

Some Problems on Glass Melting Process

Ikutaro SAWAI

(Sawai Laboratory)

Received July 4, 1952

1. Homogenization of Molten Glass

Although we can trace back the history of glass making to many thousand years, it is the matter of the last half century that glass industry has been so completely modernized as to be numbered among the modern chemical industries.

The first remarkable development in the glass industry has been brought about by the brilliant success in the mechanization of the glass forming process. By the flow system combined with the Ribbon machine it is now possible to produce daily about 300,000 pieces of electric bulbs, the amount which is approximately equal to that scarcely accomplished by a work-man in two years.

The enormous increase in production, came about almost simultaneously in every branch of glass industry, gave a great impetus to the modernization of the continuous melting system, the "glass tank furnace" A glass tank furnace, originated by Siemens towards the end of last century, is nothing but a box made of refractories containing from some twenty to two thousand tons of molten glass, according to the variety to be produced. In general, batch is charged at one end, at the dog house, and is molten down in tank itself by gas or oil flames covering whole surface of molten glass which is transferred to automatic machines after having been thoroughly refined to homogeneous mass sufficient for the production of articles, say plate glass or container. The quality of glass, coming out from the discharging end, differs more or less according to the items to be produced. Generally speaking, a plate glass requires more thoroughly refined and homogeneous melt than a container, whereas for an electric bulb and a vacuum tube the importance is laid to the uniformity of expansion coefficient, because the perfect sealing is most important for the finished products.

Moreover, we know by long experience that glass should be stabilized by keeping at high temperature enough time after it has been apparently well refined.

By the reasons given above it is necessary to keep a large quantity of glass, for example, as much as five times of the pulling amount, always at the temperature well above 1400°C.

In fact the heat efficiency of glass tank furnace is so low that it scarcely comes up to 15 % in average. From the economical point of view the only way, perhaps to make up this deficiency would be the increase of pull rate in combination with the use of highly productive machines.

As a matter of course the articles so produced should meet the quality standard, and be as uniform as possible in quality. This problem has been solved by the elevation of the furnace temperature, which makes possible to turn out well homogenized glass through the decrease of viscosity at higher temperatures. In the table^b given below one can retrace the change in the conditions of furnace operation for the past thirty years.

Table 1.

| Year | 1919 | 1923 | 1929 | 1933 | 1935 | 1937 | 1946 | 1948 |
|---|------|------|------|------|------|------|------|------|
| Melting area in ft ² /ton. day | 16.8 | 15.7 | 12.1 | 8.6 | 8.2 | 7.2 | 5.0 | 4.8 |
| Temperature in °C | 1350 | - | 1420 | - | 1480 | 1510 | 1570 | - |

Perhaps we are coming to the stage to be able to explain our understandings of the word 'homogenization'.

Generally speaking, a commercial glass is homogeneous to an extent as it may be regarded as an assembly of the parts more or less different in quality.

At the moment when glass has just molten down, the homogeneity will be so poor, or in the other words, the variation of properties from point to point in the mass will be so large as it might be intolerable to pull up the mass at the working end. Because, the variation of the batch composition, the inevitable segregation during transportation as well as the addition of the cullet of the unknown composition make it impossible to charge the batch at the dog house which is so homogeneous as required in the finished products.

The convection currents in the tank are responsible for the homogenization of molten glass. As will be touched in some detail in the next paragraph there exist several convection currents everywhere in the tank, and a mass of newly molten glass begins to migrate along the complex maze of flowing glass at the moment when it has just got free from the knoll of the fresh batch.

By the flow of glass moving in maelstroms the mass, considered above, will be torn off into minute portions, each moving with different velocities, some of them being pulled even backwards.

The homogenization may be interpreted as the phenomena of the splitting

off and the scattering of the masses of molten glass by agitation existing in the tank.

The melting and refining steps are the heart of the glass manufacture, but they were not responsible for the major cost of production. It has been quite before, recently, the glass technicians are forced to operate the tank with such large pull rate as the products of inferior quality are very liable to be turned out.

Theoretically the analysis of the convection currents in tank might be very interesting if it would be possible to approach the problem by simple means. The complexity of the phenomena should seem to be checking to dig into the core.

The following simple arguments, however, might serve as a guide for obtaining a rough idea on the function of agitation by convection currents existing in the tank, although it will be nothing more than the scratch on the bare surface of the problem.

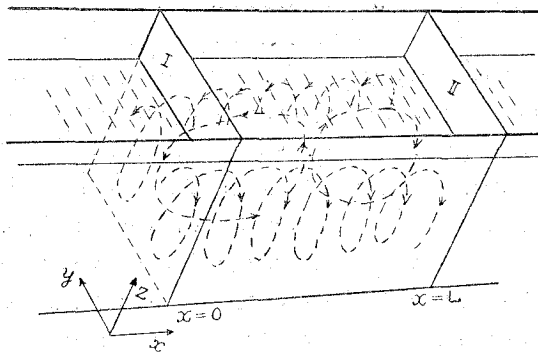


Fig. 1.

As shown schematically in Fig. 1 let us imagine a long trough containing liquid, say molten glass, extending infinitely in both direction along x axis. Two membranes I and II, permeable to the pass of liquid but impermeable to the effect of agitation, are placed at $x=0$ and $x=L$. The length L may be regarded as the length of the tank

For simplicity let us make following assumptions:

(1) While the liquid is flowing through the trough with the uniform velocity u_x from $x=-\infty$ to $x=0$, a second component is brought into this compartment with such an amount that its concentration comes up to the value c . This value c may change with time.

(2) Let us concentrate our attention to the mean value $\bar{c} = (\int_A c ds) / A$ and see how the value \bar{c} will change according to the conditions imposed at I as well as to the degree of agitation existing between two planes I and II. A is the cross-sectional area of the part of trough filled with liquid.

As the first step let us consider the simplest case, in which no agitation exists. Obviously the differential equation representing the change of \bar{c} with time t is

$$\frac{\partial \bar{c}}{\partial t} = -u_x \frac{\partial \bar{c}}{\partial x}, \quad (1)$$

whose solution obtained with the boundary conditions $x=0, \bar{c} = f(t)$ is

$$\bar{c} = f(t - x/u_x). \quad (2)$$

This states that the change of \bar{c} at I will be reproduced at II after the time x/u_x without suffering any change by passing through the trough.

Should the uniformity of velocity distribution in y - z plane be once disturbed by any cause, which may be the friction at the trough wall, the convection current or the agitation accompanied by the evolution of bubbles, the state of matters at II would no more be the reproduction of those at I. In such a case, however, the change of \bar{c} with time at I and II, although apparently quite different, are connected with some definite relationships governed by the conditions imposed between I and II.

We are now going to discuss such cases as the second step, but by passing through a short-cut leaving all mathematically rigorous proofs* almost untouched. The following limited space should rather be spent effectively in explaining the underlying principles. Our final goal is to set forth an analytical formula, which enables us to calculate the concentration \bar{c} at any time if the initial and boundary conditions are previously assigned.

For this purpose let us proceed through the following route:

(1) As the preliminary step we try to represent the actual amount, or concentration, by an expectation in order to make easy for the mathematical treatment.

(2) The second procedure is to convert the expectations for single particle to real amount or concentration by making use of the principle already established in (1).

(3) Then we pick up a particle, say n -th particle, and try to calculate the relation between two probabilities, one the probability of finding out the particle at a certain point x at the time t , and the other, that of existing at every point at a certain predetermined time $t-\tau$. If we divide the difference of these two values by τ we get the rate of change of the probability at x with respect to time.

Let a fluid system under consideration be represented by an assembly of N moving particles, whose volume V , the concentration c and the velocity u_x are different each other.

* The full mathematical treatment will be published in a separate paper.

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Let us assume that the probability of finding out n -th particle n in the space between x and $x+dx$ at the moment t will be given by the form

$$\varphi_n(t, x) dx, \tag{3}$$

where $\varphi_n(t, x)$ is the probability density for the existence of the n -th particle and varies with the conditions imposed between I and II. The higher the value of φ_n the more we are able to expect the existence, actually, of n in the space between x and $x+dx$.

If V_n, c_n be the volume and concentration respectively, of n -th particle, the amount expectable in the space $A dx$ due to the contribution by the existence of x -th particle would be

$$A c_n V_n \varphi_n(t, x) dx. \tag{4}$$

The expectable value of the total amount, which is made up with N particles, would be equal to the amount actually exists in the space, because N will generally be very large in number.

Hence the average concentration \bar{c} of y - z plane at x at the time t , will be given by

$$\bar{c}(t, x) = \sum_{n=1}^N c_n V_n \varphi_n(t, x). \tag{5}$$

If we assume that the values V_n, c_n remain unchanged during τ and also within the small distance $l = u_n \tau$, the change of the concentration with time and position could be represented by the change of the probability density alone.

Obviously the probability that the n -th particle find itself at x at the time t is equal to the product of the probability that the same particle exists at $x-l$, at t and the probability that the displacement is l at $x-l$ and at t .

Now let us designate by \bar{l} the mean value of the distances traveled during the small time interval τ by the particles which have existed on y - z plane at x , at the time t , and also by

$$g(\lambda, t, x) d\lambda. \tag{6}$$

the probability that the distance traveled by any particle has the values between $\bar{l} + \lambda$ and $\bar{l} + \lambda + d\lambda$.

Then we obtain the equation

$$\varphi_n(t+\tau, x) = \int_{-\infty}^{\infty} \varphi_n(t, x-(\bar{l}+\lambda)) g(\lambda, t, (x-\bar{l})-\lambda) d\lambda. \tag{7}$$

Assume that the value of $g(\lambda, t, x)$ would not be zero for the small values of λ , then the relation

$$g(\lambda, t, (x-\bar{l})-\lambda) = g(\lambda, t, x-\bar{l}) \tag{8}$$

would be satisfied.

Expanding the both sides of the equation (7) by Taylor's theorem we obtain

$$\begin{aligned} & \varphi_n(t, x) + \tau \frac{\partial}{\partial t} \varphi_n(t, x) + \frac{\tau^2}{2} \frac{\partial^2}{\partial t^2} \varphi_n(t, x) + \dots \\ = & \int_{-\infty}^{\infty} [\varphi_n(t, x) - (\bar{l} + \lambda) \frac{\partial}{\partial x} \varphi_n(t, x) + \frac{(\bar{l} + \lambda)^2}{2} \frac{\partial^2}{\partial x^2} \varphi_n(t, x) + \dots] g(\lambda, t, x - \bar{l}) d\lambda. \quad (9) \end{aligned}$$

Now the k -th product moment in respect of g , at t and x , will be given by

$$\int_{-\infty}^{\infty} \lambda^k g(\lambda, t, x) d\lambda = \begin{cases} A^k(t, x) & k = 2m \\ 0 & k = 2m + 1 \end{cases}, \quad (10)$$

where m is any positive integer.

$$\text{Assume that } \bar{l}^{2m} = \alpha^m A^{2m}, \quad (11)$$

in which α is a constant, then the right-hand side of the equation (9) will become

$$\begin{aligned} & \varphi_n - \bar{l} \left[\frac{\partial \varphi_n}{\partial x} - \frac{1 + \alpha/3}{2!} A^2 \frac{\partial^3 \varphi_n}{\partial x^3} - \frac{1 + 2\alpha}{4!} A^4 \frac{\partial^5 \varphi_n}{\partial x^5} + \dots \right] \\ & + \frac{A^2}{2} \left[(1 + \alpha) \frac{\partial^2 \varphi_n}{\partial x^2} + \frac{2(1 + 6\alpha + \alpha^2)}{4!} A^2 \frac{\partial^4 \varphi_n}{\partial x^4} + \dots \right]. \quad (12) \end{aligned}$$

As A^2 as well as τ in (8) and (12) might be small enough, we are able to neglect the terms higher than the second order in (8) and the similar terms in the bracket in (12), then we obtain the equation

$$\frac{\partial \varphi_n}{\partial t} = -\frac{\bar{l}}{\tau} \frac{\partial \varphi_n}{\partial x} + \frac{1 + \alpha}{2} \frac{A^2}{\tau} \frac{\partial^2 \varphi_n}{\partial x^2} \quad (13)$$

By putting $\bar{l}/\tau = \bar{u}_x$, $(1 + \alpha) A^2/2\tau = E$, multiplying to both sides with $c_n V_n$ and then summing up all terms with respect to n we obtain the expression

$$\sum_{n=1}^N c_n V_n \frac{\partial \varphi_n}{\partial t} = -\bar{u}_x \sum_{n=1}^N c_n V_n \frac{\partial \varphi_n}{\partial x} + E \sum_{n=1}^N c_n V_n \frac{\partial^2 \varphi_n}{\partial x^2}. \quad (14)$$

Referring to the equation (5) we reach to the final goal, *i. e.*

$$\frac{\partial \bar{c}}{\partial t} = -\bar{u}_x \frac{\partial \bar{c}}{\partial x} + E \frac{\partial^2 \bar{c}}{\partial x^2}. \quad (15)$$

To speak the truth the equation (15) does not contain any direct statement about the phenomena of homogenization. As long as the homogenization is understood to be the phenomena of tearing off the large mass of molten glass into minute particles and dispersing them uniformly in tank, we should have to be interested in the number N and especially in the distribution of the particles on the y - z plane and not in the change of the mean concentration $\int_A c ds/A$.

For the time being, however, this equation may serve as an alternative, because, as will be shown in next paragraph, it is able to interpret fairly well the results of experiments carried out for the purpose of investigating the homogenization in tank furnace.

Moreover, the higher is the divergency of $\lambda = l - \bar{l}$ the larger will be the distance between the particles which have previously been on the y - z plane

at x before the time t , or other words the dispersions of the material will become larger. In this case A^2 and hence E will be larger.

This term E is by no means constant, but the function of x having a profound maximum value, probably at the spring.*

For simplicity let us assume the mean value of $E(x)$ between I and II, a constant independent of x .

We are now coming to the stage to solve the equation (15) by imposing the initial and boundary conditions. We are assuming that the agitation exists only in the compartment between I and II, so that

$$\begin{aligned} &\text{at } x < 0 \quad E = 0 \\ &\text{at } 0 \leq x \leq L \quad E = E \\ &\text{and at } x > L \quad E = 0, \end{aligned}$$

where L is the distance between I and II.

The initial conditions is

$$\bar{c} = \bar{c}_0 = g(x) \text{ for } t = 0, \text{ and the boundary conditions are}$$

$$\text{at } x = 0 \quad \left. \frac{\partial \bar{c}}{\partial t} \right|_{x=0-0} = \left. \frac{\partial \bar{c}}{\partial t} \right|_{x=0+0} \quad \text{and} \quad \bar{c} \Big|_{x=0-0} = \bar{c}_1 = f(t)$$

$$\text{at } x = L \quad \left. \frac{\partial \bar{c}}{\partial t} \right|_{x=L-0} = \left. \frac{\partial \bar{c}}{\partial t} \right|_{x=L+0} \quad \bar{c} \Big|_{x=L-0} = \bar{c} \Big|_{x=L+0}$$

The concentration \bar{c}_L at $x=L$ under these conditions, assuming \bar{c}_0 and \bar{c}_1 as constant, is

$$\bar{c}_L = \bar{c}_1 - (\bar{c}_1 - \bar{c}_0) \sum_{n=1}^{\infty} \frac{2\mu_n \sin \mu_n}{\mu_n^2 + 2K + K^2} e^{K - (K + \frac{\mu_n^2}{K}) \frac{ut}{2L}} \quad (16)$$

where $K = \frac{uL}{2E}$, μ_n is the n -th root of $\tan x = \frac{2Kx}{x^2 - K^2}$.

If the agitation were so large that E would be regarded as infinity, then the equation (16) reduces to the form

$$\bar{c}_L = \bar{c}_0 - (\bar{c}_1 - \bar{c}_0) (1 - e^{-\frac{u}{L}t}), \quad (17)$$

where $u/L = a$ is the ratio of pull rate to the volume between I, II. This equation expresses the relation presented by Hampton²⁾ some twenty years ago.

If the value of E be not so large but is in the order of those obtainable with the actual tank furnace operation, the concentration at $x=L$

$$\bar{c}_L = \bar{c}_0 + \frac{\bar{c}_1 - \bar{c}_0}{2} \left\{ 1 - \phi \left(\frac{L - \bar{u}_x t}{2\sqrt{Et}} \right) + e^{\frac{ut}{2L}} \left[1 - \phi \left(\frac{L + \bar{u}_x t}{2\sqrt{Et}} \right) \right] \right\}, \quad (18)$$

solved by Wicke,³⁾ treating as semi-infinite problem, would be much more convenient, because it allows the use of probability integral table. The function

* *c f.* the next paragraph.

$\phi(x)$ is

$$\phi(x) = \frac{2}{\sqrt{\pi}} \int_0^x e^{-x^2} dx.$$

In the next paragraph the results will be compared with the experimental data.

2. Tracing the Flow of Glass in the Tank.

As the matter of fact the convection currents existing in the tank are responsible for the homogenization. The driving force of these currents, obviously, comes from the temperature differences and hence the differences of the density of molten glass at the center and near the walls of the tank. Although the phenomena seem to be so simple in principle, the actual feature of the currents is much more complicated being affected sensibly by the internal and external conditions.

The currents are so complicated, but it is very important to the practice of glass production. They are closely connected, not only with the homogenization but also with the heat transfer by convection currents and the corrosion of tank refractories. Although many glass technicians have devoted themselves to this problem during the last half century⁹⁾, and, they really have paved the way, it is also true that many important problems remain still unsolved.

In general, there are three possible ways to approach the problem, namely:

(1) During the operation some amount of indicators such as cobalt oxide or barium sulphate are added to batch. Then the whole content of tank is cooled down as it stands. The bundle of coloured lines revealed by cutting the mass of glass into pieces, are the trace marks of the flow of glass in tank. In this case the enormous amount of glass should be used for mere experimental purpose without producing any article salable. The valuable informations in anticipation, however, seem to cover such high cost in full.

(2) The second method is the use of floaters, usually a small refractory balls, whose positions during the travel in the tank are located by proper means. This method has the merit in the point being able to carry on without interrupting the operation. Plungers, or balances of special construction have often been used for similar purpose.

(3) The third one is to add some indicators such as barium carbonate⁵⁾ or ceria⁶⁾ to batch, and then analyse by chemical or physical methods the amount of indicator contained in the final products. It will be clear that this method is closely related with the items discussed in the last half of the preceding paragraph.

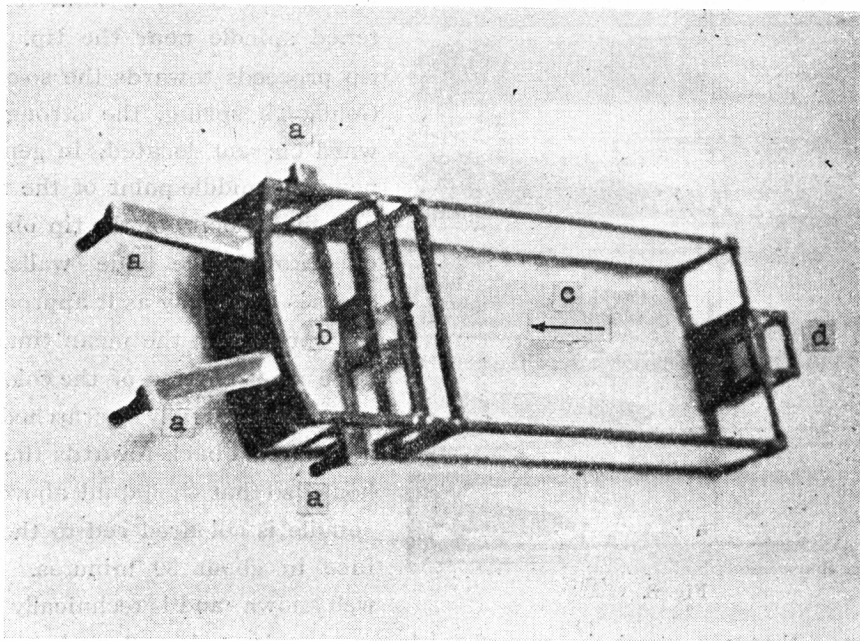
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Exact knowledge of the distribution of currents in the large mass of molten glass is difficult to secure because of the hard conditions involved in direct experiments. The observation of a floater, for example, entails long and tedious watching in the immediate neighborhood of the hot furnace wall. Moreover, the direct observation through the molten glass is impossible. Immediately, the idea must come to each one to make, in the laboratory, a model furnace, reproducing to scale the phenomena which must take place in actual furnace.

As already been pointed out by Preston⁷⁾ and Buckingham⁸⁾ a model could not represent actual tank perfectly so long as the condition of similitudes, including the similitude of external conditions, would not be satisfied. The impossibility of satisfying perfectly the condition of similitude is the serious draw back of the experiment with model, although it has a large number of merits in other points.

In fact, we are able to make direct observation of the flowing liquid through the glass walls of the model.

Now we are going to explain the results of model experiments recently carried out in our laboratory.* As the model was used a miniature tank 1/30 in size of the same which is operating in a glass factory.



a. Fore-hearth b. Throat c. Direction of pull d. Dog house

Fig. 2.

* The full text will be published in the very near future.

As it is reproduced in Fig. 2 the model was provided by a dog house, a throat and four fore-hearthes. Glycerin was used as the medium, and the conditions of the experiments were fixed so that the law of similitude could be satisfied as far as possible, using Reynolds, Prandtle and Grashof numbers. As the medium for tracing the flow of liquid in the model a part of glycerin to be charged at the dog house was mixed with a small amount of sodium hydroxide and coloured by phenolphthalein.

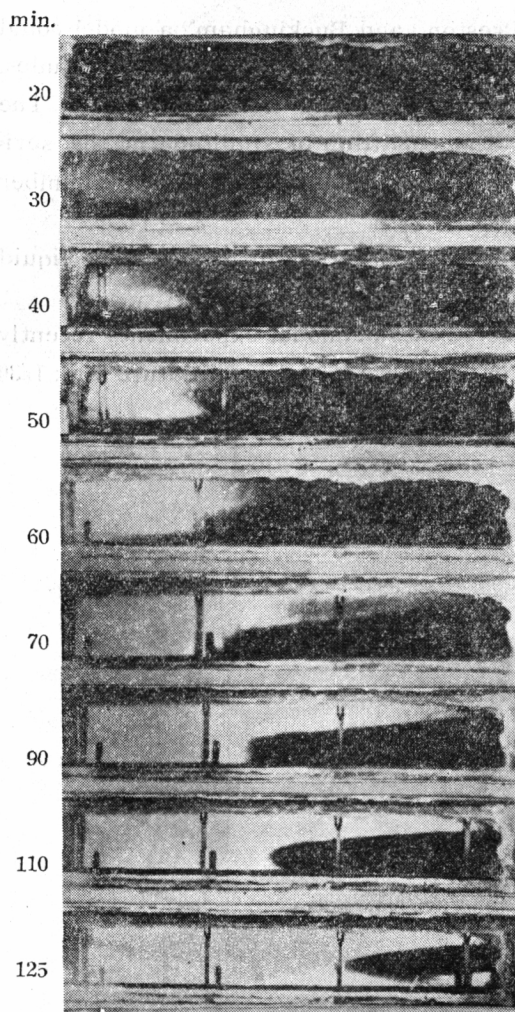


Fig. 3.

Fig. 3 is the photographic reproduction of the movement of coloured glycerin taken at intervals. The mean pull rate was 0.95 m/day. The figures attached at the left side of the photos are the time elapsed from the commencement of the experiment. With the help of the observations through the bottom of the tank the shape of the boundary, in early stages, was found to be a kind of flattened spindle near the tip. The tip proceeds towards the so-called Géhloff's spring, the strong upward current located, in general, near the middle point of the tank, and the velocity of the tip observed through the side walls, decreases gradually as it approaches the spring. In the mean time the uppermost portion of the coloured part is gradually scratched off and carried back towards the dog house so that the liquid above the spindle is coloured red to the surface in about 30 minutes. It is well known, and is technically very

important, that a part of the superficial layer of molten glass flows backward to the dog house. It is called as the superficial back current.

Due to the violent ascending current at the spring the tip is then somewhat flattened in vertical direction. This occurs during the next 10 minutes.

Meanwhile the coloured glycerin, being carried upwards by cross currents, the currents circulating in two large maelstroms in the cross sectional plane, part at the surface to right and left and then flows downwards along the both side walls. By this reason the liquid below the spindle becomes coloured after the lapse of 50 minutes. At this point of time the boundary of the tip is so flattened in the lateral direction that it looks like a straight line when observed from the bottom (Fig. 4).

About the time, when the liquid is coloured nearly uniformly from the dog house to the spring, the tip, after having gotten over the spring, begins to flow further in the direction of pull at the surface, being carried by the superficial current, which is named as superficial current of Flint, Lyle and Hausner (70 minutes). On the other hand another tip of coloured glycerin appears at the bottom. It probably comes from the influence of the direct flow bridging dog house and throat (90 minutes). In the mean time the whole space between the spring and the throat gets gradually red owing to the strong agitation by cross current. After 110 minutes from the commencement, the red tip appears in the working end after having already passed through the throat.

In Fig. 4, a, b and c are shown the sketch of the movement of such boundary observed through the side-, the back wall and the bottom, the pull rate being changed as 0.47 m, 0.95 m and 1.9 m per day.

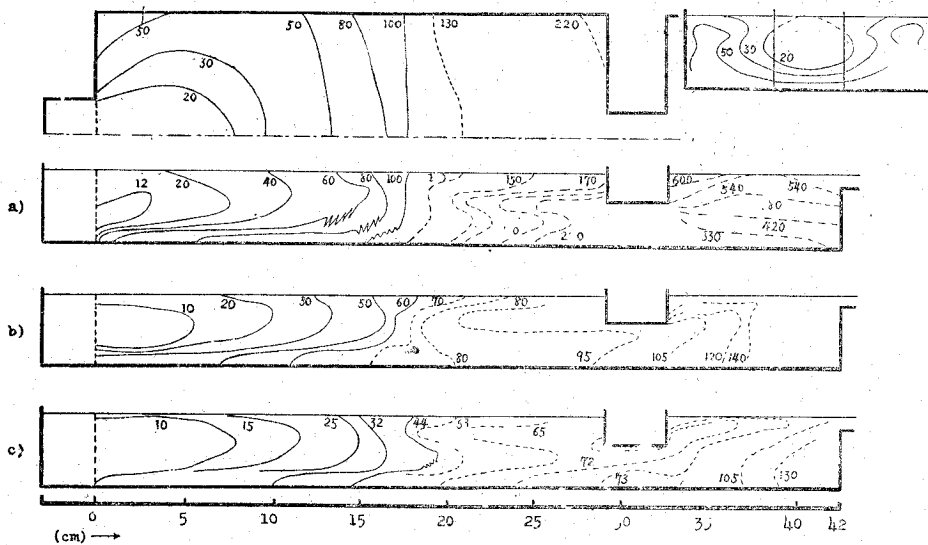


Fig. 4.

Let us compare Fig. 4 b and c with a. In both examples the increased velocity of the direct pull current reduces appreciably the time necessary

for reaching to the spring, and consequently the time during which the agitation by convection current is effective, so that the boundary of coloured glycerin remains still quite sharp. At the spring the majority of coloured glycerin is directed upwards by the spring current if the pull rate is as low as that given in Fig. 4 a. But with the increasing pull rate a larger part of coloured glycerin, being carried by the strong pull current, begins to pass rapidly through the spring, leaving smaller portion to be directed upwards at this point.

Hence the effect of the agitation by convection current, operative between the spring and throat, will be decreased with the increasing pull rate, or in the other words, the efficiency of homogenization will be lowered and, as the necessary consequence, the quality of product will become inferior.

One will be convinced that the increasing production without lowering the quality should necessarily be accomplished by more powerful agitation of molten glass. The very sensible settlement of this problem would be the elevation of the furnace temperature, which surely makes the glass less viscous.

Until recently the increasing corrosion of tank refractories at higher temperatures has checked this device to be applied in practice. Indeed, we owe much to the outstanding properties of new refractories, which have been brought to the market in the last few years.

By analysing successively the amount of alkali contained in glycerin flowing out from the fore hearthes we are able to find out the change of concentration with respect to time.

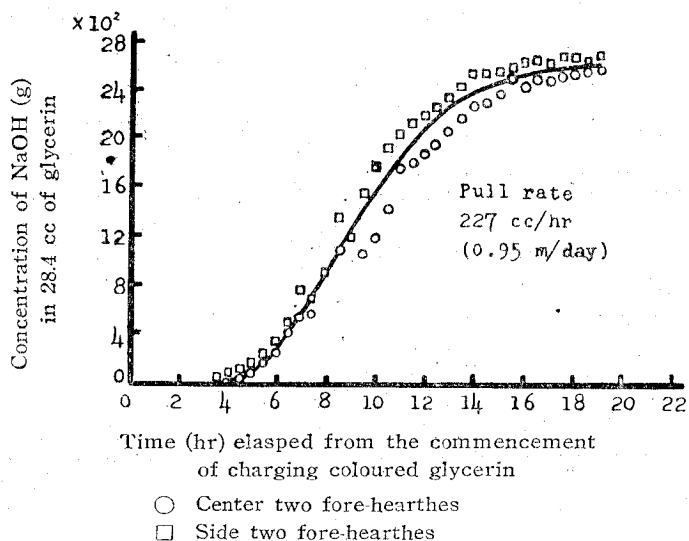


Fig. 5.

In Fig. 5 is shown a result of such experiments. The curve in full line

represents the results of the calculation with equation (18) of the preceding paragraph, using $E = 2.4 \times 10^{-2}$ m²/day and $u = 1.1$ m/day respectively. The coincidence of the experimental data with the calculated value may be regarded quite satisfactory.

Although the remaining space does not allow to discuss in detail on the correlation between the density and the glass composition, the density measurement, for example, by sink float method⁹⁾ is becoming so popular among the glass technicians for controlling the glass composition. In fact the development of simple and accurate method of measuring glass density is a prize among the recent accomplishments in the glass industry.

If the variation of the composition can accurately be known by density measurement, we are able to cite the following two examples, as shown in Fig. 6 and 7, which have been picked out from the data of tank operation.

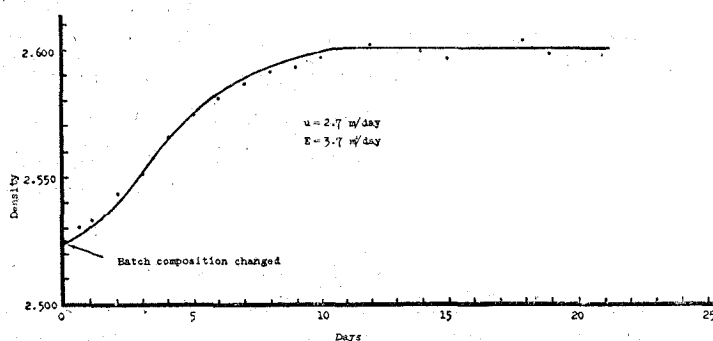


Fig. 6

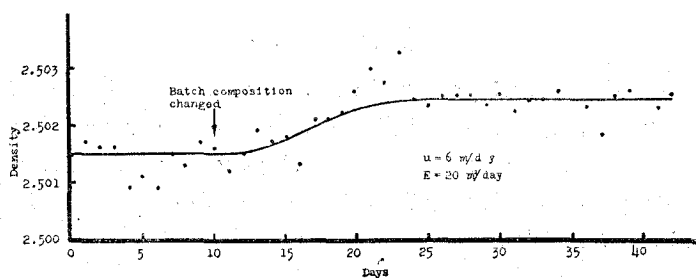


Fig. 6.

Both cases represent the variation of density of finished products after the batch composition have been slightly changed. The full lines are again the results of calculations with equation (18).

The similarity of the shape of curves with that obtained with the model experiments as well as the consistency of the measured and calculated values might serve as a proof that our present article would be of use for the practice of tank operation.

Of late a glass industry has achieved a complete modernization in factory lay out. Indeed, before ten years we little dreamed of the full automatic tank furnace of the present day. On the control panel of well equiped modern tank furnace, one will see the array of recorders and indicators, which coupled with suitable mechanism, control automatically the furnace conditions to predetermined points. All manual operations for furnace control, which have had to trust entirely to the skill of workmen, have already disappeared from the modern glass melting process. Today it is not in the least unusual thing for glass technicians that batch is fed automatically to tank furnace, whose temperature and draught are controlled with full automatic devices. The regenerators are exchanged under the control of radiation pyrometers, which measure the temperature of checker bricks. By these equipments we are now able to set the variations of glass level, furnace temperature and pressure within the very narrow allowable limits, which is becoming indispensable for the production of highest rate. The higher the rate of production and the longer the furnace life the lower will be the cost of production, although the initial investment cost might be considerably high.

The quality of the products is now controlled successfully by measuring the density, which has been found to be so valuable in giving the necessary warning on the change of glass composition.

However the sound theoretical background which bridges the conditions of furnace operation and the quality of products seems to be still very poor so that the investigations in this field is urgently needed for the future development of glass industry.

Acknowledgements

The mathematical developments in the first paragraph of this article have been worked out by Mr. Hiroshi Jinno whereas the very laborious experiments whose results appeared in the second part have been worked out by Dr. Megumi Tashiro and his collaborators to whom the author wishes to express his sincere thanks.

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