Original Paper

Optimizing the cooling parameters for annealing of glass bottles by stress simulation according to the viscoelastic theory

Cuong Phan Dang and Dieter Brüggemann

Forschungsstelle WOPAG, LTTT, Universität Bayreuth, Bayreuth (Germany)

The annealing process of glass bottles was calculated by means of the stress simulation according to Narayanaswamy, a finite element method coded in ANSYS. The glass characteristics were described according to the viscoelastic theory. The change of glass properties was computed with the help of the fictive temperature, which is described by the relaxation function of the glass structure. For a successful annealing process, the heating of the entire glass bottle to the annealing temperature is fundamental. The heating-up time depends strongly on the initial temperature and the wall thickness of the bottles. The maximum cooling stresses in the bottle are located at the bend between bottle wall and bottom. The residual stress depends particularly on the cooling rate in the critical cooling zone and for the smaller part on the cooling rate in the cooling-down zone. The maximum residual stress in the bottle was fitted as an exponential function of the cooling time.

1. Introduction

During the moulding process high stresses arise in the bottle due to the fast and uneven cooling of the glass in the metal mould. They lower the entire mechanical strength of the glass bottles. The bottles are easily fragile and not usable in this "un-annealed" state.

A well-known method to increase the mechanical strength of the bottles is the relaxation of these stresses in the bottle by a minimum value in a cooling furnace. First the bottles are warmed up to the relaxation temperature, kept there for a certain time and then carefully cooled down to room temperature. This thermal treatment is also called stress relaxation process or annealing.

Technologically, the entire annealing process can be divided into three phases:

- heating-up of the bottles to the annealing temperature and relaxation of the existing stresses;
- careful cooling (critical cooling) of the bottles to the so-called "strain point";
- cooling-down to room temperature [1].

Commonly, a long cooling furnace with a slowly running transport system inside is used for the annealing of bottles. When transported through the cooling furnace, the bottles are heated up or cooled down in different zones by circulating air to furnace temperature in the respective zone.
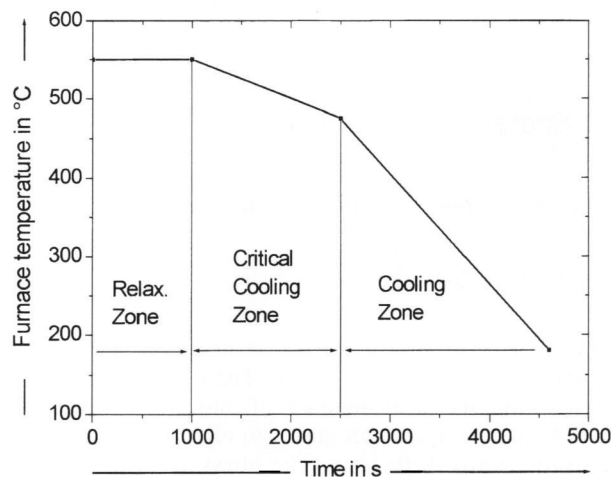
Figure 1. Typical cooling curve of a cooling furnace or target temperature of the bottles as a function of time [2].

Figure 1 shows a typical cooling curve of the cooling furnace, precisely giving the target temperature of the bottles in different zones in the furnace as a function of time [2].

During this process stresses in the bottle are diminished first in the relaxation zone, but then re-established with cooling down to room temperature. The intensity and the local distribution of the residual stresses furthermore affect the mechanical strength of the bottle. The stress calculation method of Narayanaswamy is a tool for the simulation of the evolution of thermal stresses as a function of the thermal history of the glass and for optimizing the annealing process.

2. Theory

2.1 Thermal process

The basis for the computation of the thermal stresses is the calculation of the local temperature gradients in the bottle during the whole cooling process. The heat transport in the glass and in the furnace takes place via heat conduction, convection and radiation. In this case, the heat balance equation which describes the dependence of the glass temperature T on place and time t is given as follows [3]:

$$c_p \cdot \rho \cdot \frac{\partial T}{\partial t} \equiv \nabla(\lambda : \nabla T) + \nabla q_r + \nabla q_{conv}. \tag{1}$$

Here, c_p is the specific thermal capacity of the glass, ρ the glass density, λ the heat conductivity, q_r the radiation heat flow, and q_{conv} the convective heat flow.

In most cooling furnaces hot air from a separate combustion chamber is used as the heating medium. The heat transfer is dominated in this case by the convection at the outside bottle surface. The heat transfer by radiation in the glass can be computed with the active heat conductivity λ_{activ} of the glass. λ_{activ} consists of the real conductivity and a radiation part

$$\lambda_{activ} = \lambda + \lambda_{radiation} \tag{2}$$

and can be determined by appropriate experiments [4].

2.2 Stress relaxation function

The different thermal strains in the glass cause stresses which are computed according to Hooke's law [5]

$$\sigma_t = E_t \cdot (\varepsilon - \varepsilon_{thermal}) \tag{3}$$

where σ_t is the stress, E_t is the elastic modulus, ε and $\varepsilon_{thermal}$ are the actual and thermal strain. The index t means that the stress and the elastic modulus of glass are depending on time. At high temperatures glass can relax, i.e. the stress σ_t decreases slowly with time. The stress relaxation is the faster, the higher the glass temperature is. According to the experimental results of Kurkjian [7] a typical time dependence function $M_S(t)$ of the stress [6] develops (figure 2).

According to equation (3) it is equal to the time dependence of the elastic modulus E_t. E_t can be described by the function $M_S(t)$, which is called relaxation function of the stress or elastic modulus.

$$M_S(t) = \frac{\sigma_t}{\sigma_0} = \frac{E_t}{E_0} = \exp\left[=\left(\frac{t}{\tau_T}\right)^b\right] \tag{4}$$

where σ_0 and E_0 are stress and elasticity modulus at time 0 and σ_t and E_t at time t. The constant b varies for different glasses within the range of 0.4 to 0.7 (typically 0.5) and the relaxation time τ_T depends on the temperature [6].
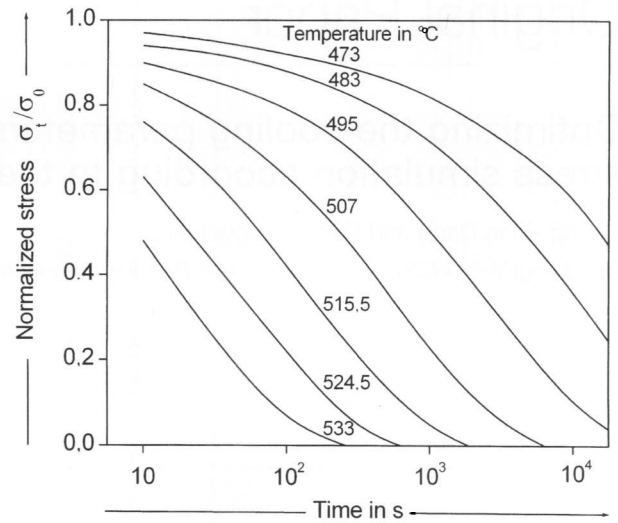

Figure 2. Stress relaxation in a soda-lime-silica glass as a function of time and temperature [7].
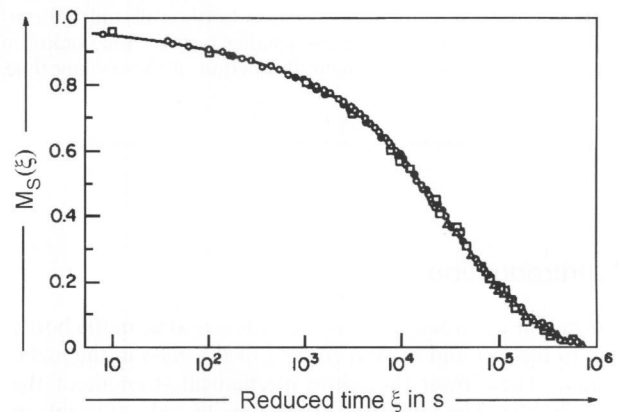

Figure 3. Master curve of the stress relaxation functions at 473 °C [7].

If we use a relaxation time τ_R at a temperature e.g. of 473 °C as a reference and compute the reduced time ξ according to equation (5)

$$\xi = \frac{\tau_R}{\tau_T} t, \tag{5}$$

all test curves can be shifted into one single curve by plotting $M_S(t)$ versus ξ (figure 3).

The function

$$\psi \equiv \frac{\tau_R}{\tau_T} \tag{6}$$

was also called shift function in this case and the obtained curve is called master curve at 473 °C. An accurate fitting of the master curve can be obtained by a sum of exponential terms

$$M_S(\xi) \approx \sum_{i=1}^{I} C_i \cdot e^{\left(-\frac{\xi}{k_i}\right)} \tag{7}$$
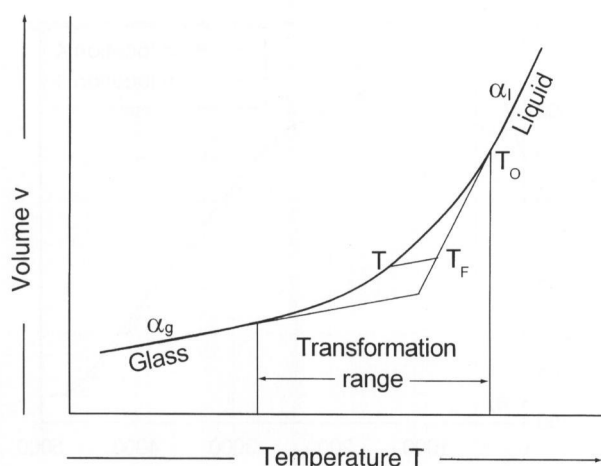
Figure 4. Evolution of the specific volume of glass cooled at a given rate.

Figure 5. Stress profile in x, y, z direction in the transient area from the bottle wall to the bottom; stress is given in MPa.

and the weighting factors C_i

$$\sum_{i=1}^{I} C_i = 1 . \tag{8}$$

I is the number of the Maxwell elements and k_i are fit parameters.

2.3 Relaxation function of the glass properties

Other glass properties in the transformation range (e.g. refractive index, viscosity, specific volume ...) show similar time dependence, if glass is warmed up or cooled down to a certain temperature within the transformation range. The reason for this viscoelastic behaviour is the dilatational structure movement in the glass with changes of temperature or under mechanical loads. This behaviour can be determined, as with the stress relaxation, by suitable experiments and can be described mathematically. For the appropriate computation of the thermal residual stress, the volume relaxation function was determined. The concept of the fictive temperature T_F was used in order to compute the relaxation of the glass characteristic during change of temperature. The relationship between volume relaxation function $M_V(t,T)$ and the fictive temperature during change of temperature of T_0 to T is given by [8]

$$M_V(t,T) = \frac{V(t)-V(\infty)}{V(0)-V(\infty)} = \frac{T_F-T}{T_0-T} . \tag{9}$$

$V(t)$ is here glass volume at time t at temperature T. $V(0)$ and $V(\infty)$ are measured volumes at time 0 and "infinite" time at temperature T. For illustration, the change of specific volume is represented in figure 4.

Application to thermal expansion results in

$$\varepsilon_{\text{thermal}} \equiv \alpha_l \cdot (T_F - T_0) + \alpha_g \cdot (T - T_F) \tag{10}$$

with α_l the thermal coefficient for glass in the liquid state and α_g the thermal coefficient for glass in the glassy state. Both are measured at temperatures with relaxation time
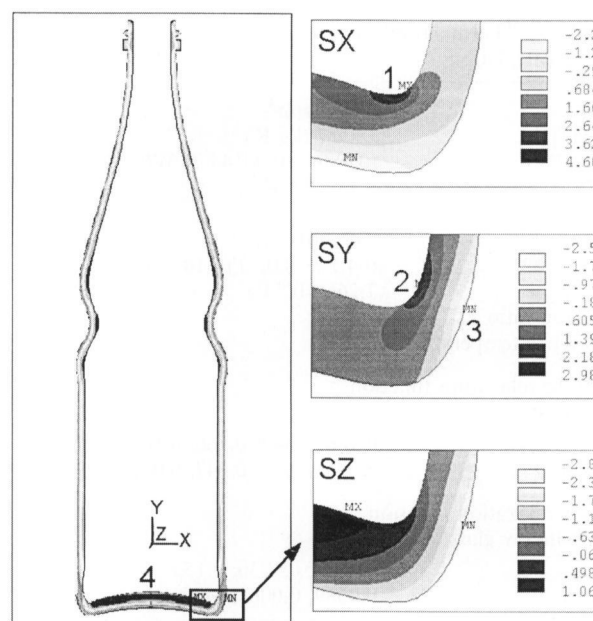
equal to zero or an infinitely large relaxation time. They are therefore constant [8]. In order to compute the reduced time, it was supposed that similarly to glass viscosity, the Arrhenius equation is applied between the relaxation time τ_T and the temperature T

$$\tau_T \equiv \tau_0 \cdot \exp\left(\frac{H}{R\,T}\right) \tag{11}$$

where τ_0 is a constant, H is the activation energy and R is the gas constant. Similarly to equation (10), the activation energy H within the transformation range is composed of the activation energy of the glass in the glassy condition H_g and the part in the liquid condition H_l. Then, equation (12) holds for the relaxation time

$$\tau_T \equiv \tau_0 \cdot \exp\left[\frac{H_g}{R\,T} + \frac{H_l}{R\,T_F}\right] . \tag{12}$$

The shift function according to Narayanaswamy for glass is [9]

$$\ln \frac{\tau_R}{\tau_T} = \frac{H}{R}\left[\frac{1}{T_R} - \frac{x}{T} - \frac{(1-x)}{T_F}\right] \tag{13}$$

with $H = H_g + H_l$ and $x = \dfrac{H_g}{H}$.

In order to compute the residual stress remaining after the relaxation the "memory integral" is used [10]

$$\sigma(\xi) = \int_0^{\xi} E(\xi-\xi') \frac{\partial(\varepsilon-\varepsilon_{\text{thermal}})}{\partial \xi'} \, d\xi' . \tag{14}$$

As a simulation object, the standard bottle for mineral waters of Gesellschaft Deutscher Brunnen (see figure 5) was

Table 1. Thermal and mechanical glass parameters [9] for stress calculation

ρ	2500 kg/m^3
c_p [11]	1300 J/(kg K)
λ_{activ} [12]	$(0.483 + 0.00144\ T)$ W/(m K)
H/R	$6.45 \cdot 10^4$ K
x	0.53
α_l	$343 \cdot 10^{-7}$ K^{-1}
α_g	$(64.7 + 0.02\ T) \cdot 10^{-7}$ K^{-1}
E	$7.26 \cdot 10^{10}$ Pa
Poisson ratio	0.3
reference temperature	891 K

volume relaxation function
for window glass:

$C_{i=1,2,3,4,5,6}$	0.108; 0.443; 0.166; 0.161; 0.046; 0.076
$\lambda_{i=1,2,3,4,5,6}$	3.000; 0.671; 0.247; 0.091; 0.033; 0.008

stress relaxation function
for window glass:

$C_{i=1,2,3}$	0.4220; 0.4230; 0.155
$\lambda_{i=1,2,3}$	0.0689; 0.0065; 0.0001

selected. The furnace temperature in the relaxation zone was specified at 550 °C and at the end of the critical cooling zone at 475 °C. The cooling time in different cooling zones, the convective heat transfer coefficient and bottle temperature were varied. The convection at the internal bottle surface was neglected. In table 1 the data are arranged for the computation. The simulation was accomplished with the FEM code ANSYS.

3. Results and discussion

First the change of stress in the bottle during the entire annealing process was examined. The cooling curve of the furnace is indicated as in figure 1. For the calculation, the following conditions have been assumed:

– a starting temperature of 450 °C for bottles entering the furnace,
– this temperature is homogeneous in the entire bottle,
– the convection coefficient in the whole furnace is constantly supposed to be at 200 W/(m^2 K).

The simulation results show that during the entire process particularly high tensile and compressive stresses located at the transient area from the bottle wall to the bottom. Figure 5 shows the stress profiles in the x, y and z direction in this place at the end of an annealing process. The computed maximum stresses and their locations are relatively well corresponding to some measured stresses in beer and champagne bottles, where the maximum tensile stress on the internal surface and the maximum compressive stress on the external surface at about 5 MPa were determined [13].

Figure 6a shows the temperature evolution in the locations 1 and 4 in the bottle and figure 6b the corresponding stress evolution in the locations 1 to 3 at the transient area (see figure 5) between wall and bottom, where maximum stresses in x and y direction arise. Positive stresses mean tensile stresses and negative ones mean compressive stresses. The occurrence of an extremely high tensile stress during the heating-up of the bottle to the relaxation temperature
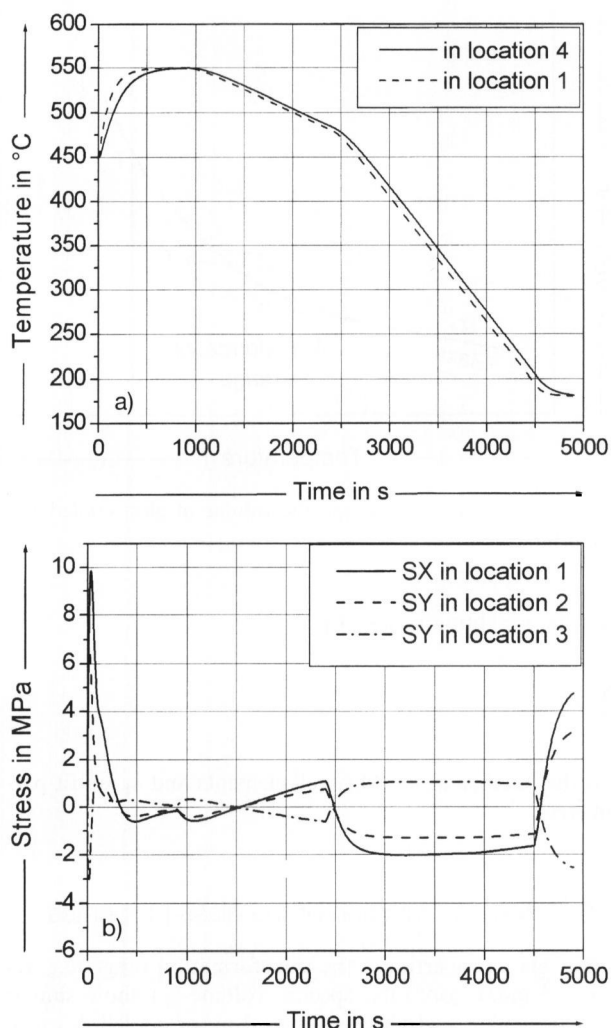
Figures 6a and b. Temperature evolution in locations 1 and 4 (figure a); and stress evolution and relaxation in the annealing process, locations 1 and 2 with high tension in x and in y direction, respectively, and location 3 with high compression in y direction (figure b).

must be taken into consideration. The intensity of this stress depends on the wall thickness, on the convective heat transfer and on the initial temperature of the bottle (figure 7) and can cause the breakage of the bottle. It is advisable to put the bottles into the furnace as hot as possible in order to avoid a high temperature shock.

After reaching the annealing temperature (> 520 °C) the glass stresses are rapidly relieved in a few minutes. For a successful relaxation procedure, the coldest place in the bottle must therefore be heated up to the annealing temperature.

High bottle temperature at the beginning shortens this necessary heating-up time. In figure 8 the dependence of the necessary heating-up time on the convective heat transfer coefficient in the furnace for bottle thicknesses of 3 and 7 mm is shown.

The duration of the annealing process can also be derived from the experimental data of Kurkjian [7] (figure 2). Glass relaxes e.g. at 533 °C already after a few minutes and at 473 °C only after several hours. At the end of the relax-
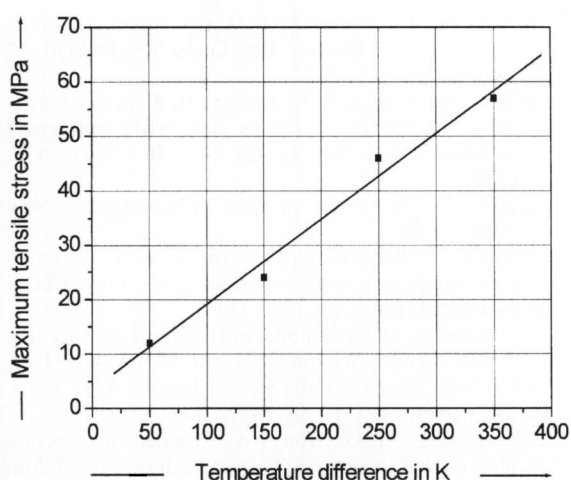
Figure 7. Maximal tensile stress (in location 1) as a function of the temperature difference between bottle and furnace at the start of the process.
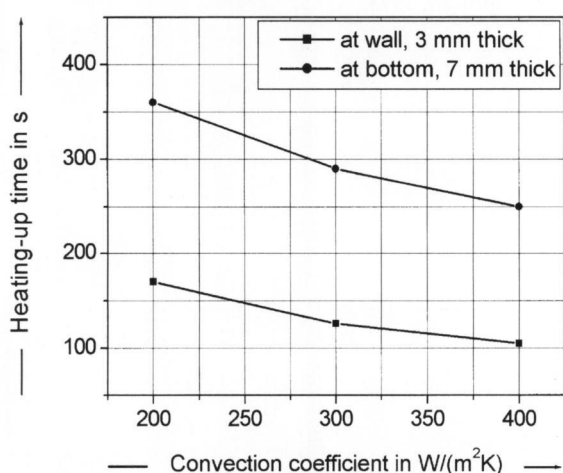

Figure 8. Necessary heating-up time as a function of convection coefficient and bottle thickness.
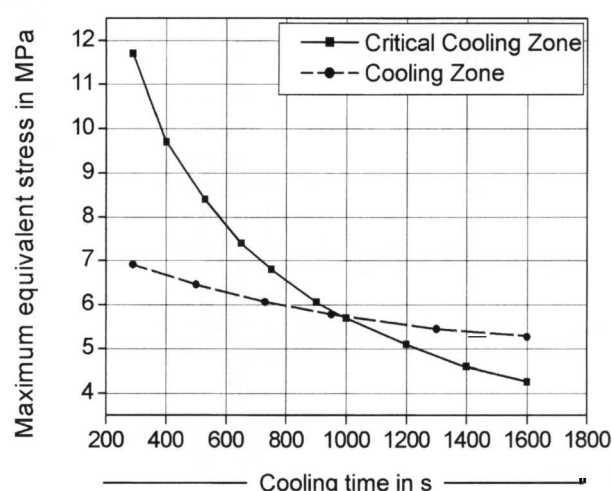

Figure 9. Dependence of the maximum equivalent stress on the retention time in the critical cooling zone and in the last cooling zone.
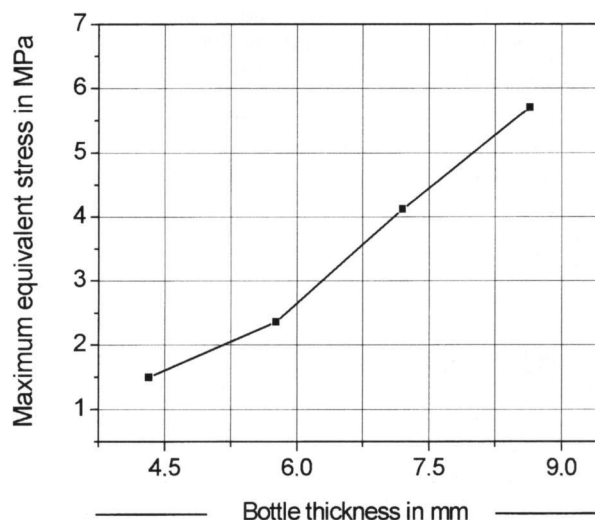

Figure 10. Dependence of the maximum equivalent stress on the bottle thickness measured at the bottom centre.

ation zone the bottle should be stress-free. The residual stress in the bottle at the end is only determined by the retention time in the following cooling zones.

When examining the dependence of the residual stress on the retention time in the cooling zones, ideal convection conditions were supposed so that the surface temperature of the glass bottle is equal to the furnace temperature in the respective place in the furnace. As a measure for the break criterion of a solid material, one uses the equivalent stress which is computed from all six stress components according to

$$\sigma = \left\{ \frac{1}{2} \left[(\sigma_x\text{-}\sigma_y)^2 + (\sigma_y\text{-}\sigma_z)^2 + (\sigma_z\text{-}\sigma_x)^2 \right. \right. \tag{15}$$
$$\left. \left. + 6(\sigma_{xy}{}^2 + \sigma_{yz}{}^2 + \sigma_{zx}{}^2) \right] \right\}^{\frac{1}{2}}.$$

Here, σ_x, σ_y and σ_z are the main stresses in x, y and z direction and σ_{xy}, σ_{yz} and σ_{zx} the shear stresses in the xy, yz and zx area of the coordinate system [9].

Figure 9 shows a strong dependence of the equivalent stress on the retention time in the so-called "critical" cooling zone t(critical) and a small dependence on the retention time in the last cooling zone t(fast). By fitting the curves equation (16) is obtained for the estimation of the cooling tensions in the bottle from the retention times:

$$\sigma_{\text{maximal}} = \left(4 + 14 \cdot e^{\frac{-t(\text{critical})}{467}} \right) \left(0.79 + 0.49 \cdot e^{\frac{-t(\text{fast})}{865}} \right). \tag{16}$$

If the wall thickness of the bottles is proportionally increased, the maximum equivalent stress increases approximately proportionally. Figure 10 shows this relationship. The bottle thickness here is measured in the centre of the bottle bottom.

4. Conclusions

The stress simulation according to viscoelastic theory is suitable to estimate thermal stresses in glass. It enables one

to determine and optimize cooling parameters in particular for new glass products with complicated geometry.

The maximum cooling stresses are located in the transition range from wall to bottom when bottles are cooled. For a successful annealing process it is important to heat up the entire glass bottle to the relaxation temperature. The heating time depends strongly on the initial temperature and the wall thickness of the bottles. The residual stress becomes particularly dependent on the cooling speed in the critical cooling zone and insignificantly on the cooling speed in the last cooling zone. In order to reduce the residual stresses in glass, the best way is to extend the critical cooling zone. A long last cooling zone is not necessary.

5. References

[1] Brückner, R.: Kühlung der Gläser. Glashütten-Handbuch. Hüttentechnische Vereinigung der Deutschen Glasindustrie (HVG). Copyright 1965.

[2] Soukup, W.: Pers. report, Company HORN Thermo Process and Machinery GmbH, 2002.

[3] Krause, D.; Loch, H.: Mathematical Simulation in Glass Technology. Berlin et al.: Springer, 2002.

[4] Fotheringham, U.; Lentes, F.-T.: Active thermal conductivity of hot glass. Glastech. Ber. Glass Sci. Technol. **17** (1994) no. 12, pp. 335−342.

[5] Burger, A. M.; Soules, T. F.; Busbey, R. F. et al.: Finite-element calculation of stresses in glass parts undergoing viscous relaxation. J. Am. Ceram. Soc. **70** (1987) no. 2, pp. 91−95.

[6] Scherer, G. W.: Relaxation in glass and composites. New York: Wiley, 1986.

[7] Kurkjian, C. R.: Relaxation of torsional stress in the transformation range of a soda-lime-silica glass. Phys. Chem. Glasses **4** (1963) no. 4, pp. 128−136.

[8] Narayanaswamy, O. S.: A model of structural relaxation in glass. J. Am. Ceram. Soc. **54** (1971) no. 10, pp. 491−498.

[9] ANSYS 8.0, Theory Reference, Chapter 4.7. Viscoelasticity. Canonsburg, PA: ANSYS, Inc.

[10] Lee, E. H.; Roger, T. G.; Woo, T. C.: Residual stresses in a glass plate cooled symmetrically from both surfaces. J. Am. Ceram. Soc. **48** (1971) no. 9, pp. 491−498.

[11] Coenen, M.: Spezifische Wärme von Schmelzen und Gläsern. Glastechn. Ber. **50** (1977) no. 5, pp. 115−120.

[12] Bansal, N. P.; Doremus, R. H.: Handbook of Glass Properties. Orlando, FL et al.: Academic Press, 1986.

[13] Aben, H.; Guillement, C.: Photoelasticity of Glass. Berlin et al.: Springer, 1993.

■ E405P001

Contact:

Dipl.-Ing. C. Phan Dang
Forschungsstelle WOPAG
LTTT, Universität Bayreuth
Universitätsstraße 30
D-95440 Bayreuth
Email: cuong.phandang@uni-bayreuth.de