Model for batch-to-glass conversion: coupling the heat transfer with conversion kinetics

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ABSTRACT
This study describes the development of a batch-to-glass conversion model for a container-glass melting furnace. The model accounts for the relationship between the temperature history of the batch particles, batch properties, and the rate of melting by coupling the heat transfer and batch conversion kinetics models. The heat transfer within the batch is modeled by a spatially one-dimensional, convective-conductive heat balance, while the conversion kinetics is described using stretched exponential, differential Avrami, and Šesták-Berggren models based on silica dissolution data. We show that the simulated melting rate significantly changes when the conversion kinetics is considered, indicating a critical importance of the temperature history of the feed particles for the glass melting process. Finally, we summarize the limitations of the batch model and discuss the key factors to be accounted for in more advanced versions.

1. Introduction

Over the past decades, mathematical modeling has become a tool for design and operation optimization of glass-melting furnaces aiming at reducing glass manufacturing costs, controlling glass quality, and complying with the ever-more-stringent environmental constraints. Yet simplifications necessitated by the complexity of the glass melting process continue to limit the ability of current models to predict the glass production rate, and consequently the furnace productivity [1–4]. Model performance would improve if crucial aspects of the batch conversion kinetics were adequately addressed.

In glass melting furnaces, the unreacted batch is converted into molten glass within the batch body. As the material is heated, it undergoes various processes, such as the creation of molten salt phase, gas evolving reactions, silica dissolution, foam growth, and foam collapse. In fossil–fuel fired furnaces, these processes occur in the top and bottom reaction zones [5–7], shown in Figure 1, that are separated by a core zone of unreacted batch material, which gradually shrinks as the batch moves from the loading port (the doghouse) across the melt pool [8]. If the batch is charged in the form of logs or piles, the top zone is inclined and is subject to an ablation process [9,10]. If the batch is charged in the form of a layer, the horizontal top zone gradually turns to molten glass as Maehara et al. have demonstrated using computed tomography [11–13]. In electric glass melting furnaces, the batch blanket is of a virtually uniform thickness and melting proceeds solely in the bottom reaction zone. In nuclear waste glass melters to which the batch is charged in the form of aqueous slurry, the cold cap can exhibit some degree of nonuniformity.

The effect of the conversion kinetics on the batch-to-glass conversion, the major point of this study, will manifest similarly both in the top and bottom reaction zones displayed in Figure 1. However, to simplify the situation and avoid for now the mathematical treatment of the ablation process occurring at the batch surface, we focus here on the bottom reaction zone in which batch material moves vertically down while its thickness remains virtually constant after a steady state is established. A steady state bottom reaction zone persists in an electric melter, to which glass batch is uniformly charged on the cold cap surface. In a gas-heated furnace, the bottom reaction zone develops soon after the batch is loaded on the glass melt level and continues to exist until the core zone thickness shrinks to zero at the tip of the batch body. As the batch moves horizontally on the glass melt surface, the heat input from the melt below may gradually change, but the steady state is still a reasonable approximation.

The reaction zone contains four main phases – gas phase, molten salt phase, glassy phase, and solid phase consisting of one or more crystalline phases.
originating from the batch or temporarily produced by melting reactions. Most gases escape from the reaction zone through open porosity to the furnace atmosphere, but residual fraction is captured in the glass-forming melt, producing primary foam. The drainage of molten salts has been observed [14,15], but rarely occurs in batches because capillary forces keep it in place by wetting and bridging solid particles [16,17]. Thus, the condensed phases move within the batch body at the same velocity.

Although the batch conversion processes are too complex, multiple, and interdependent to be described using fundamental first principle models [18] even for simple commercial batches, their conversion kinetics can be comprehended using semi-empirical models based on experimental data. Experimental studies of glass batch melting have been conducted over a century [19–23]. Experimental studies on kinetics of conversion processes have been performed isothermally or at a constant heating rate. Data thus obtained generally suffice for developing kinetic models, which have a form of differential equations that allow solving the conversion progress for a wide range of temperature histories, including the non-linear temperature history experienced by materials in the reaction zone.

Whereas the heat transfer has been thoroughly explored for fossil-fuel fired furnaces [18,24,25] as well as electric melters [26–28], the batch melting kinetics, which is an important aspect of the glass melting process, has been insufficiently understood. In this study, we evaluate basic kinetic models for batch-to-melt conversion. The best fitting kinetic model is then coupled with a simplified model for heat transfer from the molten glass to the glass batch. The conversion kinetics and the heat transfer to the glass batch are inextricably linked through the batch bottom temperature [29–31]. Estimating the temperature of the batch–melt interface at which the batch conversion is complete and its response to furnace operating conditions is an important outcome of this study.

2. Conversion kinetics models

Recently, a kinetic analysis of glass-batch melting was reported by Ueda et al. [29] who demonstrated that the processes occurring during the batch conversion, such as batch reactions, gas evolution, silica dissolution, and foaming, depend on the heating rate in a correlated manner, thus making it possible to select one key process as a gauge of the batch conversion extent (alternatively, conversion extent can be based on reaction heat [32]). For soda-lime batches, such as a container glass batch that Ueda et al. [29] investigated, the progress in silica dissolution appears a suitable candidate because silica is present throughout the whole melting process, existing everywhere in the batch body, while the residues of silica particles continue to dissolve within the glass melt, where they assist melt fining [33,34]. Based on data by Ueda et al. [29], Figure 2 displays the fraction of dissolved silica as a function of temperature in a glass batch heated at a constant heating rate, annotated with several characteristic temperatures.

The fraction of dissolved silica particles is defined as 
\[ f = 1 - \frac{w_S}{w_{S0}} \]
where \( w_S \) is the solid (i.e. undissolved) silica mass fraction in batch and \( w_{S0} \) is the SiO₂ fraction in final glass (in which all components are dissolved). When batch samples were heated at a constant rate, \( \Phi \), the temperature, \( T \), at which the fraction of dissolved silica reached any given value, i.e. \( T(\Phi)_f \), was found to
be directly proportional to the square root of the heating rate \( \dot{Q} \) [29], i.e. \( T(\dot{Q}) \sim \dot{Q}^{-1/2} \). Characteristic temperatures for other processes, such as those indicated in Figure 2, were also linear functions of \( \dot{Q}^{1/2} \), regardless of the analytical tool (thermal analysis, x-ray diffraction, volume expansion, etc.) by which these characteristic temperatures were determined [29].

Silica particles initially react with molten carbonates, producing silicates, intermediate crystalline compounds, and carbon dioxide [35–38]. At higher temperatures, silica grains dissolve in the silicate melt. According to Ueda et al. [29], most of the carbon dioxide evolves below \( T \approx 900 \) °C, a temperature that corresponds to \( f \approx 0.5 \). At this point, the mass fraction of glass-forming melt in the batch is \( w_M = 1 - \frac{w_{SiO_2}}{1 - w_{SO_2}} \approx 0.64 \). The fraction of \( SiO_2 \) dissolved in this melt is \( w_{M,S} = \frac{w_{SiO_2}}{1 - w_{SO_2}} \approx 0.56 \).

The characteristic temperatures displayed in Figure 2 include \( T_{FO} \), the foam onset temperature, \( T_{FM} \), the foam maximum temperature, which are associated with the primary foam in the batch, and \( T_{SO_2} \), the temperature of the maximum rate of \( SO_2 \) evolution, which is associated with glass melt fining (removal of tiny bubbles by diffusion of \( SO_2 + O_2 \), so the bubbles grow and leave the melt via buoyancy). Not shown in Figure 2 is the batch bottom temperature, \( T_B \), which can only be determined using the model of the melt flow in the glass melting furnace through estimating the point where the condensed phase motion is no longer vertical with respect to the batch. The interval at which \( T_B \) can exist is limited by \( T_{FM} \), as for high-level nuclear waste vitrification [30,31,39], and \( T_{SO_2} \), because \( SO_2 \) is mostly nucleated on silica particles floating in molten glass outside the batch. In Figure 2, these limits correspond to the \( f \) values of 0.5 and 0.8 (they depend on the rate of heating). Solid silica dissolves completely (\( f = 1 \)) in molten glass at temperature \( T_D \).

As Figure 2 suggests, characteristic temperatures of the batch conversion process are associated, at least in study of Ueda et al. [29], with well-defined values of dissolved silica fractions. Accordingly, understanding the kinetics of silica dissolution is key to understanding the batch-to-melt conversion process. Whether this assumption is applicable to other glass batches and nonlinear temperature histories needs to be determined by future studies.

2.1. Models based on chemical kinetics and phase change kinetics

The dissolution of silica is affected by the shape, size distribution, and aggregation of the silica particles, the diffusion and solubility coefficients of silica in the glass-forming melt, and the concentration distribution of dissolved silica around dissolving particles. This complexity precludes describing silica dissolution with a first-principles model that would account for all the physical processes occurring during the whole conversion process. For a temperature history, \( T(t) \), where \( t \) is the time, the corresponding function \( f = f[T(t)] \) to describe the silica dissolution can be determined by an empirical model, typically as a solution of a differential equation of the form \( df/dt = F[f,T(t)] \), the numerical coefficients of which are obtained by fitting to experimental data.

Non-isothermal thermal analyses can be performed for various temperature histories, but it is conventionally done for a series of constant heating rates. The data
generally suffice for determination of an appropriate kinetic equation. In this paper, we consider two fundamental approaches, one for the kinetics of solid-state reactions and the other for the kinetics of phase change. The former is represented by the Šesták–Berggren (SB) model and the latter by the differential Avrami (DA) model. Finally, we design a stretched exponential (SE) model tailored to the relationship \( T(\Phi) \sim \Phi^{1/2} \).

2.1.1. Šesták–Berggren model

The SB model \([40,41]\) was designed for complex solid-state reactions without the precise knowledge of underlying mechanism. For dissolved silica as a reaction product, SB equation can be written as

\[
\frac{df}{dt} = A^m(1-f)^n \exp\left(\frac{-E}{RT}\right),
\]

where \(t\) is the time, \(T\) is the temperature, \(R\) is the universal gas constant, \(A\) is the pre-exponential term, \(E\) is the activation energy, and \(m\) and \(n\) are constants. Although all four parameters \(A, E, m,\) and \(n\) can be determined directly by fitting Eq. (1) to data, Kissinger method \([42]\), which provides the activation energy independently of the reaction mechanisms, is typically preferred; the remaining parameters are then determined in the second step using the nonlinear fitting method \([43]\). One disadvantage of the SB model is that it only describes irreversible dissolution. Thus, it is not applicable close to phase equilibrium.

2.1.2. Differential avrami model

The original Avrami equation \([44–46]\) was derived for the kinetics of a phase change, such as crystallization, in a system approaching equilibrium at a constant temperature. The DA model \([47,48]\) can be applied to nonisothermal processes, including phase disappearance, such as crystal dissolution \([49]\). Using the dissolved silica fraction as a measure of the phase change degree, the DA equation can be written as

\[
\frac{df}{dt} = n k_A (f_e - f) \left[-\ln\left(1 - \frac{f}{f_e}\right)\right]^{1/2},
\]

where \(k_A\) and \(f_e\) are temperature-dependent parameters \([48]\)

\[
k_A = k_{A0} \exp\left(-\frac{B_k}{T}\right),
\]

\[
f_e = 1 - \frac{w_{\text{max}}}{w_{\text{so}}} \left[1 - \exp\left(-B_w \frac{T}{T_L} - 1\right)\right].
\]

In Eqs. (2) to (4), \(k_{A0}, B_k, w_{\text{max}}, B_w, T_L,\) and \(n\) are six adjustable parameters that are determined by fitting to experimental data. Here, \(f_e\) is the dissolved silica fraction in equilibrium with silica saturated melt and \(T_L\) is the liquidus temperature. The model can mathematically describe both dissolution and growth of silica particles depending on whether \(f\) is smaller or larger than \(f_e\) at given temperature. Equilibrium is commonly reached in isothermal thermocatalytic studies.

2.2. Stretched exponential model

The SE model is based on an empirical approximation function:

\[
f = f_0 \left(1 - \exp(-\kappa^n)\right)
\]

where \(\kappa\) is a dimensionless temperature function scaled in such a way that experimental data obtained for different constant heating rates fall on the same \(f(\kappa)\) curve regardless of the heating rate, \(\Phi\), value; \(f_0\) and \(p\) are fitting parameters of which \(f_0 > 1\) allows the total silica dissolution \((f = 1)\) to occur at a final temperature.

Although the fundamental nature of the square root dependence is not yet clear and is being subject to future verification, we adopt it here for simplicity. Hence, for the \(T(\Phi) \sim \Phi^{1/2}\) dependence found by Ueda et al. \([29]\), the appropriate temperature scale is

\[
\kappa = \frac{\tau / \tau_0}{1 + \sqrt{\Phi/\Phi_t}}
\]

Substituting for \(\kappa\), Eq. (6), to Eq. (5), we obtain

\[
f = f_0 \left\{1 - \exp\left[-\frac{T}{\tau_0 \left(1 + \sqrt{\Phi/\Phi_t}\right)}\right]\right\}^p.
\]

Eq. (7) has four adjustable parameters, \(f_0, \tau_0, p,\) and \(\Phi_t\), to be determined by fitting to experimental data, compared to six of DA model.

By Eq. (6), \(\Phi = \frac{\Phi_t}{\tau_0} (\frac{T}{\tau_0} - \kappa)\). Since \(\frac{d\Phi}{dT} = \frac{1}{\tau_0} \cdot \frac{1}{\sqrt{\Phi/\Phi_t}}\), we obtain a general kinetic SE model

\[
\frac{d\kappa}{dT} = \frac{\Phi_t}{K_T} \left(\frac{T}{\tau_0} - \kappa\right)^2,
\]

in which the dissolution rate is a function of temperature and conversion degree without explicitly depending on heating rate. Eq. (7) is a solution of Eq. (8) for constant heating rate.

Unlike the SB model and similar to DA model, the SE model allows expressing the equilibrium fraction, \(f_e\), as a function of \(T\) simply by extrapolation to infinitely slow heating rate, \(\Phi \to 0:\)

\[
f_e = f_0 \left(1 - \exp\left[-\left(\frac{T}{\tau_0}\right)^p\right]\right).
\]

Such an extrapolated value is subject to high uncertainty and should be checked with the phase diagram if available.
Because the right-hand side of Eq. (8) is positive, the SE model predicts only dissolution, \( df/dt > 0 \), for any \((f, T)\) value. This reflects the irreversibility of gas-evolving reactions in an open system but does not describe the diffusion-controlled growth process in the case of a temperature decrease below the liquidus temperature. To allow silica particles to grow when \( f > f_e \), Eq. (8) can be modified as

\[
\frac{df}{dt} = \text{sgn} \left( \frac{T}{T_0} - 1 \right) \frac{f}{f_e T} \frac{q}{kT} \left( \frac{T}{T_0} - 1 \right) \frac{T}{T_0} - 1 \right)^2 . (10)
\]

Because the growth dynamics might not be captured adequately with parameters based solely on constant-heating-rate data, silica crystallization data may be necessary for fitting Eq. (10). This work is focused on conversion kinetics in the batch body, where growth of silica particles is unlikely to occur. Only after entering the circulating melt below, silica particles may experience periods of decreasing temperature before they completely dissolve [50–52].

### 2.3. Fitting kinetic models to data

To test the ability of models to represent experimental data, we used the \( f[T(t)]_\varphi \) dataset from Ueda et al. [29] who used x-ray diffraction to measure fractions of crystalline materials, including silica, in a container glass batch heated at a constant rate. To make 100 g glass of composition (in mass fractions) 0.7233 SiO\(_2\), 0.0159 Al\(_2\)O\(_3\), 0.1120 CaO, 0.0086 MgO, 0.0009 K\(_2\)O, 0.1290 Na\(_2\)O, and 0.0022 SO\(_2\), the batch contained 62.00 g sand, 13.49 g feldspar, 20.33 g limestone, 21.84 g soda ash, 0.39 g Na\(_2\)SO\(_4\), and 0.04 g carbon.

The adjustable parameters of kinetics models were obtained using the least squares method. Fitting polynomial trendlines to data allowed us to determine the temperature, \( T_m \), at which the dissolution rate reached maximum and then produce the Kissinger plot [42] shown in Figure 3. The data points lie on a parabola-shaped line, which indicates that glass batch conversion kinetics does not show simple Arrhenius dependence on temperature. The slope of the curve varies from \( E = 128 \) kJmol\(^{-1}\) at \( \Phi = 40 \) Kmin\(^{-1}\) to \( E = 401 \) kJmol\(^{-1}\) at \( \Phi = 2 \) Kmin\(^{-1}\). By the linear trendline, \( E = 238 \) kJmol\(^{-1}\), a value close to that obtained by direct fitting. Simultaneously fitting all four parameters of the SB model we obtained: \( A = 335.7 \times 10^3 \) s\(^{-1}\), \( E = 249.5 \) kJmol\(^{-1}\), \( m = -6.996 \), and \( n = 1.528 \), with \( R^2 = 0.939 \) (Figure 4a). Alternatively, fitting Eq. (1) with a temperature-dependent activation energy, \( E = E_0 + E_1 T \), to dissolution rate data yields \( A = 1.540 \times 10^3 \) s\(^{-1}\), \( E_0 = 382.9 \) kJmol\(^{-1}\), \( E_1 = -5.999 \times 10^4 \) kJmol\(^{-1}\)K, \( m = -7.377 \), \( n = 2.124 \), and a higher \( R^2 = 0.988 \) (Figure 4b). Because Eq. (1) does not admit equilibrium, at which \( df/dt = 0 \), for \( 0 < f < 1 \), silica particles would always dissolve regardless of the \( T(t) \) scenario and would never completely dissolve at a constant heating rate \( (f \to 1 \text{ as } T \to \infty) \). This results in poor fitting for the slow heating rate of 2 K/min.

When fitted to the silica dissolution data, the DA model parameter values are as follows: \( k_{AD} = 10315 \) s\(^{-1}\), \( B_A = 18443 \) K, \( w_{max} = 0.3061 \), \( B_L = 2213 \) K, \( T_L = 1586 \) K, and \( n = 0.2183 \), with \( R^2 = 0.892 \) (Figure 4c).

![Figure 3. Kissinger plot. Points correspond to the temperature at maximum dissolution rate for the 6 different heating rates (2 to 40 K/min). The solid green line represents linear and the solid blue line quadratic fits to data. Dashed blue lines are tangents to the quadratic function at points corresponding to the smallest and the largest heating rates.](image-url)
The SE model parameter values are: \( f_0 = 1.031, T_0 = 1220 \text{ K}, p = 4.044, \) and \( \Phi_f = 25.06 \text{ K s}^{-1}, \) with \( R^2 = 0.992 \) (Figure 4d). The SE model fits the data better than SB and DA models at both low and high heating rates. The difference between equilibrium lines, which are given by Eq. (4) and Eq. (9) for DA and SE models, and displayed in Figure 4c and Figure 4d, respectively, can be resolved with experiments conducted under isothermal conditions. Figure 5 shows that SE model predicts a slower initial conversion than DA and SB models. Whereas the SB model converges to \( f = 1, \) the SE and DA models generally converge to \( f_e, \) the value of which is lower for the SE model. By Eq. (8), \( \kappa \to T / T_0 \) as the dissolution rate approaches zero.

The kinetic model based on Ueda et al. data [29] has two nice features, which allowed us to obtain a simple mathematical treatment, but may not adequately describe the general behavior of batches. The square root in Eq. (6), which lacks fundamental justification, can be replaced with \( (\Phi / \Phi_f)^q, \) where \( q \) is an adjustable parameter. The fixed value of \( 1/2 \) was chosen, instead of regression, because of the codependence of silica dissolution with the gas–evolving reactions, in particular the foam onset and collapse, which are indicative of the batch bottom. Also, if silica dissolution is controlled via multiple reactions, the simple form of Eq. (7), would be replaced with a series of terms reflecting successive reactions [53–56].

3. Conversion kinetics in the bottom batch-reaction zone

3.1. Heat transfer equation

As mentioned in Introduction, we focus on the bottom reaction zone (see Figure 1) in which batch material moves vertically down while its thickness remains virtually constant after a steady state is established. The temperature distribution, \( T(x), \) in the reaction zone and the nonlinear temperature history, \( T(t), \) that silica
particles experience while passing through it are obtained by solving the heat transfer equation coupled with the kinetic equation. For the reaction zone, the one-dimensional steady-state heat-transfer equation assumes the form

\[ j c_{p,e} \frac{dT}{dx} + \frac{d}{dx} \left( \lambda \frac{dT}{dx} \right) = 0 \]  

(11)

where \( j \) is the nonvolatile condensed phase mass flux (part of the condensed phase that eventually turns to molten glass), \( c_{p,e} \) is the effective heat capacity (the sum of the sensible, reaction, and phase transition heats), and \( \lambda \) is the heat conductivity (including thermal radiation).

Equation (11) is based on several simplifications: (i) As mentioned in Introduction, within the frame of reference that moves horizontally with the floating batch, heat and mass transfer occur solely in the vertical direction (x). (ii) The heat exchange between evolving gases and condensed phases is neglected – a reasonable assumption considering the fast flow of gas through open pores in the batch [25,28]. (iii) Neglecting the unlikely phenomenon of molten salt drainage, the condensed phases (molten salts, glass-forming melt, and solids) are treated as a single reacting phase. (iv) For simplicity, the heat source term for chemical reactions is absorbed in the convective term.

The nonvolatile condensed phase mass flux is independent of x and equals the melting rate defined as the glass production rate per unit batch-melt interface area. In electric furnaces, \( j = Q_b / \Delta H \), where \( Q_b \) is the heat flux delivered from the melt below the batch and

\[ \Delta H = \int_{T_s}^{T_B} c_{p,e} dT \]  

(12)

is the conversion enthalpy that accounts for sensible heat, reaction heat, and phase transition heat; \( T_s \) is the temperature at which the batch particles are charged into the furnace (the core zone temperature).

### 3.2. Boundary conditions

The position at which \( x = 0 \) (the reaction zone bottom) is where silica particles no longer move vertically with respect to the batch body and start being carried away in the circulating melt. Laboratory studies indicate that primary foam collapses at a certain specific content of undissolved solids [29]. In this work, we assume that \( x = 0 \) is located at \( f = f_B \), where \( f_B > f_{FM} \) (see Figure 2) is a value obtained through the laboratory data analysis. Since \( f_B < 1 \), a fraction \( 1 - f_B \) of solid silica enters the melt under the batch where it circulates with the melt flow until it is fully dissolved. The temperature, \( T_B = T(f_B) \), of the interface between the melt and the batch is thus determined by the silica dissolution kinetics. Accordingly, the boundary conditions for solving Eq. (11) are

\[ T|_{x=0} = T_B \]  

(13)

\[ -\lambda \frac{dT}{dx}|_{x=0} = Q_B \]  

(14)

### 3.3. Experimental data for a soda-lime glass batch

The material properties, \( c_{p,e} \) and \( \lambda = \alpha c_p \rho \), where \( \alpha \) is the thermal conductivity, \( c_p \) is the true heat capacity, and \( \rho \) is the density, are functions of temperature and the temperature history [30,57,58] the batch had experienced before reaching a spatial point \( x \). The experimental methods used to obtain their values include thermal analysis, evolved gas analysis, differential scanning calorimetry, batch expansion test, in-situ high-temperature neutron diffraction [36], and laser flash method [59]. Figure 6 displays the thermal diffusivity and effective heat capacity versus temperature as reported by Doi et al. [60] for a soda-lime glass. Note that the batch melting reactions end at 950°C.

### 3.4. Core zone-reaction zone interface

The reaction zone spans the temperature interval between \( T_B \) and the temperature, \( T_R \), at which conversion reactions start, i.e. where the fraction of dissolved silica is virtually zero. Even though there is no well-defined physical interface between the core zone and the reaction zone, the \( T_R \) value can be estimated using experimental data. In the container-glass batch studied by Ueda et al. [29], CO2-evolving reactions started just above 600 °C, which could be the lowest temperature at which silica particles were attacked by carbonates. Note that the line of \( f(T) \) in Figure 2 indicates that some silica reacted below 600 °C, but this is an extrapolation from the temperature interval from 900 °C to

![Figure 6. Effective heat capacity and thermal diffusivity of soda-lime-silica powder batch versus temperature by Doi et al. [60].](image-url)
1400 °C at which dissolved silica fraction was measured in Ueda et al. study [29] (Figure 4).

For convenience, we define the reaction zone thickness, \( L \), by the relationship

\[
Q_R = -\lambda \frac{dT}{dx} \bigg|_{x=L} \tag{15}
\]

where \( Q_R \) is the residual heat flux from the bottom reaction zone to the core zone. Its scaled value, \( \gamma = Q_R/Q_{Rb} \), we call the heat flux ratio. The steady state implies that \( L \) is constant except near the doghouse and batch body edges.

### 3.5. Temperature distribution and dissolved silica fraction distribution in reaction zone

Figure 7 displays the results of solving Eq. (11) coupled with the SE kinetic model, Eq. (8), for material properties listed in Sections 2.3 and 3.1 and the parameter values \( f_b = 0.5 \) and \( \gamma = 0.05 \). Eq. (11) subjected to boundary conditions, Eqs. (13) and (14), was solved numerically in an iterative process, in which \( L \) was initially guessed and then progressively refined until Eq. (15) was satisfied.

The temperature history of silica particles, \( T(t) \), in the reaction zone was obtained by integrating the heating rate function \( \Phi = \frac{\partial T}{\partial x} \), where \( v = -J/\rho \) is the nonvolatile condensed phase velocity. The melting rate was estimated as \( J = 1.69 \times 10^3 \text{ kgm}^{-2}\text{d}^{-1} \) [61] and the batch density was taken as \( \rho = 1150 \text{ kgm}^{-3} \) [29].

As Figure 7 shows, the reaction zone thickness is \( L = 54 \text{ mm} \), the temperature at the reaction zone upper boundary is \( T_R = 84.5 \text{ °C} \), and only 1 \% of silica has reacted. The temperature gradient is mild, \( \frac{dT}{dx} \bigg|_{x=L} = -4.73 \text{ K mm}^{-1} \), the heating rate is \( \Phi(L) = 4.63 \text{ K min}^{-1} \), and the rate of silica dissolution is close to zero. The batch temperature gradually increases and more and more silica is dissolved as the reacting batch particles move through the reaction zone toward the batch bottom at \( x = 0 \), whereas the heating rate reaches a maximum value \( \Phi = 22.3 \text{ K min}^{-1} \) at \( x = 12.9 \text{ mm} \). The “bumpy” heating rate is a response to exothermic reactions (see the peaks on the \( \Phi \) line in Figure 6). At the batch bottom, \( T_b = 956 \text{ °C} \) and silica is dissolving at the rate of \( \frac{df}{dx} = 0.0231 \text{ min}^{-1} \) (\( df/dx_b = -22.6 \text{ m}^{-1} \)). Only a half of the silica is reacted at \( x = 0 \), the rest is dissolving in the melt flowing under the batch.

### 3.6. Effects of internal parameters and external conditions on reaction layer and melting rate

The major internal parameters that affect the thickness and the bottom temperature of the reaction layer are those of the batch properties, such as \( c_{p,e}, \lambda \), and, those connected with the batch melting kinetics, such as \( f_b \). In this section, we explore the effects of \( f_b \) and \( \lambda \). The major external parameter is the bulk melt temperature (\( T_{MB} \)).

First, we pay attention to a minor parameter \( \gamma \), which defines the reaction zone thickness and affects the temperature of the reaction zone upper boundary, but because almost no conversion occurs at \( T < T_R \), the batch bottom temperature and the rate of melting are unaffected (Figure 8). As Figure 8 illustrates, the melting rate remains unchanged (\( J = 19.6 \text{ gmm}^{-2}s \)) as long as the value of \( \gamma \) is sufficiently low.

Figure 9a shows the effect of \( f_b \) on the reaction zone thickness and the upper temperature. The sooner the
solid silica leaves the batch into the melt (the lower the \( f_B \)), the thinner is the reaction layer. The upper reaction layer temperature becomes lower, which is a consequence of the constant value of \( \gamma \) and the decrease of \( T_B \). To understand the effect of \( f_B \) and hence \( T_B \) on the melting rate, see Figure 9b, let us consider the following equation for the heat delivered to the batch bottom from the melt:

\[
Q_B = \xi (T_{MB} - T_B)
\]  

Here \( \xi \) is the heat transfer coefficient and \( T_{MB} \) is the bulk temperature of melt circulating under the batch. By Eq. (16), provided that \( \xi \) and \( T_{MB} \) do not change, \( Q_B \) increases as the batch bottom temperature \( T_B \) decreases. Since \( Q_B = j \Delta H \), we can write

\[
j = \xi (T_{MB} - T_B)/\Delta H
\]

Accordingly, \( TB \) affects \( j \) through the \( T_{MB} - T_B \) difference. Thus, the sooner dissolving silica leaves the batch to melt (the lower \( f_B \) and, hence, the lower \( T_B \)), the higher is the rate of melting.

The \( T_{MB} \) value can be measured by a thermocouple and adjusted by furnace control. The heat transfer coefficient, which depends on the material properties of the melt and on the hydrodynamic conditions below the batch [39,62], can be determined by mathematical modeling [63] or estimated from empirical data [39]. To assess its typical value, let us consider the following values: \( j = 1.72 \times 10^3 \) kgm\(^{-2}\)d\(^{-1}\), \( T_B = 1000 \) °C, \( T_{MB} = 1450 \) °C, and \( \Delta H = 1.31 \) kJ/g. Using Eq. (17) we obtain \( \xi = \frac{j \Delta H}{T_{MB} - T_B} = 57.3 \) Wm\(^{-2}\)K\(^{-1}\).

With this value of \( \xi \) and \( T_{MB} = 1450 \) °C, Figure 10 shows the dependence of \( j \) and \( T_B \) on the conversion enthalpy and heat conductivity in terms of the multiple of the nominal heat conductivity of the batch, \( \lambda_n \), equal to the \( (T) \) function based on the \( a(T) \) curve in Figure 6. Increasing \( \lambda_n \) leads to a lower temperature gradient and thus a slower rate of heating. At a slower rate of heating, the fixed \( f_B \) is reached at a lower temperature, \( T_B \) (see Figure 9). Thus, as \( \lambda \) increases, \( Q_B \) and \( j \) increase (Figure 10).

As expected, a lower conversion enthalpy leads to a higher glass production rate because less heat is required for the conversion. Note that \( j \) is not inversely proportional to \( \Delta H \) (\( j \) versus \( \Delta H^{-1} \) is not a strictly linear function) as one would anticipate based on the relationship \( j = Q_B/\Delta H \). This is because when \( \Delta H \) increases, the melting rate decreases, leading to a slower average heating rate and thus, lower \( T_B \) (reached at a fixed value of \( f_B \) see Figure 4). With a lower \( T_B \), by Eq. (16), \( Q_B \) is higher. Hence, when \( \Delta H \) increases \( Q_B \) also increases, resulting in the slightly concave shape of

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Figure 8. Melting rate, reaction zone thickness, and temperature of top and bottom of reaction zone as functions of \( \gamma \) (\( T_B = 25 \) °C and \( f_B = 0.5 \)). The effect of batch conversion kinetics was determined using the SE model.

Figure 9. Melting rate, reaction zone thickness, and temperature of top and bottom of reaction zone as functions of \( f_B \). The effect of batch conversion kinetics was determined using the SE model.
the conversion rate, shown in Figure 10a. By Eq. (12), there is also a direct effect of \( T_B \) on \( \Delta H \) through the \( c_{p,e}(T) \) function, see Figure 6. However, this effect is minor as most of the reaction heat is produced at lower temperatures, and \( c_{p,e} \) is small above 900°C.

While the increase in \( j \) with \( \lambda \) seems intuitive, the shapes of curves in Figure 10b would be different if the conversion kinetics was not considered. In models that use \( \lambda \) as a tuning parameter (often far from its physical value) to obtain the expected value of \( Q_B \), the feedback loop between the heat transfer to the batch and the conversion kinetics is absent. Because of the conversion kinetics, \( T_B \) responds to changes in both internal and external parameters, and thus does not stay constant, affecting \( j \). By recognizing the response of \( T_B \) to the processing variables, the batch model provides a realistic boundary condition for the computational fluid dynamics models of the glass melting furnace [64]. Therefore, disregarding the batch melting kinetics and treating \( T_B \) as a constant can lead to errors. With a constant \( T_B \), the batch–melt interface is no longer defined by \( f_B \); so, a constant \( T_B \) isotherm does not represent the batch-melt interface.

Figure 11 compares the effect of the bulk melt temperature, \( T_{MB} \), on the melting rate estimated with and without the consideration of batch melting kinetics. As follows from Eq. (17), the melting rate increases as \( T_{MB} \) increases. However, the conversion kinetics weakens this effect: the melting rate increases with \( T_{MB} \) at a lower rate. Without considering the kinetics (by assuming constant temperature \( T_B = 1000°C \)), the effect of \( T_{MB} \) on the melting rate is overestimated. At an increased melting rate, the batch materials experience faster heating, resulting in shift of the kinetic processes to higher temperatures. Thus, \( T_B \) increases as \( T_{MB} \) increases. For example, an increase in \( T_{MB} \) from 1400°C to 1500°C resulted in 23% increase in the melting rate when kinetics was neglected; when kinetics was considered, \( T_B \) raised by 17°C and the melting rate increased by only 16%.

![Figure 11](image_url)
4. Conclusions

A mathematical model for batch conversion in a glass-melting furnace has been developed. The model considers simplified heat transfer in the conversion zone and dissolution of silica as a characteristic process defining the conversion kinetics and the batch–melt interface. Three kinetic equations were applied based on a set of silica dissolution experiments performed at a constant rate of heating: a stretched exponential model, a differential Avrami model, and a Šesták–Berggren model. Based on the goodness-of-fit and model properties, the stretched exponential model was found to be the most suitable for our data.

The coupled model implements the feedback loop between the heat transfer within the batch, conversion kinetics, and the heat transfer to the batch. Our study demonstrates that, by considering the temperature history of the batch, the changes in melter operating conditions, e.g., changes in the bulk melt temperature, affect the kinetic processes inside the batch body, which in turn affect the temperature of the batch–melt interface and the resulting melting rate. Neglecting the effect on the temperature of the batch–melt interface leads to errors in the estimated melting rate. As discussed, a more detailed batch conversion model is needed to employ additional aspects, especially for identifying the batch–melt interface and acquiring accurate temperature-dependent properties of batch materials.

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Nomenclature

| Symbol | Description |
|--------|-------------|
| \( a \) | thermal diffusivity \( (m^2s^{-1}) \) |
| \( A \) | pre-exponential factor \( (s^{-1}) \) |
| \( c_p \) | heat capacity \( (Jkg^{-1}K^{-1}) \) |
| \( E \) | activation energy \( (Jmol^{-1}) \) |
| \( f \) | fraction of dissolved silica |
| \( g \) | reaction mechanism function |
| \( j \) | melting rate \( (kgm^{-2}s^{-1}) \) |
| \( k \) | reaction constant \( (s^{-1}) \) |
| \( L \) | conversion zone thickness \( (m) \) |
| \( Q \) | heat flux \( (Wm^{-2}) \) |
| \( R \) | universal gas constant \( (8.314Jmol^{-1}K^{-1}) \) |
| \( t \) | time \( (s) \) |
| \( T \) | temperature \( (K) \) |
| \( w \) | mass fraction |
| \( x \) | position \( (m) \) |

Greek symbols

| Symbol | Description |
|--------|-------------|
| \( \gamma \) | heat flux ratio |
| \( \Delta H \) | enthalpy \( (Jkg^{-1}) \) |
| \( \kappa \) | modified fraction of dissolved silica |
| \( \lambda \) | heat conductivity \( (Wm^{-1}K^{-1}) \) |
| \( \zeta \) | heat transfer coefficient \( (Wm^{-2}K^{-1}) \) |
| \( \rho \) | density \( (kgm^{-3}) \) |
| \( \Phi \) | heating rate \( (Ks^{-1}) \) |

Sub- and superscripts

| Symbol | Description |
|--------|-------------|
| \( B \) | batch bottom |
| \( D \) | total dissolution |
| \( e \) | effective |
| \( FM \) | foam maximum |
| \( FO \) | foam onset |
| \( MB \) | bulk melt |
| \( R \) | core zone-react zone interface |
| \( S \) | core zone |
| \( T \) | batch top |

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