Processing, Properties, and Applications of Glass and Optical Materials

Edited by

Arun K.Varshneya Helmut A. Schaeffer Kathleen A. Richardson Marlene Wightman L. David Pye







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Proceedings of the 9th International Conference on Advances in the Fusion and Processing of Glass (AFPG9) and Symposium 15—Structure, Properties and Photonic Applications of Glasses held during PACRIM-9, Cairns, Australia, July 10–14, 2011

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Preface

The proceedings at hand are a bound volume of some of the presentations made at both "The 9th International Conference on Advances in the Fusion and Processing of Glass" (AFPG9) as Part A and Symposium 15, "Structure, Properties and Photonic Applications of Glasses" as Part B.

The AFPG9 was held July 10-14, 2011, co-located in Cairns (Australia) with the 9th PACRIM and the annual meeting of the Australian Ceramic Society. The organizers of PACRIM-9, namely Dan Perera and Philip Walls, thought (wisely) that they needed to approach some of the undersigned members of the Glass & Optical Materials Division of The American Ceramic Society to organize the glass and related technical program portion of the PACRIM-9.

We thought that a clear opportunity existed to expand the glass technical program to include the 9th AFPG which hitherto had missed its target of having a meeting about once every three years.

The "AFPG" conferences have a strong history of bringing together highly skilled professionals who have been contributing scientifically, educationally or technologically in direct support of the world-wide glass industry. Beginning with the first conference in this series at Alfred University in 1988, the conferences have been organized in rotation between Alfred and the Deutsche Glastechnische Gesellschaft (DGG): Dusseldorf (1990), New Orleans (1992), Wurzburg (1995), Toronto (1997), Ulm (2000), Rochester (2003), Dresden (2006).

The AFPG meeting in Australia was therefore a departure from the tradition, yet an experiment worth trying. With China, Japan, Korea and India representing better than half the gross tonnage of glass production and Australia beginning to establish itself as an important new supplier of raw materials, it didn't take much convincing to bring the AFPG to where the action may be in the foreseeable future and co-locate with the PACRIM meeting in Australia.

New challenges were foreseen, however; in particular, how do we expect to draw attendance to a faraway part of the globe while the world economy was not doing well with unemployment rates hitting highs not seen since the Great Depression and the Australian dollar had appreciated nearly 60% over a matter of months! No challenge is fun until it is met head-on. A sound technical program and megafunds are essentially the winning combination. That is just what the AFPG9 organization aimed to do. Technical programming focus, of course, was on new ways of glass fusion and raw materials that improve the efficiency of the fusion process and hence are energy conserving, sustainable, and leave a smaller carbon footprint. In addition, new thoughts were sought for container lightweighting, increasing the usable strength of the glass product. An equally important focus was on bringing the pharmaceutical glass packaging professionals to the attention of the glass industry. Because of the low tonnage involved, glass industry has barely paid attention to the needs of the pharma industry; however, because of the potential life-saving issues, the pharmaceutical industry invests a great deal in solving its own problems without a significant interaction with the traditional glass professionals. AFPG9 sought to change that. To complete the technical program package, new research on the photonic applications of glass were also sought. Presentations on optical materials and applications were organized as "Symposium 15".

Some of us donned our fundraising caps and looked for financial support. Surprisingly, we did well. We would like to thank the following corporations for their generous financial support: Ivoclar Vivadent (Volker Rheinberger), NEG (Akihiko Sakamoto), Corning (Ivan Cornejo), Owens-Corning (Manoj Choudhary) and PPG (Mehran Arbab). Internal organizational support from the Australian Ceramic Society made it possible to support several invited speakers at least partially and underwrite the expense of these proceedings. Between AFPG9 and Symposium 15, a total of six keynote lectures, 75 oral and 13 poster presentations were scheduled. The following seven students were judged to receive awards up to \$1500, plus registration, towards their travel: Fumitake Tada (Japan), Laura Adkins (USA), Sefina Ali (USA), Zhuoqi Tang (UK), Sebastian Krolikowski (Germany), J. David Musgraves (USA), and Rolf Weigand (Germany). These students received an opportunity to present posters as well as a brief oral presentation.

As it happens, the Cairns region was hit by the category 4 cyclone Yasi in early February 2011. For a brief moment in time, it appeared that the PACRIM/AFPG conferences may need to be relocated. Fortunately, early fears were unfounded. Yet, to add insult to injury, five weeks later, Japan was hit by an earthquake and a resulting nuclear catastrophe which triggered new concerns over attendance by Japanese professionals (more than what bird flu did to PACRIM 8). Undoubtedly, the attendance by glass professionals from China and India lacked luster. However, our friends from Japan and Korea made up for this shortage. In particular, we would like to thank Akio Makishima of Japan Advanced Institute of Science & Technology and Satoru Inoue of National Institute for Materials Science (NIMS) who encouraged the participation by the conglomerate of Japanese glass industry, university and government labs to bring their work on in-flight melting of glass to AFPG9. We would also like to thank Ron Iacocca of Eli Lilly to have understood the need for greater interaction between the traditional glass professional and the pharmaceutical glass packaging professional.

Special thanks go to our longtime friend, Fabiano Nicoletti of the Stevanato Group and president of the International Commission on Glass. Dr. Nicoletti delivered the AFPG9 Premiere Lunch lecture entitled, "The Global Role of the International Commission on Glass", Tuesday July 12, 2011, and presented the student awards.

Since the metropolitan Cairns had escaped the wrath of Yasi, the travel, transportation and sightseeing were not an issue. For many of us, it was the first time visiting Australia. Through the tireless efforts of our primary hosts, namely Phil Walls (President, The Australian Ceramic Society), Dan Perera (The Australian Ceramic Society), Nick Koerbin (Materials Australia), Hussein Hamka (Materials Australia), and Yi-Bing Cheng (Monash University), the conference was a roaring success. Their hospitality seems to have left no stones unturned.

Thanks to Dr. Doreen Edwards, Dean, School of Engineering, Alfred University for making the university infrastructure available for launching the conference. Endorsements by the Glass & Optical Materials Division of The American Ceramic Society and the International Commission on Glass did much to help; they are gratefully acknowledged.

To all of you, the members of the Organizing Committee extend their sincere thanks for your help, encouragement, and steadfast support during those times of uncertainty while planning AFPG 9.

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PART A: THE 9TH INTERNATIONAL CONFERENCE ON ADVANCES IN THE FUSION AND PROCESSING OF GLASS

NEW CONCEPTS FOR ENERGY EFFICIENT & EMISSION FRIENDLY MELTING OF GLASS

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ABSTRACT

The paper starts with a short analysis of the performance of currently applied glass melting processes/furnaces concerning their energy consumption levels (benchmarking), as well as NOx emissions. The best practice situations will be presented for few glass products (e.g. container glass). Potential improvements in furnace designs to obtain more intensified melting or smaller glass furnace sizes and improved-controlled process steps (melting-in, complete raw material (e.g. sand) digestion of the melt, fining, homogenization & conditioning) will be discussed. Important aspects per process step are control of the applied temperature level, finding optimum glass melt flow regimes and residence times in each compartment of the melting tank, dedicated for a specific process step. Controlled & intensified heat transfer to the batch blanket area is decisive for the possibility to reduce glass furnace size, and consequently lowering structural energy losses. Mathematical modeling studies support the development of new furnace designs and heating methods in industrial glass furnaces with targeted high energy efficiency and moderate / low NOx emissions. New technological elements, such as application of CFD models, innovative oxygen-firing glass furnace designs, improved refractory materials, and advanced process control systems support the development of new glass melting concepts and furnace designs.

INTRODUCTION

Glass industries are continuously searching for production cost reductions. Energy costs are an increasingly growing part of the total production costs. In Europe, since 2005, emission trading of greenhouse gas allowances has been in place. The costs for CO_2 emissions will grow in future, due to a limited availability (on purpose) of CO_2 -allowances in the European Union.

In container glass melting, glass melting processes in Europe use about 45-50 % of all the (primary energy) energy [non-published LCA studies and references^{1,2}], required in the production chain of that glass, including raw material supply. Benchmarking in the container glass industry shows that regenerative or oxygen-fired glass melting tank furnaces with capacities above 300 metric tons per day show lowest specific energy consumption $(3.5 - 4.25 \text{ GJ/ton molten glass, depending on cullet% and quality demand). In most other sectors, oxygen-fired furnaces are often the most energy-efficient options, even including energy consumption of the oxygen generative fired container glass furnaces with batch preheating), average residence times of the melt in the glass furnaces are much longer than required for batch-wise melting processes as can for instance be performed in laboratory crucible tests.$

Most industrial glass furnaces suffer from very wide Residence Time Distributions (RTD) of the glass in the melting tank, due to the poorly controlled glass melt flow patterns in conventional glass tank furnaces and because of the method of heat transfer to the glass forming raw material batch. The high levels of average residence times, require relatively large melt tank volumes and furnace sizes. Average residence time (hrs.), multiplied with pull rate (m³ melt/hr) is the content of a melting tank. Although, approximately 20% energy savings are possible when comparing the average of the energy efficiency of EU glass furnaces to the best practice energy efficient furnace, see figure 1 for the container glass sector, the problem of the wide RTD and poorly controlled heat transfer and flow patterns is not solved by best practice today. Even the most efficient container glass furnace shows very wide RTD's and average residence times of 20 hours or more. Therefore new furnace designs are necessary. Such furnaces should split the melting process for the different functionalities: melting-in of batch (here most energy is required), dissolution of sand grains (strong convection and mixing is preferred, here temperatures should not be too high), removal of gases (bubbles, dissolved) from the melt (highest temperature level, shallow tank depth), secondary fining and conditioning (homogenization of composition and temperature) of the melt. Furthermore, for advanced glass furnace designs, the control of high intensity heat transfer, especially into the sections where highest energy supply levels are needed, should be improved, however without excessive evaporation from the melt of volatile glass species.

Although, modern glass production plants are equipped with Air Pollution Control systems (APC),² generally consisting of a scrubber and filter (to remove dust particulates), emissions of CO_2 and NOx are still of high concern. Primary measures are mostly preferred to reduce these emission levels of glass furnaces. NOx emission reduction methods have successfully been developed in the last 25 years. For instance in the Netherlands specific NOx emission levels dropped from 5.24 kg/ton molten glass down to 1.8 kg/ton melt from 1992 until 2009. This has been achieved by primary measures, including all oxygen-firing, only, and further reductions are expected.

Even with energy efficient regenerative glass furnaces, with high air preheat temperatures, specific NOx emission levels due to fossil fuel combustion can be kept at much lower levels (container glass furnaces < 1-1.2 kg NOx/ton molten glass around the year 2010) as was shown in the 1970-ties (> 4 kg/ton molten glass).³ A combination of modified combustion chamber design, burner port geometry adaptations and optimized dimensions, type of burner, number of burners in use, burner settings (velocity of fuel injection, direction of fuel injection jets, creation of multiple jets from burner nozzles, positions of burners in burner ports), air number control (oxygen excess), and CO monitoring is a proven package of primary measures, being successful to reduce NOx emissions, often without the direct need for a DeNOx system. Emission levels in regenerative soda-lime-silica glass furnaces below 700-850 mg/Nm³ are feasible for modern glass furnaces with high energy efficiency.^{2,3}

ENERGY BENCHMARKING

Figure 1 shows the ranking of the energy efficiency of 168 container glass furnaces, starting from the lowest specific energy consumption (most energy efficient) up to the highest specific energy consumption. The energy values are normalized for the situation of 50% recycling cullet in the batch and re-calculating to total primary energy consumption per metric ton molten glass, taking into account the primary energy required for generating electric power (in case of electric boosting) and oxygen production.¹ From this figure 1, assuming it being a representative sample for worldwide container glass production, we can conclude that the average glass furnace consumes about 21% more energy than the best practice (most energy efficient) furnaces (average value for number 1-17) in the ranking of figure 1. However, glass quality demands may be different for the different furnaces, representing figure 1.

The theoretical energy demand, for heating the raw material batch and the endothermic fusion reactions for normal batch and bringing the fresh glass melt to temperatures of about 1350°C (throat temperature) is about 2.55-2.60 MJ/kg for a soda-lime-silica (SLS) glass melting process. For only cullet melting this would be 1.65 MJ/kg glass melt from this cullet. These are the lower limits, but even very well insulated glass furnaces will show some structural heat losses and fossil fuel firing is always associated with flue gas production. The flue gas temperatures of at least 200-250°C will remain. Therefore the minimum achievable energy consumption level is at least 1.5 times the value determined by the conversion of raw materials into a hot glass melt and emitted batch gases.



Average value of total set compared to average of 10 % most energy efficient: shows a potential of 21% energy savings on average

Figure 1. Example of ranking of energy efficiency from most efficient to lowest efficient container glass furnace of a set of 168 furnaces (excluding forehearths). Each data point refers to one existing container glass furnace. Specific energy data are normalized to 50% cullet and primary energy consumption electricity & oxygen production taken into account.

For normal batch this would be about 3.8 MJ/kg molten glass and for 100% cullet, about 2.5 MJ/ton molten glass. Today the most efficient container glass furnaces show energy losses / energy distributions similar to figure 2, in the case of charging room temperature (non-preheated) batch into the furnace. In all cases today, average residence times (typically 20-60 hours) of the glass in industrial melting tanks are much more than minimum melting times required for laboratory melts (typically 3-5 hours).

End-port fired regenerative container glass furnace 3.62 GJ/ton molten glass - 83 % cullet



Figure 2. Energy consumption and distribution of energy (losses) in an energy efficient container glass furnace (top 10% in the benchmark pool).

NOx EMISSIONS

In this paper, only the effect of primary measures will be shown. In the 1980's and 1990's, many glass furnace combustion systems have been modified by changing the types of burners and adjusting the nozzle sizes to reduce fuel injection velocities. Mixing between fuel (natural gas and/or fuel oil) and combustion air can be retarded by lower gas velocities and fuel jets not directing sharply into the combustion air flow.³ Furthermore, combustion control systems have been developed to limit the excess of combustion air or oxygen, preventing oxygen rich conditions in the flames, that usually lead to high NOx conversion rates (so called thermal NOx).



Figure 3. NOx emission levels in container glass industry for regenerative furnaces, depending on specific combustion space volume, curves are derived as best fit from many data.

Today, new furnace designs are considered to be important for further decreasing the NOx formation levels in fossil fuel fired glass furnaces, especially for regenerative furnace types. NOx benchmark studies, correlating furnace design parameters with specific NOx emissions and Computational Fluid Dynamic modeling of combustion processes in glass furnaces show that modifications in combustion chamber, burner port designs and number of burners are essential for a combination of high heat transfer and low NOx formation levels.

Table I summarizes some results from an inventory from approximately 40 end-port fired furnaces. This table shows clearly that an increase in combustion chamber volume, tighter control of oxygen excess and slower mixing by burner angle adjustments can result in strong reductions of NOx emissions of regeneratively fired glass furnaces. Further optimizations seem to be possible by optimizing burner port design and burner types.

Modern regenerative glass furnaces show a relatively high crown, spacy regenerators and wide/high burner ports with a small slope and a limited number of burners per port. The burner position, burner angles and nozzle size are important elements for flame shaping, for flame length, heat radiation to batch blanket & melt and NOx formation levels. Figure 3 shows results of measured specific NOx emissions in container glass sector for regenerative furnaces, depending on the relative size of the combustion chamber.

Table I. Effect of changes in air excess (oxygen residue in flue gas), burner angle (underport) and relative volume of combustion space on NOx emissions values normalized to base case (100%) with high NOx level.

End-port fired regenerative glass furnaces	"Low"-NOx glass furnaces	"Mid" NOx-group glass furnaces	"High"-NOx glass furnaces
8	·	5	Base-case
NOx emission level range	500 – 800 mg/Nm ³	~1200 -1600 mg/Nm ³	~2000 mg/Nm ³
	8% O ₂ , dry	8% O ₂ , dry	8% O ₂ , dry
Typical O ₂ content in top regenerator	64 %	80 %	100 %
Vertical burner angle compared to base case	50 %	85 %	100 %
Relative volume combustion chamber compared to volume flow fuel	125 %	110 %	100 %

ENERGY TRANSFER TO MELT AND BATCH BLANKET

One of the most important drawbacks of continuous melting tank furnaces, as used today in almost all glass sectors, is the strong recirculation ("return flow") flow of glass melt from the hot-spot zone of the melting tank along the glass melt surface, back to the batch blanket tip, (see figure 4). In the hot-spot zone of the melting tank, with high heat transfer from the combustion chamber to this zone, the glass melt receives its quality. Here bubbles are released from the melt, viscosity is very low and homogenization is effective. This high temperature melt is suitable for making high quality glass products. However, most of the well (re)fined glass melt from this section of the tank is flowing upstream to the batch blanket area instead of being transported to the conditioning zones of the melting process and after that to the forming processes. The "return flow" can be 5 to 8 times stronger than the net pull. The heat input from the combustion space directly above the batch blanket to this blanket is generally not sufficient to heat a batch effectively in order to keep the surface area of the melt covered by the blanket as small as possible. Thus, the return flow is essential to limit the surface area covered with non-molten batch. This return flow of "hot-spot" glass melt is providing most of the heat required to heat and melt (fuse) the batch materials. Without the return flow, batch blankets could extend over the whole furnace length. These very massive return flows, necessary for batch heating in conventional furnaces however lead to very wide residence time distributions for the glass in the melting tank. Some volumes of melt will not re-circulate (often this gives the minimum residence time), but some parts may re-circulate up to 10 times, giving very large residence time values. Figure 4 schematically shows this situation. Fig. 5 shows determined residence time distributions for 3 industrial furnaces. A fundamental aspect associated with this behavior is that this strong re-circulation flow determines the necessary volume of a melting tank for a certain production capacity while still keeping a certain level of minimum residence time for glass melt quality. In the case that the part of the heat supply by this recirculation flow can be replaced by another heating source for melting the batch blanket, the recirculation can be retarded and the required tank volume can be decreased.



Figure 4. Scheme of main flow patterns in glass melting tank for continuous glass production. The "return flow" from spring zone/hot spot to batch blanket can be more than 6 times the pull. Batch absorbs within about 1 hour, 80-90% of the total net heat input in the melting process.

Modeling calculations for a typical container glass furnace showed that a reduction of the wellinsulated container glass furnace volume by 30% will result in about 5% less energy consumption. For very well-insulated furnaces, which operate already at high specific pulls, potential energy savings by an even more compact melter is rather limited. But, smaller furnaces require lower capital investments. In the theoretical extreme case of infinite large regenerators and a perfectly insulated furnace the energy consumption is about 3.0 MJ/kg molten glass, this is the theoretical limit for normal batch (assuming no batch preheating). This limit however is not of practical value, since perfect insulation is not possible and infinite large regenerators are not feasible.



Minimum residence time versus average residence time = 0.15 to 0.20

Figure 5. Typical residence time distributions for the melt in different glass melting tanks for float glass, TV panel glass and soda-lime-silica container glass production.