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Use of Concentrated Radiation for Solar Powered Glass Melting Experiments S.Q.S.Ahmad¹, R.J. Hand¹, C. Wieckert² ¹Department of Materials Science and Engineering, University of Sheffield, Sir Robert Hadfield Building, Mappin Street, Sheffield, S1 3JD, UK ²Solar Technology Laboratory, Paul Scherrer Institute, Villigen, CH-5232 Villigen PSI, Switzerland

Abstract

To investigate the feasibility of using concentrated solar radiation to provide process heat for glass production, a high flux solar simulator was used to melt glass forming batches. Initial experiments involved melting various glass forming batches which demonstrated that rapid and full conversion of the crystalline raw materials into an x-ray amorphous vitreous state was possible. A pure silica batch produced an x-ray amorphous product but it was not possible to refine the melt in these exploratory tests. A powdered, ternary soda-lime-silica (SLS) glass forming batch melted vigorously, with rapid gas removal, resulting in a completely transparent glass. Industrial SLS pellets were subsequently used in semi-continuous melting experiments, whereby the batch was intermittently fed into the melting zone while the beam was kept on. Additional secondary heating and insulation around outlet of the melting zone was required to achieve a semi-continuous flow of molten glass to an output crucible.

Key words: Concentrated solar thermal, Glass production, High flux solar simulator, High temperature melting.

1. Introduction

Glass manufacture is a highly energy intensive process, requiring primarily, high temperature process heat, usually provided by burning fossil fuels. The most common type of glass produced is soda-lime-silica (SLS); this consists of three main components: silica sand, which forms the glass network; sodium oxide, a network modifier, usually supplied as sodium carbonate (soda ash), which reduces the temperature at which a melt is formed; and calcium oxide, also usually supplied as a carbonate which improves the chemical stability of the glass produced. Other more minor components are also added to commercial melts, either deliberately or as unavoidable components in the raw materials. The theoretical energy required to convert these raw materials into glass is around 2.7 GJ/t (Trier 1987a), (Scalet et al. 2013). This theoretical value is based only on the chemical heat of reaction plus the enthalpy changes associated with heating up the raw materials ('batch') and the gas produced, from ambient temperature to the generally assumed melting temperature of 1500°C. However, in practice, significant additional energy is required to overcome heat losses associated with maintaining the glass melt at temperature to enable homogenisation and bubble removal (fining) to occur, in order to produce the required high quality products expected by the modern consumer. This real energy consumption for modern industrial glass melting, with typical production in the order of hundreds of tonnes per day (tpd), can vary from 3.5 to 40 GJ/t (Scalet et al. 2013) depending on furnace design and scale, with larger furnaces generally being more efficient.

Environmental degradation and depletion of natural resources necessitates a shift towards more sustainable energy sources for large scale and continuous industrial processes such as glass manufacture. Solar energy is virtually unlimited and can be concentrated to directly provide high temperature process heat. Use of concentrated solar radiation to provide process heat for industry has so far been primarily been limited to low–medium temperature processes (Kalogirou 2003), (Mekhilef et al. 2011). A major part of the research into the use of concentrated solar radiation at very high temperatures has been motivated by the desire to generate high energy density thermochemical energy storage systems, see e.g. Diver et al. (1992), Kodama (2003), Wieckert et al. (2007), Neises et al. (2012), Martinek & Weimer (2013), Tescari et al. 2013), and the solar upgrading of carbonaceous materials

(Zgraggen et al. 2007), (Rodat et al. 2010) and (Piatkowski et al. 2011). These reaction schemes usually involve solar heating of combinations of gaseous and solid reactants to generate gaseous and solid products such as the two step water and CO₂ splitting reactions involving intermediate metal oxides to generate hydrogen and/or synthesis gas (Steinfeld 2005), (Meier et al. 2012), (Villasmil et al. 2014) and (Neises et al. 2012).

Significant research work has also been devoted to the use of concentrated solar radiation for substituting fossil and electrical energy in energy intensive commodity production (Meier et al. 2012). This includes production of lime from limestone (Meier et al. 2005), melting of aluminium from aluminium scrap (Funken et al. 2001) and recovery of zinc from zinc containing materials (Schaffner et al. 2002), (Tzouganatos et al. 2014). Very few activities have been reported aiming in producing a viscous high temperature fluid as in glass melting. Felix Trombe (1954), (1961) conducted a series of experiments melting high temperature refractory materials, including use of a 1 MW solar furnace to fuse hundreds of kilograms of silica sand in a rotary furnace. However the material produced was opaque, inhomogeneous and with too high viscosity for any useful forming processes. This was probably due to the very high temperatures needed to enable silica to be fluid enough for easy bubble removal which would be a challenge for the furnace refractories.

This paper outlines initial research which aims to investigate the feasibility of using concentrated solar radiation to provide a sustainable source of process heat for glass manufacture. In particular, a high quality, transparent glass with minimal bubble content and sufficient fluidity for forming processes is required. There are several motivations to justify research into the use of solar energy to manufacture glass:

- The major environmental challenges for glass industry are energy consumption and emissions to air from combustion of fossil fuels and from high temperature oxidation of atmospheric nitrogen i.e. sulphur dioxide, carbon dioxide, and nitrogen oxides (Scalet et al. 2013). Replacing fossil fuel combustion with concentrated solar radiation to provide the process heat for glass melting, directly addresses some of these issues, although the use of carbonate raw materials means that not all CO₂ emissions can be readily prevented.
- 2. In certain developing countries, such as The Gambia, sufficient infrastructure for conventional industrial glass production facilities does not exist while solar energy and glass making grade silica sand are abundant (International Business Publications 2011). In such situations, solar powered glass manufacture could enable the sustainable use of local resources to meet local demands.
- 3. In the future, it is envisaged that solar powered glass production could enable in-situ production of the glass elements (mirrors or lenses) required for new Concentrated Solar Power (CSP) plants in remote sandy desert regions, similar to the "Sahara Solar Breeder" concept set out by Stambouli & Koinuma (2012). This could enable significant savings because currently the cost of these glass components alone account for around 6.1-8.4% of the total cost of a CSP plant (Kolb et al. 2011)(IEA-ETSAP and IRENA 2013). Also there could be additional cost savings associated with the logistics of transporting the heavy, fragile glass components, since around 12 thousand tonnes of glass is required in building a typical 100 MW CSP plant (Müller-Steinhagen 2009).
- 4. NASA (Ho & E. Sobon 1979) considered the use of concentrated solar radiation for in-situ glass production of glass from lunar based materials. Such research is motivated by the extremely high cost of transporting materials from earth, the abundance of silicate raw materials on the moon which could be used to make glass and the versatility of glass such as its ability to be drawn into fibres for structural and communications applications.

Glass melts exhibit very complex and non-linear thermodynamic/optical properties, that depend on composition, temperature and redox state (Prokhorenko 2005), which vary significantly in time and space in a furnace. A

substantial body of literature exists relating to these properties with respect to heating in conventional fossil fuel fired furnaces (Pye et al. 2005). However, extrapolating this available data to predict results for the case of heating with solar radiation is difficult because:

 The most commonly assumed regime in mathematical modelling of heat transfer phenomenon in glass melting furnaces is the 'optically thick limit' (Chaudhary 1985), (Chaudhary & Potter 2005). This basically assumes that the radiation from the heat source is rapidly absorbed/attenuated by the glass melt. Comparison of the spectral power distribution of solar radiation (approximated by a 5505°C black body) to the absorption spectrum of molten glass (Figure 1) suggests that the glass is mostly transparent to the solar radiation.



Figure 1: The spectral power distribution of solar radiation and the absorption spectrum of molten glass at 1400°C (after Chaudhary & Potter 2005).

2) A conventional glass furnace uses large flames which provide heat over a substantial area the melt by radiation and convection over large areas. However, concentrated solar radiation generally provides more focussed heating over a much smaller area. This raises issues regarding the feasibility of producing substantial flows of homogeneous molten glass as would be required for any useful manufacturing processes.

This paper describes some proof-of-concept experiments designed to obtain some initial insight into the practicalities of using concentrated solar radiation to melt glass on a reasonable scale.

2. Methods

2.1 Materials

Standard glass-making grade raw materials (purities: SiO₂ 99.5%, Na₂CO₃ 99.1%, CaCO₃ 99.3%) obtained from Glassworks Services Limited, were used to prepare a ternary simple soda-lime-silica (SLS) batch in powder form to produce a 15Na₂O 15CaO 70SiO₂ (mol%) glass. Pure silica batches were also prepared. Industrial SLS pellets to produce a commercial SLS glass were obtained from Apollo Furnaces Ltd. The average pellet mass was 673 mg. Glass melting was conducted using either mullite or alumina crucibles. The former were made in-house by slip casting and firing at 1000°C. Alumina crucibles were sourced from Almath Crucibles Ltd.

2.2 High Flux Solar Simulator (HFSS)

The PSI High Flux Solar Simulator (HFSS) (Petrasch et al. 2007) (Figure 2) was used to investigate the use of concentrated radiation for glass melting. The HFSS consists of an array of ten, independently controlled, xenon arc bulbs (Ushio UXW 15000 W, 15 kW_{el}). Each bulb was coupled with an ellipsoidal reflector aiming towards a common focal point. Shutters positioned in the path of the beam enable gradual adjustment of the power supplied to the sample.



Figure 2: Photograph of the High Flux Solar Simulator (HFSS): a) Xenon arc bulb, b) ellipsoidal reflector, c) shutters.

The spectral power distribution of solar radiation generally approximates that of a black body at 5505°C which is similar to that of the xenon arc bulbs, but with some significant divergence, mostly in the infra-red region due to the Xe emission lines, as discussed by Petrasch et al. (2007) and Alxneit & Schmit (2012). Also, there is some slight variation of the spectrum of the concentrated radiation from the HFSS over the area of the focus which would not be expected for real concentrated solar radiation (Alxneit & Schmit 2012). The discrepancy between the spectral power distributions of the solar simulator compared to real solar radiation was considered a reasonable compromise, in order to allow for easily controlled and repeatable experiments as required for the proof of concept experiments.

2.3 Static melting experiments

Powder SLS and pure silica batches were contained inside mullite and alumina crucibles respectively. The crucibles were insulated with glass fibres and placed inside a cavity reactor with a water-cooled compound parabolic concentrator (CPC) at its aperture. The horizontal beam from the HFSS was directed vertically into the sample by a 45° water cooled mirror, as shown in Figure 3. The sample was accurately positioned at the focal point of the beam using an x-y-z table.



Figure 3: The HFSS facility setup for the static glass melting experiments with a crucible containing the glass forming batch inside the cavity reactor.

2.4 Semi-continuous melting experiments

Further experiments to investigate the feasibility of generating a semi-continuous flow of molten glass using the concentrated radiation were conducted using the same setup as described above (Figure 3) with some modification to the crucible containing the melt and the removal of the cavity reactor and CPC to allow access by a batch feeder.

2.4.1 Modified crucible

Shallow, round, mullite and alumina crucibles, ranging in size, but all around 2-3 cm deep and 4-6 cm diameter, were modified with an additional spout to allow molten material to flow out and a weir to prevent un-melted material from passing out. The modified crucibles were filled with the industrial SLS pellets, surrounded by fibreglass insulation and contained within a larger mullite crucible as shown in Figure 4. In most tests a K-type thermocouple was attached to the outer surface of the crucible and hence provided an approximate information about the temperature level in the crucible/melt.



Figure 4: Typical modified crucible with 1) spout and 2) weir, surrounded by 3) insulating wool and contained within a larger crucible. 4) Industrial SLS pellets contained inside feed compartment of crucible.

2.4.2 Batch feeder

To enable a semi-continuous glass melting process, a batch feeder (see figure 6) consisting of a dispenser and a hopper containing the pelleted batch was fabricated. A linear actuator (Gimson Robotics, stroke length 700mm) was used to move the feeder to the melting region to allow batch feeding and then away again, so as to avoid

overheating due to prolonged exposure in the path of the concentrated "solar" beam. The temperature at the tip of the batch feeder was monitored to avoid overheating.



Figure 5: Remotely controlled intermittently operated batch feeder consisting of: 1. dispenser from which a small number of pellets at a time were dispensed into the melting zone; 2. agitator which vibrated the feeder to ensure pellets progressed to the end of the dispenser; 3. hopper which contained a store of pellets to gradually replenish the number of pellets in the dispenser; 4. linear actuator that moved the dispenser tip to the melting zone for feeding and away from the melting zone after feeding.

A video camera with protective filters, positioned behind the shutters, was used to observe the surface of the melting zone via the reflected image in the 45° mirror.

In a separate experiment, a TG1000-58F thermogage was used to provide the flux distribution over an area corresponding to the surface of glass melt for the semi-continuous melting experiment. The thermogage was calibrated by the manufacturer (Vatell Corp., Christiansburg, VA, US) for ±3% accuracy with a repeatability of ± 1%. 16 flux measurements were taken at equal intervals covering a region in and around the surface of the melt.

Glass samples produced were crushed to a fine powder and analysed on a Siemens D5000 X-ray diffractometer using CuK_{α} radiation, in order to determine whether the raw materials has been converted into glass.

3. Results

3.1 Static melting experiments

Pure silica melted very slowly even when the intensity of the concentrated beam was increased up to the thermal shock limit of the crucibles used. The input radiant power was gradually increased until the onset of melting was observed by a change in colour at a localised spot corresponding to where the radiation flux was presumed to be greatest. The melting zone gradually increased in size to eventually envelope the entire surface of the batch. Upon cooling, the silica glass produced appeared a translucent milky white and full of small bubbles (see Figure 6a), appearing somewhat similar to the material produced by Trombe (1954).

A 232 g batch of SLS powder was also successfully melted. The beam intensity was gradually increased until bubbling was observed in a section of the crucible. As intensity was increased further, the bubbling became more vigorous and enveloped the entire contents of the crucible. Then, the beam intensity was controlled to maintain a temperature

around 1450°C for an hour until the bubbling gradually subsided and the surface of the melt appeared to stabilise. A clear mirror image of the solar simulator could be seen in the surface of the molten SLS glass. Upon cooling, the resultant SLS glass (see Figure 6b) was observed to be completely transparent and contained few bubbles. Repeating this experiment with the pelleted batch resulted in a glass with even fewer bubbles, as shown in Figure 6c. The improved quality of glass produced by the pelleted batch was probably due to better homogenisation of the raw materials in the pellets as well as the inclusion of fining agents to aid bubble evolution in the pellets.



Figure 6: Glass produced from a) pure silica; b) powdered SLS batch b) industrial SLS pellets.

3.2 Semi-continuous melting experiments

A series of semi-continuous glass melting experiments were completed to iteratively optimise pellet feed rate, radiant power intensity and crucible geometry. Radiant heat supply was required to be sufficient to overcome the chemical heat of reaction to produce the glass from batch while maintaining sufficiently high temperatures so that the glass would be of low enough viscosity to flow out consistently. The beam intensity had to be carefully controlled in order to prevent the crucible cracking due to thermal shock. Batch feed rate also required careful control in order to provide a sufficient weight of material to force a flow of the hot glass from the melting zone out through the cooler spout but adding to much batch would cause the temperature of the glass to drop too rapidly and therefore cause a plug glass to form downstream, thus prevent a consistent outflow. The crucible geometry required adjustments in order to ensure all of the glass contained, especially the end of the spout, stayed well inside the focal spot of the beam in order to maintain the high temperatures throughout as required for the consistent outflow of the molten glass produced. The set-up was designed to feed any output glass to a pre-heated tube furnace for annealing. The final successful experimental setup included an additional heat shield and secondary heating around the outlet, provided by an open tube furnace set at 1200°C.

Radiant power intensity was gradually increased to until all the pellets initially present in the crucible had fully melted and then new pellets were fed. While the batch feeder was in the path of the beam the shutters were partially closed to reduce the risk of overheating the batch feeder tip. Once one set of new pellets had completely melted and the glass appeared sufficiently fluid, another set of pellets were fed. The interval between consecutive feeding cycles was usually around 3 minutes. These feeding cycles were repeated until a semi-continuous flow of molten glass was clearly demonstrated as shown in Figure 7.



Figure 7: Time-lapse snapshots from video of the successful semi-continuous melt flow experiment. This shows the 4th out of 7 feeding cycles completed in this experiment with the time (in minutes and seconds) from the instant when new pellets were fed (00:00) through till the moment just before the next pellets were fed (03:15). Crucible \emptyset =45mm. The black line at the left edge of the first image is due to the temporary partial closing of the shutters (required to prevent the batch feeder from overheating) behind which the camera is situated.

The output of three of the solar simulator's bulbs with a combined electrical power consumption of 32.7kW and the shutters open at 100% (corresponding to the flux distribution shown in Figure 8) was found to be adequate to consistently melt the new pellets added in each feeding cycle and rapidly recover the melt temperature to provide a sufficiently low viscosity to allow the glass to flow out properly.



Figure 8: Incident flux (kW/m²) distribution covering area corresponding to melt surface inside the modified crucible during semi-continuous melting experiment

Figure 8 shows the flux distribution at the focal plane over the 45mm diameter circular area of the crucible when three of the bulbs were on and the shutters were opened 100%. Integrating over the flux distribution over this surface area yields a total incident radiant power supply of 1.588 kW and an average flux of 999 kW m⁻². For comparison, the heat flux intensity inside a conventional glass melting furnace is usually around 90-200 kW m⁻² (Trier 1987b). However, commercial glass making furnaces are several orders of magnitude larger and extremely well insulated so therefore would be expected to utilise heat far more efficiently. Calculating black body radiative heat loss over the exposed melting surface area at 1450°C ($E_b = A\sigma T^4$), yields an approximate value of 796 W, which is around half of the total incident radiative power (1.588 kW). Furthermore, since the layer of the semi-transparent molten glass was quite thin (around 20 mm), a significant proportion of the incident radiation could have been transmitted through the melt and reflected off the bottom of the crucible and back out without being absorbed by the glass. Finally, the reflected image of the solar simulator bulbs, clearly visible on the melt surfaces (see Figure 7 at 03:15) suggests a yet further significant proportion of the incident radiative yetlected.

Figure 9 shows the temperature measured by a thermocouple touching the outside surface of the modified crucible and the temperature measured at the tip of the feeder over the course of the semi-continuous melt. Figure 9 also shows the incident radiative power supplied over the surface of the melt. This was derived by calculating the incident radiative power, as described above, from the flux distributions for the cases of 2 bulbs and 3 bulbs with the shutters open at 100% and then adjusting these values according to the logged shutter opening e.g. we know the incident power for 3 bulbs at 100% was 1.588 kW, therefore when the shutters were 90% open the estimated incident power would be $0.9 \times 1.588 = 1.429$ kW. During this experiment, seven consecutive feeding cycles were completed, which

can be seen in Figure 9 by the seven peaks in the batch feeder tip temperature and corresponding troughs in the crucible temperature profile. The batch feeder tip temperature increased during feeding as expected, because it came into the path of the beam. With the exception of the first feed, a reduction in the radiative power supplied can be seen at the beginning of each feeding cycle, as the shutters were temporarily partially closed in order to prevent the feeder tip from overheating. It can be seen in Figure 9, that where the incident radiative power was held constant for a while, the temperature of the crucible soon began to approach a corresponding constant temperature. This is because as the temperature of the melt increased, so would the thermal losses and eventually a point would be reached where the incident radiative power input would equal the thermal losses. As the temperature profiles for the feeding cycles can be seen to be approaching a horizontal line by the end of each cycle, this suggests it was approaching this equilibrium limit. The 4th feeding cycle (see Figure 9) corresponds to the sample photographs from the video of this experiment shown in Figure 7.



Figure 9: Temperature and power supply profiles for successful semi-continuous melting experiment with the 1-7 marking the instants where pellets were fed.

Feeding cycles marked 5 and 6 in Figure 9 appear the most consistent in this experiment, in both cases 5 pellets were added (total mass approximately 3.37g) and melted to become sufficiently fluid for the next feed in around 150 seconds (± 10 seconds). This yields a melting rate of 0.0224 g/s, a specific energy consumption (based on incident radiative power of 1.579 kW as discussed above) of 70.39 kJ/g and an overall thermal efficiency (assuming a theoretical specific energy requirement of 2.7 GJ/t (Trier 1987a) of 3.8%. The small scale of these experiments and lack of any insulation covering the top surface of the melt, would lead to large energy losses which could explain this low level of thermal efficiency.

XRD analysis showed that all the static melting and semi-continuous melting experiments resulted in complete conversion of the crystalline raw materials into x-ray amorphous glasses as shown in Figure 10.



Figure 10: XRD results comparing soda-lime-silica crystalline raw material to glass produced from a static melting experiment

An issue that has not been addressed in these experiments is the discontinuous nature of solar irradiation, given that most commercial glass melting is usually conducted on a continuous basis, although more intermittent melting schedules for smaller scale production should be possible. Demonstration of a more efficient, scaled up glass melting system, utilising real concentrated solar radiation for the manufacture of useful glass products is proposed. As discussed in the previous section, due to the constraints of the current, small scale, poorly insulated design, a significant proportion of the input radiation would have been reflected out and a large proportion of the absorbed radiation would be also wasted by re-emission, resulting in the low overall thermal efficiency of 3.8%. The following recommendations are proposed for future scale up experiments:

- A cavity type solar receiver, with internally reflective walls and a small aperture for the incoming radiation, could be used, rather than leaving the top surface of the melt completely exposed, in order to significantly minimize radiative heat losses by causing most of the reflected and re-emitted radiation from the surface of the melt to be deflected off the cavity walls back into the melt. These multiple internal reflections could also enable a better distribution of heat over a larger contained volume.
- 2) Use of much higher solar concentration ratios (>2000) would maximise the ratio of the volume of molten glass inside the reactor to the size of the aperture for incident radiation, thus reducing the radiative heat losses.
- 3) Careful control of batch feed rate according to instantaneous incident solar radiative power could enable better control of process temperature.
- 4) Possibly molten glass could be accumulated and maintained at a high temperature inside the well insulated cavity receiver which could act as a thermal energy store enabling the temperature inside the cavity receiver to remain sufficiently hot overnight to prevent thermal shock of refractory walls and solidification of glass so that glass production could continue the following morning, with minimal secondary heating.

- 5) The size of the system must be scaled up not only to provide a good production rate but also to benefit from efficiency gains associated with the decrease in surface area to volume ratio as well as increasing the thermal mass of the system required for overnight heat storage and to minimize temperature fluctuations which can otherwise cause to refractory materials to fail.
- 6) Alternating electrical heating and water cooling may be required at the outlet section in order to provide better control of flow rate and retention time as required to produce a suitable quality of glass for the manufacture of useful products. For a solar based system the electrical power could in principle be generated via solar cells.
- 7) Materials with better thermal shock resistance should be used to maximise the durability of the system, to enable longer term experiments with exposure to variable heat supply by solar radiation.

4. Conclusions

A series of experiments using a high flux solar simulator to investigate the feasibility of use of concentrated solar radiation to provide process heat for bulk glass production has been successfully completed. Initial, proof of concept static melting experiments involving irradiating pure silica, a ternary simple soda-lime-silica (SLS) glass batch, and an industrial pelleted SLS glass forming batch demonstrated that the raw materials could be melted, to produce x-ray amorphous glasses. Pure silica melted slowly to produce a highly porous glass but the SLS batches rapidly produced fluid melts, resulting in glasses which appeared completely transparent and with surprisingly very few bubbles considering the short residence time in the beam. The industrial SLS pelleted batch produced the best quality glasses, so these were used for the raw material feed in subsequent semi-continuous melting experiments. These experiments involved use of a remotely controlled, intermittently operated batch feeder to enable new pellets to be fed into the melting zone while the beam was still on. Also, the crucible was modified, incorporating a spout and weir to enable fully molten glass to flow out. Secondary electrical heating and a heat shield around the crucible spout was required to keep the glass melt flowing out sufficiently hot to run out consistently.

Due to the non-linearity of productivity and efficiency on scale-up, significant further experimental work on a larger scale is required before the commercial feasibility of solar powered glass melting can be evaluated to any reasonable degree of certainty. Also, the experiments described in this paper used the HFSS to provide continuous heating to maintain required temperatures for controlled and repeatable glass melting experiments whereas real solar radiation is naturally intermittent and therefore problems related to rapid temperature fluctuations would be expected. Accordingly, a follow-up series of up experiments are scheduled, again using the HFSS beam, this time aiming to melt larger quantities of glass inside a well-insulated cavity with the aid of additional secondary heating to maintain sufficient temperatures to overcome thermal shock issues when the beam intensity is reduced or turned off in order to investigate dealing with issues related to the intermittent nature of real solar radiation.

Overall, the results of these exploratory experiments provide crucial input for the next step of development of glass production with concentrated solar radiation, mainly by providing approximate values of the relation between flux and melting rate as well as insight how to realise a continuous outflow of the formed glass. Suggestions as to how these experiments could be scaled-up have also been made.

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