. D., Dickinson G.: *Time-Energy Savings and RCB&NOx control on the Color Cell of Liberty Glass in Sapulpa Oklahoma*, 9th Annual Glass Technology Seminar, Nemaocolin Woods, Farmington, Pa, August 18, 2003.
19. PBI-report R93.002: *Glass production with plasma's, investigation into possibly large energy savings and the technical and economical feasibility*, June 1993 (for NOVEM project: 32205/7085).
20. Ehrig R, Wiegand J., Neubauer E.: *Glass Sci. Technol. (Glastech. Ber.)* 68, 73 (1995).

21. Pieper H.: *The Flex-Melter: A new melting furnace for the glass industry, an alternative to pot furnaces*. Glastech. Ber. Sonderband LXIII K (1990). pp. 144-1562, 1990.
22. Beerkens R. G. C., Limpt van H. A. C., Jacobs G.: *Glass Sci. Technol.* 77, 47 (2004).
23. Kawaguchi T, Okada M., Ishimura K., Kokubu Y.: *Refining of glasses under subatmospheric pressures-IV*, Proceedings XVIII International Congress on Glass, pp. 39-44, San Francisco, July 5-10, 1998.
24. Gonterman R et al.: *High Intensity Plasma Glass Melter Project*. Final DOE (USA Department of Energy) Report. Award number: DE-FC36-03GO13093, Plasmelt Glass Technologies, LLC report. Boulder Co, USA, October 27, 2006.
25. Beerkens R. G. C.: *Silica Loss in Crowns by Reactions with Glass Melt Vapours*, Proc. 66th Conference on Glass Problems, pp. 69, Wiley-Interscience, John Wiley & Sons Inc. Hoboken NJ, USA, 2006.
26. Lenhart A., Schaeffer H. A.: *Glastechn. Ber.* 58, 139 (1985).
27. Claussen O., Rüssel C.: *Glastechn. Ber. Glass Sci. Technol.* 70, 231 (1997).
28. Bauer J.: *Measuring the Sulfur Content of Industrial Glass Melts Using Square Wave Voltammetry*, Proc. 61st Conference on Glass Problems, pp. 205-219, The Amer. Ceram. Soc. 2001.
29. Laimböck P. R., Beerkens R. G. C.: *Am. Ceram. Soc. Bull.* 85, 33 (2006).
30. Beerkens R. G. C.: *Analysis of industrial glass melting processes and clues for new melting concepts*, XX. ATIV Conference Modern Technologies and Techniques for Glass Manufacturing, Parma Italy, September 14-16, 2005.
31. Beerkens, R. G. C.: *A deeper look into industrial glass melting processes - Evaluation of the key processes in industrial glass melting tanks*, Proc. XXth International Congress on Glass, Kyoto Japan, September 26 - October 1, 2004.

ANALÝZA ELEMENTÁRNÍCH PROCESNÍCH KROKŮ
V PRŮMYSLOVÝCH SKLÁŘSKÝCH TAVICÍCH VANÁCH
- NĚKTERÉ PŘEDSTAVY PRO INOVACI
PRŮMYSLOVÉHO TAVENÍ SKLA

RUUD BEERKENS

*TNO Science & Industry, Glass Group,
De Rondom 1, P.O. Box 6235, 5600 HE Eindhoven,
The Netherlands*

Konvenční průmyslové sklářské pece vykazují značné doby zdržení skloviny v tavicí vaně a střední doba zdržení může být u vysoce kvalitních sklářských výrobků, jako je plavené sklo nebo sklo na televizní obrazovky, i více než dva dny, přestože minimální doby zdržení se pohybují v intervalu 8-10 hodin (nebo i méně než 4 hodiny u pecí na obalové sklo). Dlouhé doby zdržení jsou spojovány s velkými poměry objemu taveniny k rychlosti odběru a s vysokými konstrukčními tepelnými ztrátami. Recirkulační proudy nezbytné pro dostatečnou dodávku energie do vrstvy kmene pro proces tavení materiálu kmene jsou často špatně kontrolovány a způsobují tyto vykazované rozdíly v dobách zdržení. U každého elementárního procesního kroku jsou analyzovány nejdůležitější procesní parametry, pokud jde o požadovaný charakter proudění, chování při míchání, teplotu, chemické děje a čas potřebný na daný procesní krok. Porovnáváme-li tyto procesy mezi sebou, z analýzy vyplývá, že různé kroky vyžadují velmi různé podmínky. Proto by úpravy konstrukce sklářské pece měly být zaměřeny na vývoj segmentovaných pecí, každý segment by měl být vyhrazen pro určité stadium tavení a měl by být pro toto stadium optimalizován: roztavení kmene, rozpuštění pískových zrn, odplynění (bubliny a rozpuštěné plyny), reabsorpce zbytkových bublin a homogenizace (chemická a tepelná) skloviny. Budou ukázány a krátce diskutovány některé pokroky ve vývoji zaměřené na rychlé čerání a rychlé roztavení kmene.