

## Mathematical Description of Non-Newtonian Flow of Polymeric Melts in Thermoforming Process

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### ABSTRACT

Wide applications of thermoforming are due to its high performance, simplicity, compactness and relatively low-cost equipment. These issues make it possible to produce complex, large-scale configurations and free form shapes of products.

The fundamental defects inherent to the thermoforming technology are wall-thickness variation and warpage of the products due to inadequate thermal processing and frozen-in stresses during production of polymer. A nonlinear viscoelastic rheological model is implemented for developing the process model. This model describes deformation process of a sheet in thermoforming process. The results shown in our work not only leads to the understanding of the cause of these problems, but also enables us to formulate calculation of parameters that affect warpage and wall-thickness variation of polymeric sheets during plug-assist vacuum thermoforming. The gained experimental results confirm our presented model.

### INTRODUCTION

The polymer processing the production of all forms of polymeric articles has found a great place in chemical industries. Thermoforming process is one of the most popular techniques in this field. It applies thermoplastic sheet or film-forming

techniques for various packaging applications such as medical devices, food containers, and pharmaceuticals [1-4]. Wide applications of thermoforming are due to its high performance, simplicity, compactness and relatively low-cost equipment. These issues make it possible to produce complex, large-scale configurations and free form shapes of products. In thermoforming, a heated plastic sheet is stretched into a mould cavity by applying pressure, eventually assisted by direct mechanical loading [5,6]. Upon contacting of a sheet with the cold surface of the mould, the sheet prevents from undertaking any further deformation. The forming sequence induces a thickness variation in the final part. Besides wall-thickness variation, other problems facing the thermoforming industry lie mainly with physical instabilities during inflation – rupture of sheet and shrinkage [7,8] exhibited in the final parts. There are many ways to stretch sheets: vacuum, air pressure and mechanical aids such as implementation of a plug. For increasing the quality of products such as narrow wall-thickness tolerance or elimination of frozen-in stresses, a combination of mechanical and vacuum or pressure forming methods may be implemented. In early step, it involves the usage of mechanical pre-stretching with plug and then vacuum or pressure forming is applied. Figure 1 illustrates this process.

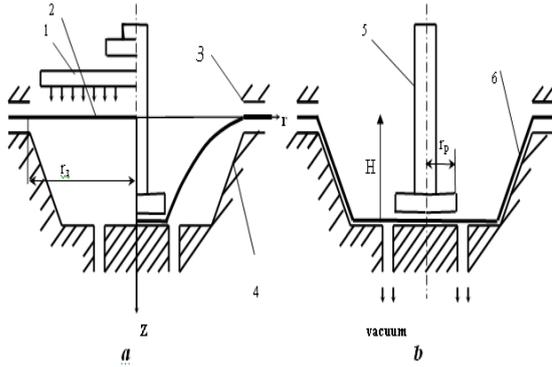


Fig. 1. Flow diagram of mechanical-vacuum thermoforming (a - heater and plug-assist ; b - vacuum thermoforming ): 1 - heater, 2 - sheet prior to forming, 3 - clamp frame, 4 - mold, 5- plug, 6- deformed sheet.

Another fundamental defect inherent to the thermoforming technology is warpage of the products during their application which becomes particularly apparent under high temperatures (Fig.2). The warpage defect is understood as the process of non-uniform (heterogeneous) change of the geometrical dimensions of products in time resulting in a change (distortion) of their original form. Unfortunately, this problem is an overlooked area in thermoforming research.



Fig.2. The warpage of a polymeric product during its application

The results of this work allow us to find out the causes of this warpage and ascertain the conditions that give rise to this defect, thereby making it possible to work out valid

recommendations for its partial and, in some cases, complete elimination.

### RHEOLOGICAL MODEL OF PLUG-ASSIST THERMOFORMING

As mentioned above, shrinkage and warpage prediction is very important due to processing constraints and finally may cause system failure. Successful solution to it directly depends on the right selection of a proper rheological model. One of the theoretical models in this area was developed by Leonov [9]:

$$\begin{cases} \bar{\sigma} + p\bar{\delta} = 2\bar{c}W_1 - 2\bar{c}^{-1}W_2 & (1) \\ \bar{e}_f = \frac{1}{\theta_0(T)G_0(T)} \cdot [f(I_1, I_2)]^\psi \cdot \left[ \left( \bar{c} - \frac{I_1}{3}\bar{\delta} \right) W_1^* - \left( \bar{c}^{-1} - \frac{I_2}{3}\bar{\delta} \right) W_2^* \right] \\ \frac{d\bar{c}}{dt} + \bar{\omega}\bar{c} - \bar{c}\bar{\omega} - \bar{c}(\bar{e} - \bar{e}_f) - (\bar{e} - \bar{e}_f)\bar{c} = 0 \end{cases}$$

Where:

$\bar{\sigma}$  is stress tensor; P is Lagrange multiplier, determined by boundary condition;  $\bar{\delta}$  is identity tensor;  $\bar{c}$  is Cauchy strain tensor;  $\bar{e}_f$  is flow strain rate tensor;  $\bar{\omega}$  is vortex tensor;  $\bar{e}$  is strain rate tensor;  $\theta_0(T)$  is relaxation time; T is temperature;  $G_0(T)$  is tensile modulus; W is strain energy function  $W = W(I_1, I_2)$ ;  $I_1$  and  $I_2$  are primary and secondary strain tensor invariants respectively,  $\psi$  is dimensionless parameter ( $\psi = 0$  at  $\bar{\omega} = 0$  and  $\psi = 1$  at  $\bar{\omega} \neq 0$ );  $f(I_1, I_2)$  is dimensionless function that defines relaxation time, and  $2W^S = W(I_1, I_2) + W(I_2, I_1)$  represents the symmetric function of W.

The last one can be shown by:

$$\begin{aligned} W_1 &= \frac{\partial W}{\partial I_1} & W_2 &= \frac{\partial W}{\partial I_2} & W_1^S &= \frac{\partial W^S}{\partial I_1} \\ W_2^S &= \frac{\partial W^S}{\partial I_2} \end{aligned}$$

But in practice there is a problem for application of Eq.(1). This problem arises from the choice of strain energy function  $W = W(I_1, I_2)$ . Most researchers use Mooney-Rivlin potential, but there are differences between experimental and theoretical results for prediction of stress and strain. Results of recent research show that in various kinematical deformations, the following potential can be used [10]:

$$W = 0.25G_0(I_1 + I_2 - 6) \quad (2)$$

Now, by utilizing Eq.(1) with regards to strain energy function (2), a mathematical description for deformation process in mechanical pre-stretching thermoforming can be developed (figure 1a).

#### MATHEMATICAL DESCRIPTION

An analysis of the produced results shows that warpage is caused by the process of relaxation of residual (frozen-in) stress. This stress was accumulated during thermoforming process. This can be proved by examination of the abstract task of describing the aftereffects of a deformation process on a viscoelastic polymer. It preliminarily was subjected to a plug with a determined intensity in the conditions of pure shear. In this case, the kinetics of the development of general (viscoelastic) and elastic deformations resulting from a mechanical pre-stretching with plug on the polymer. Assuming that at a certain moment of time  $\tilde{t}_\phi$ , the mechanical pre-stretching was instantly ceased by the removal of any external influence (plug) on polymer. At this time, due to deformation, a quite certain level of general and elastic deformations is accumulated -  $\varepsilon^H(\tilde{t}_\phi)$ ,  $\varepsilon_e^H(\tilde{t}_\phi)$ . This condition corresponds to the "free" condition (not influenced from the outside) of the polymer in the molded product. According to the rheological model (1), for a polymeric medium in a "free" condition the relaxation process will occur at varying

speeds as the level of accumulated elastic deformations reduces. The carrying out of this process is characterized by the following natural kinematic conditions:

$$\bar{e} = -\bar{e}_f \quad (3)$$

The physical meaning of the last condition is that a polymer in absence of an external influence, the speed of deformation changes are determined only by the typical speed of the relaxation processes.

Using the condition (3) from the rheological model (1) for the examined and kinematically determined process of deformation aftereffects in the viscoelastic polymeric sheet, the following system of differential equations can be obtained that describes the kinetics of the process:

$$\begin{cases} \frac{dc}{dt} = -(c^2 - 1) \frac{1}{2\theta_0(T)} \exp[-\beta(c + c^{-1} - 2)] \\ \frac{d\varepsilon^H}{dt} = -(c - c^{-1}) \frac{1}{4\theta_0(T)} \exp[-\beta(c + c^{-1} - 2)] \end{cases} \quad (4)$$

Where :

$$c \equiv \lambda_e^2 = \exp(2\varepsilon_e^H),$$

$\varepsilon^H$  :: Hencky strain,

$\varepsilon_e^H$  :: elastic Hencky strain,

$\lambda_e$  :: elastic strain ratio.

The initial conditions for solving the system of equations (4) -  $\varepsilon^H(\tilde{t}_\phi)$ ,  $\varepsilon_e^H(\tilde{t}_\phi)$  are determined by solving of deformation influences on the polymer. From the analysis of the equations (4) it follows that

the elastic deformations accumulated ( $\varepsilon_e^H$ ) during deformation influence on the polymer relax (the first equation) with

varying speed upon the removal of the mechanical field (plug) from the polymer. This leads to the process of a reduction in accumulated general deformations (second equation) which is going to be proved. For rigid-chain polymers ( $\theta_0 \rightarrow 0$ ) or for a small level of accumulated elastic deformations ( $\exp(\varepsilon_e^H(\tilde{t}_\phi)) \rightarrow 1$ ), the solution of the system of equations (4) will look as follows:

$$\left\{ \begin{aligned} \varepsilon_e^H(\tilde{t}) &\approx \frac{1}{2} \ln \frac{1 + \exp(2\varepsilon_e^H(\tilde{t}_\phi)) \exp[(\tilde{t} - \tilde{t}_\phi)] - 1}{1 - \exp(2\varepsilon_e^H(\tilde{t}_\phi)) \exp[(\tilde{t} - \tilde{t}_\phi)] + 1} \\ \varepsilon_e^H(\tilde{t}) &\approx \varepsilon_e^H(\tilde{t}_\phi) - \frac{1}{4\tilde{t}_\phi} \int_{\tilde{t}_\phi}^{\tilde{t}} [\exp(2\varepsilon_e^H(\tilde{t})) - \exp(-2\varepsilon_e^H(\tilde{t}))] d\tilde{t} \end{aligned} \right. \quad (5)$$

Where  $\tilde{t} = \frac{t}{\theta_0(T)}$  :: dimensionless time,

$\tilde{t}_\phi$  :: dimensionless time of plug-assist forming.

Figure 3 represents the kinetics of the free relaxation process of elastic deformations in viscoelastic polymers resulting in the parallel process of changing its original dimensions.

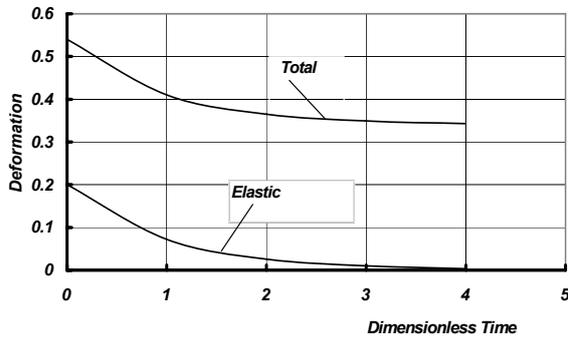


Fig.3. The kinetics of the free relaxation process of elastic deformations.

## RESULTS AND DISCUSSION

The stated above clearly proves that the cause of the warpage of molded products during their application is the accumulation of elastic deformations in the polymer. Since the accumulated elastic deformations in the polymer can essentially be distributed non-uniformly, in this case the relaxation process will run at essentially different speeds. This will in turn result in noticeably different speeds of dimensional change in various parts of the product, i.e. in the warpage which is evidenced by the equation (5). Thus, on the basis of this analysis one may state that the warpage originates under the condition of accumulation of heterogeneous elastic deformations in the polymer during the process of thermoforming. This issue manifests itself through the relaxation process of these deformations.

Gained equations (5) show that the process of warpage is essentially determined by one of the fundamental rheological characteristics of polymers, the typical relaxation time, ( $\theta_0(T)$ ), which is temperature-dependent. By the reduction of required time for the relaxation at the expense of increasing in the polymer's temperature, the relaxation processes in polymers run faster. Thus the speed of warpage consequently increases in such conditions.

By introduction of a quantitative characterization of the warpage with time, the notion of a coefficient of dimensional change which integrally characterizes the relative change in the original dimension of a product, determined as follows:

$$k_{u\phi}(\tilde{t}) = 1 - \frac{S(\tilde{t})}{S(\tilde{t} = \tilde{t}_\phi)} \quad (6)$$

where  $S(\tilde{t})$ ,  $S(\tilde{t} = \tilde{t}_\phi)$  – respectively are the current area of the lateral surface of the distorted article and original area of the lateral surface of the product.

By using well-known equations  $(S(t) = 2\pi \int_0^H r(z) \sqrt{1 + \left(\frac{dr}{dz}\right)^2} dz)$  and the mathematical results in this work, the following determining expression is obtained.

$$S(\tilde{t} = \tilde{t}_\phi) = 2\pi r_3 \sqrt{b} \int_0^{\tilde{H}} \frac{\sqrt{(\sqrt{b} + \tilde{z})^4 + b}}{(\sqrt{b} + \tilde{z})^3} d\tilde{z} \quad (7)$$

$$S(\tilde{t}) = 2\pi r_3 \sqrt{b(\tilde{r}_0 = 1, \tilde{t})} \int_{\tilde{r}_0 = \tilde{r}_p}^{\tilde{r}_0 = 1} \frac{\exp(\varepsilon^H(\tilde{r}_0, \tilde{t}))}{\tilde{r}_0} d\tilde{r}_0 \quad (8)$$

Where

$$b(\tilde{t} = \tilde{t}_\phi) \equiv \lambda^2(\tilde{z} = 0) - 1 = \left( \tilde{H} \cdot \frac{\tilde{r}_p}{1 - \tilde{r}_p} \right)^2,$$

$$\tilde{r}_p \equiv \frac{r_p}{r_3}, \tilde{H} \equiv \frac{H}{r_3}, \tilde{z} \equiv \frac{z}{r_3}, \tilde{r}_0 = r_0 / r_3$$

$b(\tilde{r}_0 = 1, \tilde{t}) \equiv \lambda^2(\tilde{r}_0 = 1, \tilde{t}) - 1 = \exp(2\varepsilon^H(\tilde{r}_0 = 1, \tilde{t})) - 1$   
 $r_p$  :: plug radius;  $r_3$  :: initial radius of sheet;  $r_0$  :: current radius of sheet during plug-assist;  $r, z$  :: the horizontal and vertical axes, respectively;  $H$  :: depth of deformed sheet.

And the function  $\varepsilon^H(\tilde{r}_0, \tilde{t})$  is specified by the solution of the differential equations system (4).

By using the expressions (7) and (8), for equation (6), the required equation for describing of the kinetics of the warpage in the product is obtained. The characteristic of the gained expression in conjunction with experienced data is presented in figure 4.

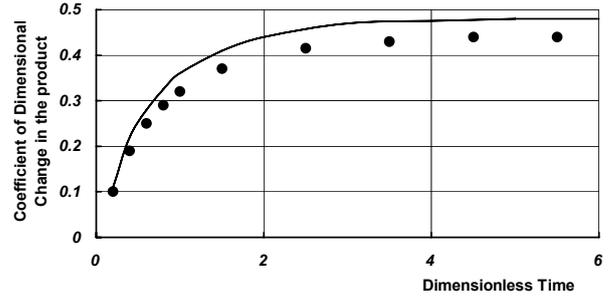


Fig.4. The kinetics of the warpage of HIPS at  $T=353K$ ,  $\square \square \square \tilde{t}$  sec. (curve-Eq.(6) and points-experimental data).

Now by assigning an allowable tolerance value for dimensional change coefficient of a product, ( $[k_{u\phi}]$ ), in a specific application, the maximum permissible time can be determined based on the following condition:

$$[k_{u\phi}] = k_{u\phi}(\tilde{t}) \quad (9)$$

where the function  $k_{u\phi}(\tilde{t})$  is determined in equation (6).

Based on the analysis of the condition in equation (9), it becomes clear that the application period of a polymeric product under given temperature conditions with the allowable coefficient of dimensional change is essentially determined by the elastic deformations accumulated, ( $\varepsilon(r_0, \tilde{t}_\phi)$ ), in the product in its formation process. Smaller elastic deformations accumulated mean longer operational period of the product. Based on this concern, a completely practical task for organizing the plug-assist vacuum-forming methods arises, so that the total level of elastic deformations (i.e. the accumulated deformations during both stages of the article's molding) in polymeric materials must be minimal. As the molding technology is consisted of two stages, it is necessary to analyze each of the stages.

It is well known that the stage of vacuum-forming with a pre-stretched sheet

occurs too quickly for relaxation processes in the polymeric sheet to be experienced. This will result in this fact that almost all accumulated deformations in polymeric materials within this stage will be elastic. Consequently, the minimization of the accumulation may be realized only by minimizing of the general deformations accumulated in polymeric sheet during this stage. For practical purposes, this means that the profile of the pre-stretched sheet should be maximally approximated by the profile of the final product which could be assured based on the application of a plug with the respective radius.

In contrast to the second stage, the stage of mechanical pre-stretching by a plug can be regulated in the sense that it is technically possible to control the motion of the plug. This creates a practical opportunity that at this stage to organize the relaxation process of elastic deformations accumulated in a polymeric sheet during the process of mechanical pre-stretching with plug. The essence of the simplest of many variants used in the realization of this process lies in the creation of a relaxation pause period between the first, mechanical pre-stretching, and the second, vacuum forming, stages. During this period the accumulated elastic deformations can completely or partially are relaxed. The only problem may arise here is that during this pause period the pre-stretched sheet would not have time to cool to a certain temperature, thereby making a realization for the second technological stage of the thermoforming process impossible. Evaluation of the necessary duration for this pause that would ensure the realization of the relaxation processes.

Since during the relaxation pause, a plug holds a stretched sheet, one of the kinematic conditions of this process corresponds to the absence of an external deforming influence will be in the following form:

$$\bar{\varepsilon} = 0 \quad (10)$$

By using the condition (10) from the rheological model (1) for the kinematically determined process of the relaxation in a viscoelastic polymeric medium, the following equations can be obtained that describes its kinetics:

$$\begin{cases} \frac{dc}{dt} = -\frac{1}{2}(c^2 - 1) \frac{1}{\theta_0(T)} \exp[-\beta(c + c^{-1} - 2)] \\ \frac{d\varepsilon^H}{dt} = 0 \end{cases} \quad (11)$$

Figure 5 presents the kinetics of change of elastic deformations in a sheet stretched with a plug during the relaxation pause.

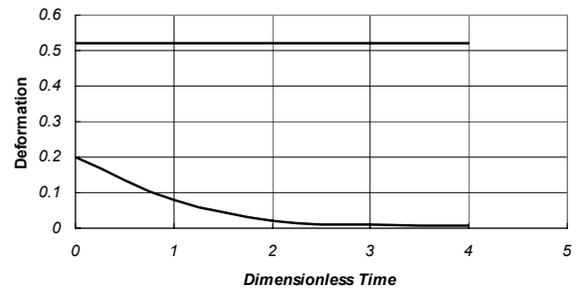


Fig.5. The kinetics of the relaxation process of elastic deformations in viscoelastic polymer during the relaxation pause.

Comparison of the differential equation systems (11) and (4), it is not difficult to conclude that the relaxation processes in polymers with limited conditions and free aftereffects are distinguishable only by speed.

This is quite logical considering that the limited conditions of the relaxation aftereffect do not completely correspond to the removed mechanical field that superimposed on a polymeric medium which also slows down the development of the relaxation processes. For rigid-chain polymers, or at a moderate level of the accumulated elastic deformations ( $\exp(\varepsilon_e^H(r_0, t_\phi)) \rightarrow 1$ ), the analytical

solution of the equations system (11) will be in following form:

$$\left\{ \begin{array}{l} \varepsilon_e^H(r_0, \tilde{t}) \approx \frac{1}{2} \ln \frac{\frac{1 + \exp(2\varepsilon_e^H(r_0, \tilde{t}_\phi))}{1 - \exp(2\varepsilon_e^H(r_0, \tilde{t}_\phi))} \exp[(\tilde{t} - \tilde{t}_\phi)] - 1}{\frac{1 + \exp(2\varepsilon_e^H(r_0, \tilde{t}_\phi))}{1 - \exp(2\varepsilon_e^H(r_0, \tilde{t}_\phi))} \exp[(\tilde{t} - \tilde{t}_\phi)] + 1} \\ \varepsilon^H(r_0, \tilde{t}) = \varepsilon^H(r_0) = const \end{array} \right. \quad (12)$$

The analysis of the equations obtained by the numerical solution of differential equation (11) for the most common cases shows that even in the most unfavorable conditions, ( $\beta > 1$  and  $c(t_\phi = 0) \approx 3$ ), the accumulated elastic deformations will practically comes to a complete relax within a period of time that not exceeds from  $9\theta_0(T)$ , approximately. Considering the fact that for most thermoplastic materials at average temperature values of the recommended processing temperature range, the typical relaxation time will be about in range of 0.2 to 0.5 second, the conclusion that even in the most unfavorable situations, the maximum time necessary for the relaxation processes in polymers would not exceed 4.5 seconds.

From [11] it is not difficult to determine that, during this time period, a polymeric sheet with a certain thickness, e.g. 1.0 mm, at a temperature of  $T=140^\circ\text{C}$  is cooled down only about  $10^\circ\text{C}$  (thermodynamic figures of similarity of the examined conditions of cooling:  $Bi \approx 0.04$  and  $Fo \approx 1.75$ ). This reduction in temperature is not critical for the second stage of product formation even considering the time spent on the stage of mechanical pre-stretching.

For thinner sheets the maximum relaxation pause period will obviously become critical. But this criticality issue will arise only under very strict requirements

concerning the quality of the final products according to the admissible criterion of their distortion,  $[k_{u\phi}] \rightarrow 0$ . In less strict conditions when some level of distortion, determined by the value of the allowable coefficient ( $[k_{u\phi}]$ ), is admitted, the technological time necessary for the relaxation pause can be determined as follows:

In this case, the minimum duration of the relaxation pause period should be a value that guarantees an infinitely large application time of a product with the allowable coefficient  $[k_{u\phi}]$ .

Nominally, considering (6), this may be expressed as follows:

$$S(\tilde{t} \rightarrow \infty) \geq S(\tilde{t}_\phi) \cdot (1 - [k_{u\phi}]) \quad (13)$$

It is clear from the analysis (8) that the function  $S(\tilde{t})$  is determined by solving the differential equation systems (4). Consequently, solving the equations under different initial conditions  $-\varepsilon^H(r_0)$ , and  $\varepsilon_e^H(r_0, \tilde{t}_\phi)$ , it is always possible to find such a solution which would critically satisfy the condition (13). Based on the results obtained in this work, the following expressions determine the initial conditions specified.

$$\varepsilon_e^H(r_0, \tilde{t}_\phi) = \frac{1}{2} \ln \left[ \frac{1}{2} \left( \frac{c_0^2(\tilde{t}_\phi) - 1}{c_0(\tilde{t}_\phi)} \cdot \frac{1}{1 + b(\tilde{t}_\phi)} \cdot \frac{\tilde{r}_0^4 + b(\tilde{t}_\phi)}{\tilde{r}_0^3} + \sqrt{\left( \frac{c_0^2(\tilde{t}_\phi) - 1}{c_0(\tilde{t}_\phi)} \cdot \frac{1}{1 + b(\tilde{t}_\phi)} \cdot \frac{\tilde{r}_0^4 + b(\tilde{t}_\phi)}{\tilde{r}_0^3} \right)^2 + 4} \right) \right]$$

$$\varepsilon^H(r_0) = \frac{1}{2} \ln \left[ \left( \frac{\tilde{H} \tilde{r}_p}{(1 - \tilde{r}_p) \tilde{r}_0^2} \right)^2 + 1 \right] \quad (14)$$

It follows from (14) that the procedure of the equation systems (4) critically satisfying

the condition (13) is actually reduced to a search of the critical (maximum) value of the elastic deformation,  $(c_{*0}(\tilde{t}_\phi))$ . This should not be exceeded in a certain section of a stretched sheet by the end of the relaxation pause period. Furthermore, this will determine the fulfillment of the condition (13). As soon as the value  $c_{*0}(\tilde{t}_\phi)$  is determined, the duration of the necessary relaxation pause can be easily determined by solving the first differential equation (11). For the special case specified above, substituting the obtained value as current and corresponding to the end of the relaxation pause period in the first equation of the equations system (12), an expression for determining the operational factors of the relaxation pause period is obtained. This ensures the realization of the relaxation process of elastic deformations to the required level:

$$t_{pn} \approx \theta_0(T) \ln \left\{ \frac{c_0(\tilde{t}_\phi) - 1}{1 + c_0(\tilde{t}_\phi)} \cdot \frac{1 + c_{*0}(\tilde{t}_\phi)}{c_{*0}(\tilde{t}_\phi) - 1} \right\} \quad (15)$$

Confirmation of this model is exhibited in figure 6.



Fig.6. Comparison of warpage in PS products (**a**- initial product at T=298K without warpage, **b**- at T=353K with  $t_{pn} = 2$  sec., **c**- at T=353K without relaxation pause).

As expected, under the same conditions, the warpage of a product without a relaxation pause is much greater than the

warpage of a product with a relaxation pause. This issue shows the importance of a relaxation pause in reducing warpage of products during their application, especially under high temperatures.

Based on the results obtained in this work, another method can be developed for reducing the level of elastic deformations of sheet stretching. It calculates lower speed of plug movement. However, in this case the time of sheet stretching will exceed the total time spent on its stretching at the normal speed of plug movement and the relaxation pause.

This is quite evident considering that in the first stage a more active mechanical force is imposed to the polymeric materials than in the process of the relaxation pause period. Consequently, the speed of the relaxation processes in the first stage will be less than in the second stage. We should also mention that the time necessary for the relaxation processes in the relaxation pause period can be reduced by raising the sheet temperature. However, as it mentioned, the existing temperature range ( $T_g < T < T_m$ ) that is within the practical achievement of the examined technology is recommended, specifies a limit on practical use of this operating condition in order to solve the defect.

## CONCLUSION

The analysis conducted to the causes and conditions of the products warpage allows us to develop:

- A method for calculating the operational conditions of molding products. It would ensure operation of the final product within admissible warpage values.
- A method for calculating the maximum admissible period for application of polymeric products within which the warpage of products would not exceed the allowable values.

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