Original Paper

Diagnosis and treatment of bubbles in glass production using a numerical simulator

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A methodology to evaluate the bubble removing performance of a tank furnace was proposed in previous papers. Since the operating data of the furnace being used for actual production can be utilized to examine the effectiveness of the above proposal, this paper deals with the verification issue when this methodology is applied to a real furnace. Additionally, various indirect indices, which have been advocated to evaluate the bubble removing performance, are compared with the proposed direct simulation. The results show that the phenomena experienced in the factory can be clearly explained and the direct simulation has been proved to be very useful to diagnose and to treat bubbles during actual production.

Diagnose und Entfernung von Blasen in der Glasproduktion mit Hilfe numerischer Simulation

Eine Methode, die Wirksamkeit der Blasenentfernung bei einem Wannenofen zu beurteilen, wurde in früheren Beiträgen vorgeschlagen. Da die Betriebsdaten des Ofens zur Verfügung stehen, behandelt die vorliegende Arbeit die Anwendung der Methode bei einem realen Ofen. Außerdem werden verschiedene indirekte Daten, die zur Beurteilung des Ergebnisses der Blasenentfernung empfohlen wurden, mit der hier vorgeschlagenen Simulation verglichen. Die Resultate zeigen, daß die in der Praxis beobachteten Phänomene klar erklärt werden können und daß sich diese numerische Simulation zur Diagnose und Entfernung von Blasen während der Produktion eignet.

1. Introduction

Research on the process to remove bubbles from the molten glass is regarded as one of the most important subjects in glass melting technology. As a means to understand and to improve this process, Kawachi and Kawase [1 and 2] proposed a numerical simulator to evaluate three factors that influence bubble removal: design of glass melting furnace, glass composition and batch materials including refining agents, and the operating conditions of the furnace. The objective of this paper is to evaluate how useful information can be obtained by applying the simulation technique to solve the bubble problem encountered in a real TV glass furnace.

Since Clomburg [3] in the 1970s, a lot of research has been conducted to clarify the behavior of glass melts in a tank furnace by numerical simulation. However, research on the consequences of thermal fluid analysis related to the glass quality from actual furnaces has seldom been published even though the necessity is recognized. The reason is presumably due to company confidentiality constraints about disclosing the real furnace data.

Nolet [4] introduced a procedure for analyzing the thermal flow of a lead glass furnace by simulation. His research revealed the existence of a dead zone, which was not serviceable for glass melting, and also a strong return current from the refiner to the melter, which reduced the energy efficiency. The effect of various furnace design factors on convection current, residence time and temperature distribution was studied. A case illustrating decreased energy consumption and full utilization of the furnace volume was reported in the paper.

In addition, Krämer [5] introduced a case of applying a 'Fining Index' proposed by TNO Institute of Applied Physics (TPD), (Eindhoven, The Netherlands) to a TV glass furnace as a method for evaluating bubble-removing performance in an indirect manner. It was assumed that the bubble-removing function was in proportion to the thermal history of the bubble. It was the product of temperature and time calculated by directing attention to a lump of the glass, which arrived at the feeder entrance in the shortest time, starting from under the batch blanket. It was also hypothesized that the bubble-removing function is inversely proportional to the viscosity of the glass with the same trajectory as described above. Such an approach was also proposed by Suzuki et al. [6] and Amemiya [7].

However, all these research methods were intended to indirectly infer the bubble-removing function. The bubble-removing mechanism was not taken into consideration.

2. Tank furnace conditions for simulation

A furnace melting TV glass was selected as the object of analysis for this study. The reason for this selection was

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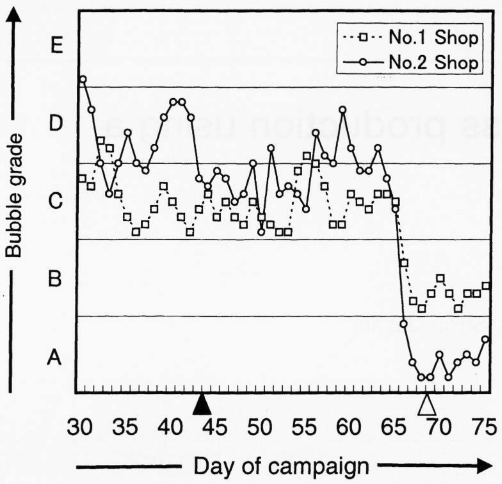


Figure 1. Transition of bubble quality (dates of case 1(▲) and case 2 (△) for simulation).

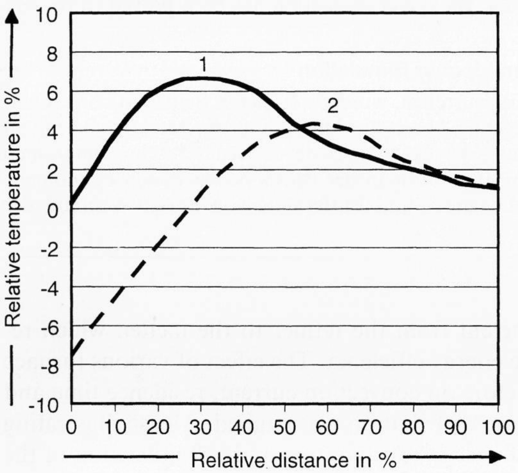


Figure 2. Surface temperature of melter; curve 1: case 1, curve 2: case 2.

due to the furnace's history. Bubble quality was dramatically improved by boldly changing furnace operating conditions after poor bubble quality had been a problem. Because the operating changes for such a short term are analyzed, there was no need to consider their influence upon simulation effects owing to changes with the passage of time in the furnace.

An oxy-fuel firing system was adopted to this furnace, a relatively small-sized furnace for TV glass with two feeders serving two shops, used to melt 50 t/d. Antimony trioxide was added as a refining agent, 0.2 wt% when converted to the oxide glass weight, and the cullet ratio was 50 %.

The transition of bubble quality from the 30th to the 75th day of the campaign is shown in figure 1. Bubble quality was classified from A through E, where A means the best quality and E means the worst. As is obvious from figure 1, the bubble quality from both feeders was C and D grade at the beginning of the period under study, but it swiftly improved to A and B grade by the 67th day. The data for the 44th day was adopted as the representative worse case in bubble quality (case 1), and the data for the 69th day was utilized as the representative better case (case 2). These two cases are marked by the symbols ▲ and △ in the date line of figure 1.

The surface temperatures of the melter were changed from case 1 to case 2 as shown in figure 2. The horizontal axis in the figure indicates the relative distance from batch charging end (0 %) to bridge wall (100 %) of the melter. Assuming the reference melting temperature is zero, the vertical axis in the figure means the temperature distribution on the superstructural walls of the melter. In case 1, the highest temperature was kept at the 30 % position of the relative distance, while in case 2 it was shifted to the 60 % position. Moreover, the highest temperature in case 2 was lowered by about 2 % in comparison to case 1. At the 0 % position of the relative distance, the temperature of case 2 was maintained 8 % lower than case 1. Because the surface temperatures of the glass melt could not be measured directly, the above temperatures were utilized as the thermal boundary condition for the thermal fluid analysis.

3. Simulation of the bubble-removing process

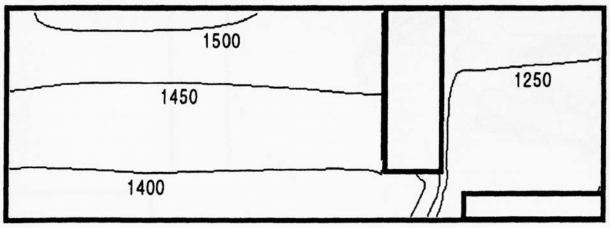
In order to evaluate the bubble-removing performance of the furnace, the following three steps were taken sequentially as described in the former papers [1 and 2][1)].

3.1 Thermal fluid analysis

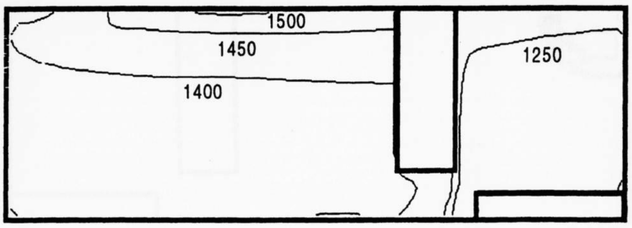
Simultaneous solutions for such equations as the Navier-Stokes' motion equation, the continuity equation and the energy equation will yield the temperature distribution and the convection current of the glass melt in the furnace. With regard to the thermal boundary conditions, the temperature condition was adopted for the glass surface, and the heat flux condition was used to determine the heat loss from the refractory wall of the glass contact area.

The computed temperature distribution and the flow vector at the furnace center section of case 1 and case 2 are illustrated in figures 3a and b and figures 4a and b, respectively. Here, the length of the depth direction is enlarged by five times. It is understood in figures 3a and b that the difference between the temperature boundary conditions on the surface reflected the temperature profile in the middle and bottom of the furnace. In figures 4a and b, the location of the hot spring was recognized as where the surface flow separated to two directions, one returning to the charging end and the other advancing to the bridge wall. The position of the hot spring is expressed by ▼ mark on the melter surface in figures 4a and b. The location of the hot spring in case 2 was moved towards the bridge wall in comparison with

[1)] In equation (14) [1], the mass transfer coefficient, k_L, should be k_{Lk} and the power of Pe_k, in the parenthesis should be $-1/3$ instead of $1/3$.



a)



b)

Figures 3a and b. Temperature distribution in degrees centigrade at the furnace center section; a) case 1, b) case 2.

case 1. Under such furnace design and the operating conditions, it was observed that there existed a forward current in the throat tunnel from the melter to the refiner, but there was no back current coming from the refiner to the melter.

3.2 Gas distribution analysis

The distribution of gas concentration dissolved in the glass melts was calculated in the next step. To compute the gas distribution, the unsteady diffusion equation concerning gas concentration was solved under the suitable boundary conditions and the source term condition.

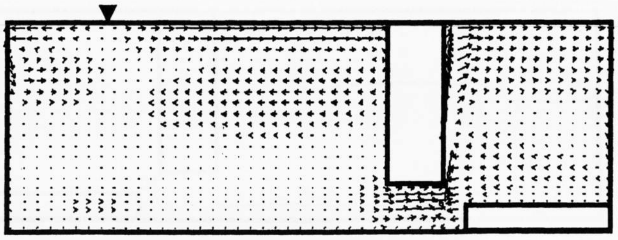
As for the source term condition, the evolved volume of the refining gas which stemmed from the decomposition of the refining agent ($Sb_2O_5 \Rightarrow Sb_2O_3 + O_2\uparrow$) was obtained in the glass melt under the temperatures and flow distribution computed in the previous section [8]. Oxygen gas distribution in the glass melt caused by the decomposition of antimony pentaoxide is presented in figures 5a and b. It is understood from these figures that the oxygen gas evolution in case 1 concentrated in around the charging end, but in case 2 it largely extended across the furnace.

Next, the saturation solubility of each gas which corresponded to the temperature under the batch blanket was regarded as the boundary condition for gas concentration at the batch/melt contact zone. In addition, the gas concentration at the glass surface calculated from the partial gas pressure in the combustion chamber using Henry's law was assumed to be the boundary condition where the glass melt had contact with the combustion atmosphere.

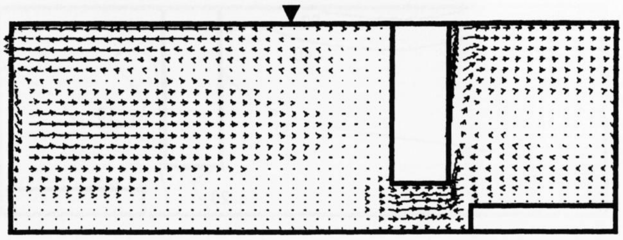
Such source term condition and boundary conditions were substituted for the unsteady diffusion equation, which gave the concentration distribution of oxygen in the glass melt as shown in figures 6a and b. The refining oxygen evolved by the refining agent was widely spread by diffusion and convection. It is obvious that the oxygen concentration in the furnace of case 2 was higher than that of case 1.

3.3 Analysis of bubble removal

Finally, the number of bubbles flowing out to the feeders was gained on the assumption that a lot of bubbles were generated under the batch blanket and were eliminated by buoyancy or absorption as they traveled through the molten glass. In the simulation, hypothesizing that the refining action in the feeders could almost be disregarded, only bubble removal in melter and refiner were considered. As described in the previous paper [2], the bubbles generated in the initial stage of TV batch melting consisted of the following gas composition (in vol.%) on the average: 15.7 CO_2, 0.3 N_2 and 84.0 O_2.



a)

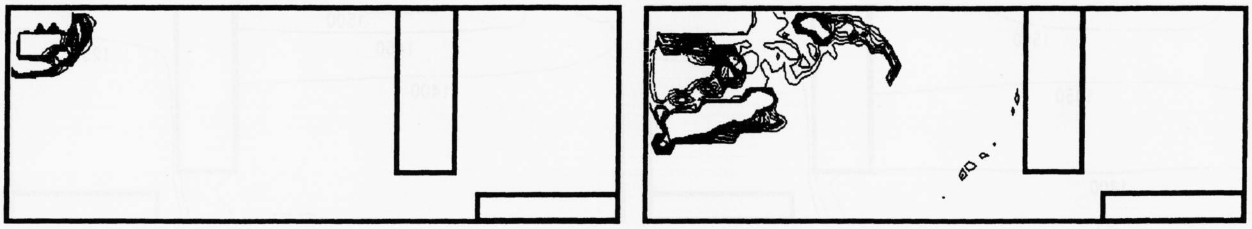


b)

Figures 4a and b. Velocity distribution at the furnace center section (▼: location of hot spring); a) case 1, b) case 2.

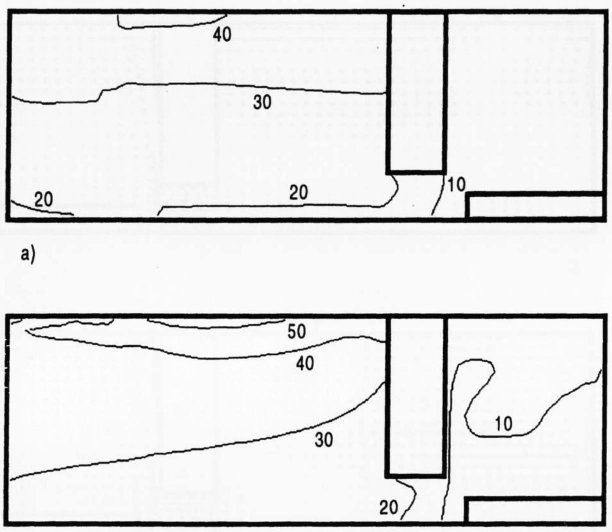
Classifying the number of bubbles by their dimension, the bubbles of 0.1 mm or less accounted for 79 %, bubbles of 0.1 to 0.2 mm accounted for 16 %, bubbles of 0.2 to 0.3 mm accounted for 4 %, bubbles of 0.3 to 0.4 mm accounted for 1 % and bubbles of 0.4 mm or more did not exist. Assuming that these bubble sizes were represented by 0.1, 0.2, 0.3 and 0.4 mm, and 2000 bubbles of each size were evolved under the batch blanket at the beginning, the whereabouts of the bubbles was pursued. The bubbles which were not eliminated in the melter and the refiner within 48 h and arrived at the feeder entrance were traced.

The outcomes are summarized in table 1. The table shows that in case 1 approximately 90 % of bubbles were removed because of buoyancy, but in case 2 about one-third of the bubbles was removed by buoyancy and



a) b)

Figures 5a and b. Distribution of refining gas (O_2) due to decomposing reaction of refining agent (Sb_2O_5); a) case 1, b) case 2.



a) b)

Figures 6a and b. Partial pressure distribution of O_2 gas in wt% at the furnace center section; a) case 1, b) case 2.

Table 1. Fate of bubbles (frequency given in %) after 48 h

class of fate	case 1	case 2
removal by buoyancy	88.4	28.2
removal by absorption	9.5	57.8
existing in furnace	0.2	9.0
flowing out of furnace	1.9	5.0

Table 2. Number of bubbles that reached feeder entrance, depending on initial bubble size

initial bubble size in mm	case 1	case 2
0.1	11	0
0.2	19	0
0.3	15	3
0.4	18	2

about two-thirds of the bubbles were removed by absorption. It can be read from table 1 that there were more bubbles in case 2 which flowed out to the feeders and had the possibility of becoming reject products when both cases were compared.

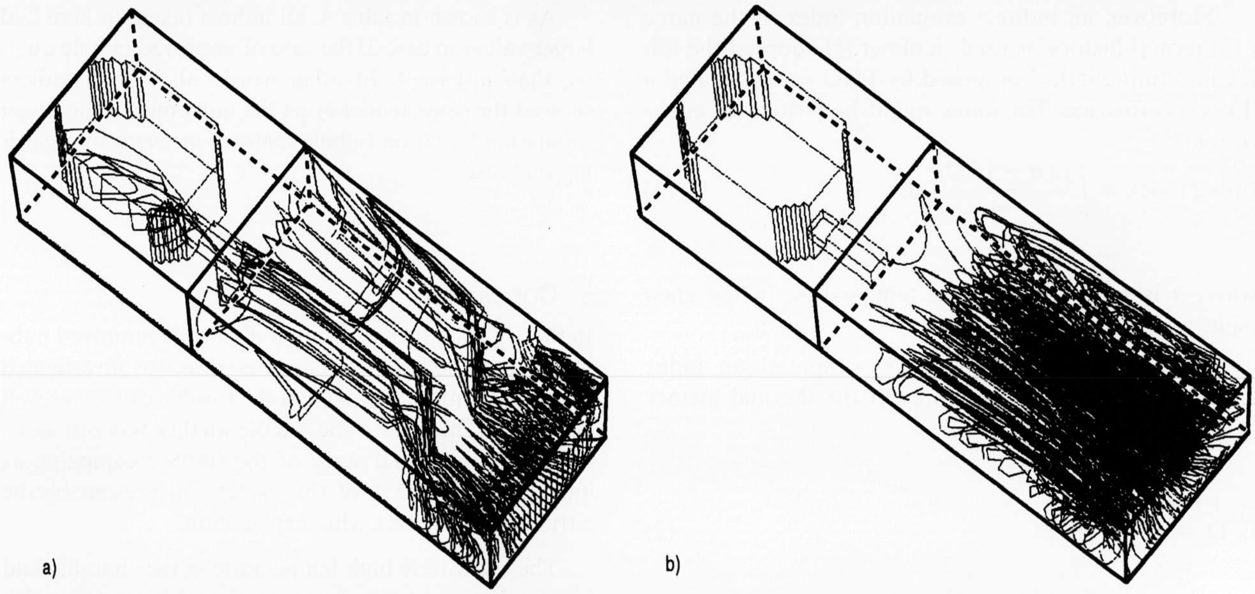
Table 3. Number of bubbles that reached feeder entrance, depending on initial bubble size and with bubble evolution rate being considered

initial bubble size in mm	existence ratio in %	case 1	case 2
0.1	79	8.69	0.00
0.2	16	3.04	0.00
0.3	4	0.60	0.12
0.4	1	0.18	0.02
sum of bubbles		12.51	0.14
ratio of sum		89.35	1.00

Since the bubbles of 0.1 mm or less are difficult to detect with the naked eye, they are rarely counted as a glass defect. Therefore, only the bubbles of 0.1 mm or more in diameter when they reached the feeder entrance became the subject for evaluation, which is tabulated in table 2. Accordingly, as an example, table 2 indicates that 2000 bubbles with 0.4 mm in initial diameter were generated under the batch blanket, and only 18 bubbles with 0.1 mm or more which reached the feeder entrance were counted in case 1.

However, because the test melting of the batch in a crucible [2] showed that the ratio of initial bubbles, depending on the diameter, was different, it was necessary to put weight on the effects by bubble size. The result is summarized in table 3. For instance, since bubbles of 0.1 mm in diameter represented 79 % of bubbles in the beginning, multiplying 11 pieces in case 1 of table 2 by 0.79 in table 3 leads to a result of 8.69. If the numbers of bubbles were calculated as stated above and were summed up, the results indicated that 12.51 bubbles in case 1 and 0.14 bubbles in case 2 flowed out of the refiner. Comparison of both cases denoted a ratio of 89.35 vs. 1.00, which means that 89 times more bubbles appeared in case 1.

With regard to the bubbles of 0.1 mm in diameter, which had the highest evolution ratio, the process of bubble removal in case 1 and case 2 is depicted in figures 7a and b. From figure 7a, a case of poor bubble quality can be observed. It is understood that many bubbles generated under the batch blanket reached the feeder entrance via melter, throat and refiner. On the other hand, in figure 7b, a case of improved bubble quality, there were no bubbles which flowed out to the feeders, because almost all were removed in the melter.



Figures 7a and b. 48-hour trajectories of bubbles of 0.1 mm in diameter when evolved; a) case 1, b) case 2.

It is known from gas chromatography analysis that the TV glass bubbles can contain four gases i.e., CO_2, N_2, O_2 and Ar. However, the gas component of the bubbles caused by improper melting or refining processes usually consists of CO_2 and N_2. If attention is paid only to CO_2, the characteristics of the bubbles can be represented. Figure 8 indicates the frequency distribution of CO_2 gas in the bubbles which reached the feeder entrance, having a diameter of 0.1 mm or more. The 'calculated value' in the figure is a summary obtained in this simulation. The 'measured value' was the result of the analysis of actual bubbles. It was judged from the figure that the calculated value and the measured value showed very good agreement. The measured values of some bubbles might have been caused by reasons which were different from factors examined in this simulation and, in particular, bubbles containing 30 to 50 % CO_2 might be attributed to the other causes besides poor melting.

4. Indirect evaluation index

Many trials have been carried out to evaluate the bubble removing performance of a tank furnace on the basis of thermal fluid analysis, but simulation based on the bubble removing mechanism has not been done, so far. In this section, the results of the direct method explained in this paper and the results of indirect method [7] are compared.

The three indices used in this study are listed in table 4. Minimum residence time is defined as the time necessary for a glass mass charged at the batch blanket area to reach the feeder entrance in the shortest possible time. It is thought as an important criterium for glass quality because, the longer the minimum residence time, the greater the chance of bubble removal increases. Accordingly, this index has been utilized as one of the guides to guessing the quality of bubbles and homogeneity by thermal fluid analysis [6 and 7]. Table 4 shows that the improved bubble quality of case 2 was the result of a 145 % longer minimum residence time than in case 1 of poor bubble quality.

Table 4. Indirect index for evaluating bubble-removing performance

index	case 1	case 2
minimum residence time in h	11.0	15.9
minimum thermal history (*[1])	$0.5153 \cdot 10^8$	$0.7757 \cdot 10^8$
minimum thermal history (*[2])	$0.3915 \cdot 10^5$	$0.4474 \cdot 10^5$

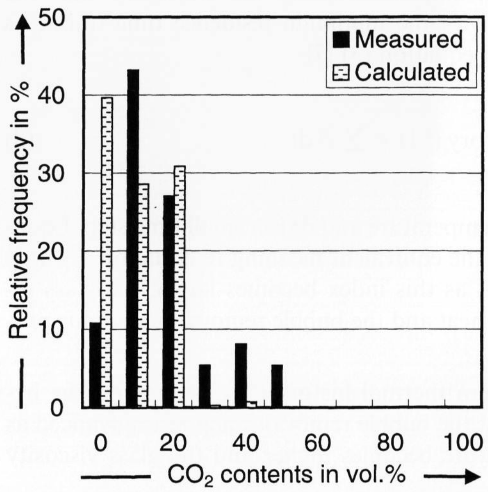


Figure 8. Frequency distribution of CO_2 in bubbles that reached feeder entrance.

Moreover, an indirect evaluation index in the name of 'thermal history' is used. Krämer [5] adopted the following 'fining index' proposed by TNO and evaluated a TV glass furnace. The index might be written as equation (1):

$$\text{fining index} = \int_0^t \frac{t\,(\vartheta - 1623)^2}{\eta}\,\mathrm{d}t \qquad (1)$$

where t is time and ϑ is the temperature in the glass melt, and η the viscosity.

In addition, Suzuki et al. [6] proposed an index called 'energy density' to represent the thermal history in the following equation (2):

$$\text{E. D.} = \int_{t_1}^{t_2} \frac{\vartheta - \vartheta_B}{t_2 - t_1}\,\mathrm{d}t \qquad (2)$$

where E.D. is the energy density, ϑ the temperature of the glass mass along the trajectory, ϑ_B the reference temperature (1300 °C adopted in their paper), and t_1 and t_2 denote time.

Although there are some differences in the above two defining equations, a common criteria is applied considering the simultaneous influence of time and temperature as the glass mass proceeds along the trajectory which governs the minimum residence time.

In this research, indices with the same concept as the above were used. "Minimum thermal history (*1)" is defined as the summation of the product, time and temperature, calculated along the trajectory that the glass mass with the minimum residence time traces, as expressed in equation (3) [7]:

$$\text{thermal history (*1)} = \sum \vartheta\,\mathrm{d}t \qquad (3)$$

where ϑ is temperature and dt is a small time step. Equation (2) has the equivalent meaning to equation (3). It is thought that as this index becomes larger, the glass receives more heat and the bubble removal function is promoted.

"Minimum thermal history (*2)" stands on the hypothesis that the bubble removal function is advanced as the temperature becomes higher and the glass viscosity decreases.

$$\text{thermal history (*2)} = \sum \frac{\vartheta}{\eta}\,\mathrm{d}t \qquad (4)$$

where η is viscosity. "Minimum thermal history (*2)" is the summed-up value of the product of time and temperature divided by viscosity concerning the glass mass with the minimum residence time.

As is shown in table 4, all indices taken up here had larger values in case 2, the case of improved bubble quality, than in case 1. In other words, all indirect indices showed the same tendency as the outcomes of the direct simulation based on bubble removal mechanism regarding this case.

5. Conclusion

In the TV glass furnace with dramatically improved bubble quality, the bubble removal process was investigated by the simulation method and the trouble cause was well explained. The reason the bubble quality was not satisfactory in the initial stage of the furnace campaign as introduced in case 1 of this paper can presumably be attributed to the following explanation.

The excessively high temperature at the charging end in case 1, caused the decomposing reaction of the refining agent to finish in a short period of time and, consequently, the concentration of oxygen in the glass melt was lowered throughout the furnace. Therefore, this prevented the bubbles generated under the batch blanket from sufficiently growing enough to escape from the glass surface or from being absorbed when they passed through the lower temperature zone.

Comparing both cases, table 1 indicates that, in case 1, there were many bubbles eliminated by buoyancy and fewer bubbles reached the feeder entrance. However, close attention must be paid to this table, noting that it shows the result of the investigation of all the bubbles generated under the batch blanket. It includes bubbles smaller than 0.1 mm in diameter, which were not recognized as bubble reject products. Accordingly, the number of bubbles larger than 0.1 mm which reached the feeder entrance is listed in table 3. It is verified that the simulator of the bubble removing process reported in the previous papers [1 and 2] functioned effectively in a tank furnace which had operating conditions appropriate for evaluation. The simulated issues were also in good agreement with the experience and observations of the plant personnel.

This analysis utilized a great deal of information obtained from the operating data of a working furnace, but the operating conditions varied from moment to moment and contributed much data noise. Additionally, the use of high temperature data of questionable reliability from the glass melt is unavoidable. Therefore, such a simulating method will be useful to evaluate the effect of big changes in the furnace design and/or the factors which have a strong influence on operating conditions.

In this case, indirect evaluation indices showed the same tendency as that of direct simulation. However, because the indirect indices are not based upon the physico-chemical theories considering bubble-removing mechanism, it is believed that direct simulation will become a powerful tool to find bubble causes and to eliminate the sources. Indirect indices are also thought to be of use when they are utilized to supplement and to interpret the results of direct simulation.

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6. References

[1] Kawachi, S.; Kawase, Y.: Evaluation of bubble removing performance in a TV glass furnace. P. 1. Mathematical formulation. Glastech. Ber. Glass Sci. Technol. **71** (1998) no. 4, p. 83–91.

[2] Kawachi, S.; Kawase, Y.: Evaluation of bubble removing performance in a TV glass furnace. P. 2. Verification using real furnace data. Glastech. Ber. Glass Sci. Technol. **71** (1998) no. 5, p. 111–119.

[3] Clomburg, L.: Mathematical and experimental modeling of the circulation patterns in glass melts. Massachusetts Institute of Technology. Cambridge, MA (USA). Ph. D. thesis 1971.

[4] Nolet, D. A.: Applications of modeling and process analysis to furnace operations and redesign. In: Proc. 46th Annual Conference on Glass Problems, Urbana, IL (USA) 1985. Columbus, OH: Am. Ceram. Soc., 1986. (Ceram. Eng. Sci. Proc. **7** (1986) no. 3–4, p. 329–339).

[5] Krämer, F. W.: Simulation of fining in speciality glass. In: Proc. 5th International Conference on Advances in the Fusion and Processing of Glass, Toronto 1997. Westerville, OH: Am. Ceram. Soc., 1998. Vol. 2, p. 7–17.

[6] Suzuki, J.; Kato, T.; Mishima, M.: Studies on refining and homogenizing progress by the two-dimensional numerical model. J. Non-Cryst. Solids **38 & 39** (1980) p. 861–866.

[7] Amemiya, H.: Problems in glass melting process and the new development. In: Proc. 25th Summer Seminar for Young Ceramists, Hachioji-city, Tokyo 1993. Tokyo: Ceram. Soc. Japan, 1993. p. 43–55.

[8] Kawachi, S.; Kato, M.; Kawase, Y.: Evaluation of reaction rate of refining agents. Glastech. Ber. Glass Sci. Technol. **72** (1999) no. 6, p. 182–187.

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