Rheological Modeling of Fracture in Thermoplastic Polymers During Thermoforming Processes

H. Hosseini[1], B. V. Berdyshev[2], and A. Mehrabani[3]

[1] University of Tabriz, Department of Chemical Engineering, Tabriz, Iran
2 Moscow State University of Environmental Engineering, Moscow, Russia
3 Isfahan University of Technology, Isfahan, Iran

ABSTRACT

Fracture of thermoplastic sheets is one of the practical problems during plug-assist vacuum thermoforming. This defect can occurs during both stage of plug-assist and vacuum forming. This paper focuses on two issues, (i) origins of fracture creation, and (ii) determination of the process parameters for removal of the defect. The results shown in our work not only leads to the understanding of the cause of this problem, but also enables us to formulate calculation of parameters that affect fracture of polymeric sheets during plug-assist vacuum thermoforming. The gained experimental results confirm our presented model closely.

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 that applies to thermoplastic sheets or plastic films. This forming technique is mainly used for various packaging applications such as medical devices, consumer electronics, food containers and pharmaceuticals[1-3]. The reasons of wide applications of thermoforming process are its high performance, simplicity, compactness and relatively low-cost equipment. These specifications make it possible to produce complex, large-scale configurations and free form shapes. In thermoforming, a heated plastic sheet is stretched into a mould cavity by applying pressure, and eventually it is assisted by direct mechanical loading.

Products of plug-assist vacuum thermoforming especially at high temperatures may contain two types of imperfections, fracture of polymeric sheet during deformation process and warpage. The main reason of these defects can be found in the nature of thermoforming process. Initially, it seems that these defects can be eliminated by performing of a simple control on state parameters of thermoforming process. But this is not a straight forward operation. For removal of the defects, at the first step the origins of imperfections must be identified. Then, for removing of the situations in occurrence of these defects, parameters of deformation process must be modified. In order to make these steps operational, it is necessary to provide a complete image on all of possible reasons to origins of the defects. Through a systematic reasoning, process parameters can be determined for removal of the defects[4-5].

This paper focuses on two issues, (i) origins of fracture creation, and (ii) determination of the process parameters for removal of the fracture. But mathematical calculation of parameters that affect this defect has found a little attention. The gained results based on our work not only leads to the understanding of the cause of this problem, but also enables us to work out on a method for calculating of the related parameters that affect polymeric sheets fracture during plug-assist vacuum thermoforming. These enable us to eliminate the defect during the deformation process.

Rheological Model and Description of Process

Phenomenon of polymeric sheet fracture is a practical problem on plug-assist thermoforming process for production of hollow symmetrical products. This defect can be created during both stages of plug-assist and vacuum forming.

Unfortunately, as mentioned, this problem is an overlooked area in thermoforming research. Its successful solution is directly depended on the right selection of a proper rheological model.

One of the theoretical models in this area was developed by Leonov[6]:

$$\bar{\sigma} + p\bar{\delta} = 2\bar{c}W_1 - 2\bar{c}^{-1}W_2$$

$$\bar{e}_f = 1/\theta_0 G_0(T)\exp\{-\beta\, w^s/G_0(\mathrm{T})\} \;.$$

$$[(\bar{c} - I_1\bar{\delta}/3)\, \mathrm{W}_1^{\mathrm{s}} - (\bar{c}^{-1} - I_2\bar{\delta}/3)\, \mathrm{W}_2^{\mathrm{s}}] \qquad (1)$$

$$\frac{d\bar{c}}{dt} + \bar{\omega}\bar{c} - \bar{c}\bar{\omega} - \bar{c}\left(\bar{e} - \bar{e}_f\right) - \left(\bar{e} - \bar{e}_f\right)\bar{c} = 0$$

where:

$\bar{\sigma}$ :: stress tensor,

p :: Lagrange multiplier, determined by boundary condition,

$\bar{\delta}$ :: identity tensor,

$\bar{c}$ :: Cauchy strain tensor,

$\bar{e}_f$ :: flow strain rate tensor,

$\bar{\omega}$ :: vortex tensor,

$\bar{e}$ :: strain rate tensor,

$\theta_0(T)$ :: relaxation time,

$G_0(T)$ :: tensile modulus,

W :: strain energy function $W = W(I_1, I_2)$,

β :: flexibility parameter of macromolecular chains,

I_1, I_2 :: primary and secondary strain tensor invariants, and

$2W^S = W(I_1, I_2) + W(I_2, I_1)$ :: symmetric function of W.

The last one can be shown by:

$$W_1 = \frac{\partial W}{\partial I_1},\ W_2 = \frac{\partial W}{\partial I_2}$$

$$W_1^S = \frac{\partial W^S}{\partial I_1},\ W_2^S = \frac{\partial W^S}{\partial I_2}.$$

Consider a polymeric sheet with radius (r_3) is heated for production of an axisymmetric article. It is deformed by movement of a plug with radius of r_p at constant velocity V_p, in direction of “z” axis. The implemented material is assumed to be incompressible and isotropic. The deformation process is carried out under isothermal condition. The deformed sheet could be considered a thin shell, thus the hot polymer can be modeled as a membrane. Therefore, the bending resistance of the hot sheet is ignored and the material thickness is assumed to be small in comparison to dimensions of the material. Three different stretch ratios involving in deformation process are as follows:

$$\lambda_1 = \frac{d\xi}{d\xi_0};\ \lambda_2 = \frac{r}{r_0};\ \lambda_3 = \frac{h}{h_0} \qquad (2)$$

where λ_1, λ_2 and λ_3 are the principal stretch ratios in the meridional, radial and thickness directions of the membrane, respectively. They are related together by the incompressibility condition $\lambda_1\ \lambda_2\ \lambda_3 = 1$ and ξ, ξ_0 are length of meridian in deformed and strainless sheet.

r, r_0 :: radii of deformed and strainless sheet, respectively.
h, h_0 :: thickness of the sheet after and before deformation, respectively.

Mechanical pre-stretching is a planar stretching (pure shear). Therefore, the following conditions exist:

$$\lambda_2 = 1 ; \sigma_3 = 0 ; \lambda_3 = \lambda_1^{-1} \tag{3}$$

With respect to the conditions and the tensors in Eq. (1), the following expression can be written:

$$\bar{e} = \dot{\varepsilon} \cdot \begin{pmatrix} 1 & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & -1 \end{pmatrix} ; \ \bar{\omega} = 0 ;$$

$$\bar{c} = \begin{pmatrix} c & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & c^{-1} \end{pmatrix} ;$$

$$\bar{c}^{-1} = \begin{pmatrix} c^{-1} & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & c \end{pmatrix} \tag{4}$$

where $\dot{\varepsilon}$ :: rate of deformation in longitudinal direction.

The primary and secondary invariants of tensor $\bar{c}$ are resulted from Eq. (4) as:

$$I_1 = I_2 = c + 1 + c^{-1} \tag{5}$$

By utilizing Eqs. (4) and (5), following form of Eq. (1) can be developed.

$$\bar{\sigma} + p\bar{\delta} = 0{,}5\, G_0(T) \cdot \begin{pmatrix} c - c^{-1} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & c^{-1} - c \end{pmatrix} \tag{6}$$

$$\bar{e}_f = \frac{1}{4\theta_0(T)} exp\left[-\beta\left(c + c^{-1} - 2 \right) \right] \cdot \begin{pmatrix} c - c^{-1} & 0 & 0 \\ 0 & 0 & 0 \\ 0 & 0 & c^{-1} - c \end{pmatrix} \tag{7}$$

Parameter p is resulted from condition (3): $\sigma_3 = 0$. By substituting expressions (4) and (7) into equation (1):

$$\frac{dc}{dt} = 2\left[\dot{\varepsilon} \cdot c - \left(c^2 - 1 \right) \frac{1}{4\theta_0(T)} exp\left[-\beta\left(c + c^{-1} - 2 \right) \right] \right] \tag{8}$$

This differential equation defines kinetics of elastic strain during the deformation process of viscoelastic media. The deformation rate is defined as follows:

$$\dot{\varepsilon} = \frac{d\varepsilon^H}{dt} = \frac{d \ln \lambda}{dt} = \frac{1}{\lambda} \frac{d\lambda}{dt} \tag{9}$$

where ε^H is Hencky strain.

Above equation may be represented in a dimensionless state form:

$$E(\tilde{t}) \equiv \dot{\varepsilon}(t) \cdot \theta_0(T) = \frac{\tilde{t}}{\tilde{t}^{\,2} + a^2} \tag{10}$$

Where $\tilde{t} \equiv \dfrac{t}{\theta_0(T)}$.

Finally, from Eq. 6 and condition shown by Eq. 3 following relationship can be derived.

$$\sigma_1 \equiv \sigma = G_0(T) \cdot \left(c - c^{-1} \right) \tag{11}$$

MODELING OF FRACTURE

It can be easily found out that a reason on fracture of polymeric sheets is reaching of meridional stress to its critical value. The critical level depends on the strength limit of polymer at its deformation temperature. The fracture of a polymeric sheet occurs under following conditions:

$$\sigma_1 \equiv \sigma_* = \left[\sigma(T)\right] \qquad (12)$$

where $\left[\sigma(T)\right]$:: is the strength limit of the polymer at processing temperature of T; σ_1:: meridional stress; σ_*:: critical value of stress.

Polymeric sheets during deformation process have accumulation of elastic deformation^7. The kinetics of this process can be specified based on parameters of deformation process. This accumulation can be a reason for destruction of sheet during this process. By using Eq. 11 and condition (12), following condition for destruction of sheet in plug-assist can be derived.

$$\sigma_1 \equiv \sigma_* = G_0(T)\cdot\left(c_* - c_*^{-1}\right) = \left[\sigma(T)\right] \qquad (13)$$

where σ_* and C_* are critical stress and elastic deformation in meridional direction, respectively.

The amount of critical elastic deformation can be specified easily based on equation 13.

$$c_* = \frac{1}{2}\left(\frac{\left[\sigma(T)\right]}{G_0(T)} + \sqrt{\left(\frac{\left[\sigma(T)\right]}{G_0(T)}\right)^2 + 4}\right) \qquad (14)$$

It should be mentioned that in above an assessment on variation of elastic deformation with time, $(\frac{dC}{dt})$, is presented.

During deformation of the sheet, the zone at $\tilde{r} = \tilde{r}_p$ is under high risk of destruction.

where $\tilde{r} = \frac{r}{r_3}$; $\tilde{r}_p = \frac{r_p}{r_3}$.

$\tilde{r}$, $\tilde{r}_p$: dimensionless radiuses of the sheet and plug, respectively,

r_3, r_p: the radiuses of the sheet and plug, respectively.

Exactly on this cross section area, the dimensionless parameter of (a) has its lowest value. Thus based on above equations, the amount of $\dot{\varepsilon}$ and accumulated elastic deformation are at their highest values. It is clear that no destruction on a sheet occurs while the accumulated elastic deformation on any of its cross section area to be less than critical limit.

The processing time period for plug-assist process can be obtained based on following relationship:

$$t_{Plug} = \frac{H}{V_P} \qquad (15)$$

where H is depth of the sheet, V_p is plug velocity, t_{plug} is duration time of mechanical stretching with plug.

Parameter of (a) at the critical cross section of the sheet is in following form:

$$a = r_3 \frac{\tilde{r}_p(1-\tilde{r}_p)}{V_p\theta_0(T)} \qquad (16)$$

By combining the above two equations :

$$\tilde{t}_{plug} = a\frac{H}{r_3\tilde{r}_p(1-\tilde{r}_p)} = \frac{H}{V_p\theta_0(T)}$$

where $\tilde{t}_{plug} = t_{plug} / \theta_0(T)$.

RESULTS AND DISCUSSION

Now, the amount of critical dimensionless parameters ($a = a_*$) can be obtained by using equation $(\frac{dC}{dt})$. Under this condition, the amount of elastic deformation reaches to its critical value $(\mathrm{C} = \mathrm{C}_* = \max)$, but not be more than it. In another word, under critical condition, i.e. the critical value of $a=a_*$, the elastic deformation reaches its critical amount (C_*). The amount of this parameter is obtained through solution of the differential equation of $(\frac{\mathrm{dC}}{\mathrm{dt}})$ by using various guesses for *a*. By using evaluated critical value of *a* in relationship of $a = r_3 \frac{\tilde{r}_p(1-\tilde{r}_p)}{V_p \theta_0(T)}$, critical velocity of plug $(\mathrm{V}_{*\mathrm{P}})$ can be obtained. Deformation of the polymeric sheet by a plug with this velocity results in its destruction. Thus, the condition for being safe from having fracture in a sheet is:

$$V_p < V_{*p} = \frac{r_3}{a_* \theta_0(T)} \tilde{r}_p\left(1-\tilde{r}_p\right) \qquad (18)$$

By considering the presented formula for parameter of “a”, this can be concluded that its amount can be altered not only through changing of plug velocity, but also it is possible by tuning of other process parameters. Adjustment of parameter “a” can be achieved by setting of the relaxation time parameter for implemented polymeric material. This parameter is temperature depended. Thus, suitable selection of thermal regimes, the occurrence of the polymeric sheet fracture can eliminate. Basically, the deformation process has a physical nature. The effects of this deformation on a polymeric sheet are (i) development of macromolecules mixing process, and (ii) adjustment of macromolecular structure direction with the direction of applied force field. This results in accumulation of elastic deformation within polymer. In addition, movement of polymer structure due to heating results in reduction of accumulated elastic deformation level. The rate of this process is defined by the parameter of relaxation time. Thus, accumulation of a certain amount of elastic deformation in a polymeric sheet is a result of two competitive processes. As the rate of the process of polymer structure alteration under deformation is temperature dependent, thus it’s increasing concludes to reducing of relaxation time and consequently increasing of this process diffusivitive rate. This issue reduces the level of the accumulated elastic deformation. The adequacy of Eq. (8) was verified through the comparison of experimental values in the fixed section of the deformed sheet with the theoretical results from (11).The comparison of the results for meridional stress at T=140C is presented in Fig. 1.

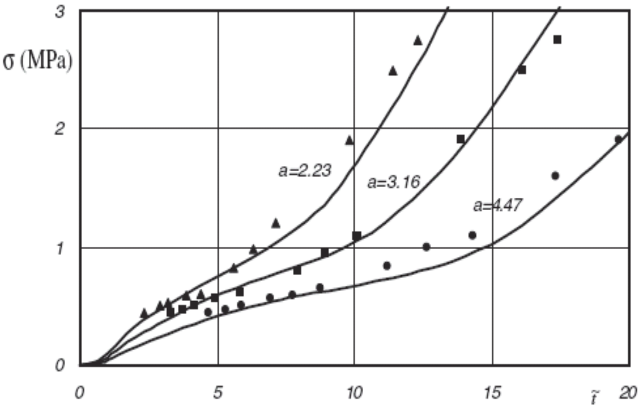

Figure 1. Stress *vs.* deformation time for ABS sheets at different plug velocities: lines- theoretical equation (11); points- experimental data.

At a certain plug velocity, an evaluation on relaxation state without occurrence of fracture phenomenon can be obtained by using Eq. (16).

$$\theta_0(T) < \frac{r_3}{a_* V_p} \tilde{r}_p\left(1-\tilde{r}_p\right) \qquad (19)$$

Theoretical and experimental information for specifying of quantitative relations between relaxation time and temperature of different polymers do not exist at present. Thus, definition of the thermal regime for polymeric sheets is only possible based on non-linear regression analysis of following equation:

$$\dot{\gamma} = \frac{1}{\theta_0(T)\cdot F(c_{12})}\cdot\frac{c_{12}}{1-c_{12}^2} \qquad (20)$$

where $\dot{\gamma} \equiv \frac{d\gamma}{dt}$ and γ is shear rate, $F(c_{12}) = \exp\left[(\beta - 7.8\sqrt{1-c_{12}^2})\left(\frac{1}{\sqrt{1-c_{12}^2}} - 1\right)\right]$, and C_{ij} is component of tensor of elastic deformation in an arbitrary coordinate.

The experimental basis for performing of such analysis is curves of the polymer flows at different temperatures. This information is obtained based on viscometer experiments. In the next step, gained relaxation times at different temperatures are compared with its critical levels, equation 19. This results in identification of a suitable thermal regime for the deformation process. The defect can be removed by specifying the regime throughout the process. Study of the obtained formula for parameter of (*a*) reveals that this parameter depends on technical-structural parameters such as the sheet and plug radiuses. The possibility of implementing these parameters on solving of the problem of sheet fracture is a theoretical issue rather than practical one, as these two parameters depends on geometrical shapes of polymeric products.

It must be mentioned that a practical solution of the sheet destruction problem during plug-assist stage can be obtained by adjusting of the parameter $\tilde{r}_p$ to its optimum value. This value can be easily obtained by means of Eq. 16 :

$$\tilde{r}_{p\,opt} = 0.5 \qquad (21)$$

Destruction of a polymeric sheet can be happened not only in plug assist stage but also in the stage of vacuum thermoforming. At the last stage, the destruction reason is same as the other stage, reaching of stress amount to its critical level. This parameter depends on the level of polymer strength in a certain temperature. In other words, the occurrence of plate destruction in this stage is same as the other stage, validating of Eq. 12.

By using Eq. 12, following equation can be derived[8]:

$$\sigma_1 = \frac{G_0(T)}{2}\left(c\lambda_1^2 - \frac{1}{c\lambda_1^2} - c^{-1}\lambda_1^{-2}\lambda_2^{-2} + c\lambda_1^2\lambda_2^2\right) - \Delta p$$

(22)

where ΔP is pressure drop.

One can conclude that the sheet destruction at vacuum stage occurs under following conditions:

$$\sigma_1 \equiv \sigma_* = \frac{1}{2}G_0(T)\cdot\left[c_*c_{*1} - (c_*c_{*1})^{-1} + c_*c_{*1}c_{*2} - (c_*c_{*1}c_{*2})^{-1}\right] = [\sigma(T)]$$

(23)

where c_{*1} and c_{*2} are the amounts of critical elastic deformation in directions of meridional and radial in vacuum stage, respectively.

By considering this fact that during destruction of polymeric sheet the following relation must be satisfied[8]:

$$cc_1(1+c_2) >> (cc_1)^{-1}(1+c_2^{-1}) \qquad (24)$$

It can be easily understood that stress-strain state of the sheet at the vacuum stage which is a non-symmetrical biaxial expansion-contraction can be assumed as the uniaxial strain with a low level of error. In other words:

$$cc_1(1+c_2) \approx c_{uniaxial} \qquad (25)$$

This provides the ability for a practical evaluation of the critical state in a polymeric sheet by implementation of relations 14 and 23. This results in following relationship:

$$[\sigma(T)] \approx [\sigma_p(T)] \qquad (26)$$

Based on Eq. 23, the following relation can be derived for accumulation of the critical values of elastic deformation at the first stage (plug-assist), and the second stage (vacuum).

$$c_* = \left(\frac{[\sigma_p(T)]}{G_0(T)} c_{*2} + \sqrt{\left(\frac{[\sigma_p(T)]}{G_0(T)} c_{*2} \right)^2 + c_{*2}(c_{*2}+1)^2} \right) \cdot [c_{*1} c_{*2} (c_{*2}+1)]^{-1} \qquad (27)$$

Based on analysis of Eq. 23 and considering Eq. 27 at the vacuum stage, a polymeric sheet destructs under following conditions:

$$cc_1(c_2+1) \geq c_* c_{*1}(c_{*2}+1) = \left(\frac{[\sigma_p(T)]}{G_0(T)} + \sqrt{\left(\frac{[\sigma_p(T)]}{G_0(T)} \right)^2 + c_{*2}\left(1+c_{*2}^{-1}\right)^2} \right) \qquad (28)$$

It is clear that at stage of vacuum, destruction of polymeric sheet occurs on a cross section surface which satisfies the following condition

$$cc_1(1+c_2) = max \qquad (29)$$

At this stage, for preventing from phenomenon of sheet destruction, the deformation process must be managed in a way that guarantees:

$$cc_1(1+c_2) = max < c_* c_{*1}(1+c_{*2}) \qquad (30)$$

It is well-known that due to speed of deformation process, there is no relaxation process at the stage of vacuum. Thus, in this stage there is accumulation of elastic. This implies that there is no possibility on management of elastic deformation. Therefore, at this stage critical conditions exist on accumulation of elastic deformation within a polymeric sheet (Eq. 29):

$$c_1 c_2 = c_{*1} c_{*2} \qquad (31)$$

Now by using Eq. (27), the critical level of elastic deformation (C_*) can be specified. This must not be misused with the critical deformation, Eq. 14, which is in support of the first stage. Since, the amount of (C_*) at the second stage must not reach to its value at the first stage.

During fracture of polymeric sheet, the order of the first term in the square root section of Eq. (28) is several times larger than the second term. Thus, based on this fact, the amount of critical deformation can be determined from following relationship:

$$c_* c_{*1}(c_{*2}+1) \approx 2 \frac{[\sigma_p(T)]}{G_0(T)} \qquad (32)$$

Two parameters of tensile modulus and strength limit for various materials at different temperatures are required for using of the last equation.

The amount of tensile modulus is obtained through rheological experiments which there are enough information on it within various references[9]. The required information on polymers strength limit at various temperatures can be found in reference[10]. In case of unavailability of this information, following theoretical method can be used:

$$\frac{[\sigma_p(T)]}{[\sigma_p(T=293K)]} \approx 1 - B \cdot \frac{T-293}{T_B-293} \qquad (33)$$

where $\sigma_P(T)$ is polymer strength limit at a specific temperature. The symbol of T_B is used for the mean softening temperature that specifies the stability of a polymer shape. Parameter of B is a dimensionless coefficient. It shows the relative variation of the polymer strength limit with respect to variation of temperature[10].

As it can be viewed, the worst destructive zone is the tinniest section of throat. Within this zone, conditions of (29) become in following form.

$$cc_1(1+c_2) \approx c \cdot 59 \cdot (1+0.25) = \max \qquad (34)$$

By substituting of relations (33) and (34) in Eq. (32), the following relationship for evaluation of critical elastic deformation amount can be derived.

$$c_* \approx 2\frac{[\sigma_p(T=293K)]}{59 \cdot (1+0.25) \cdot G_0(T)}\left(1 - B \cdot \frac{T-293}{T_B-293}\right) \qquad (35)$$

Now the thermal zone can be specified by comparison of the derived amount based on this relation with the accumulated elastic deformation at a certain cross-section area during plug-assist process in different temperatures (i.e. for various values of dimensionless parameter of $a = f[\theta_0(T)]$ and under constant plug velocities). Performing of the deformation process within this thermal zone, results in production of polymeric article without any fracture. The amount of the accumulated elastic deformation in a certain cross-section area at the end of plug-assist process is obtained by solving of differential equation $(\frac{dC}{dt})$ with various amounts for parameter of $a = f[\theta_0(T)]$.

For HIPS, $\beta \approx 2.5$, and $G_0(T = 393 \Leftrightarrow 408\,K) \approx 0.5\,MPa$. These rheological parameters are obtained through various tests by viscometer. Based on derived results, within thermal range of 398-405K, one can forecast that the produced polymeric articles can be classified in two classes of perfect or defective (with fracture). Tests are performed by using of plug-assist vacuum thermoforming equipments at different temperatures. In order to have a statistical form of destruction process, ten tests at each temperature is performed. The percent of products with fracture at each temperature is shown in Fig. 2.

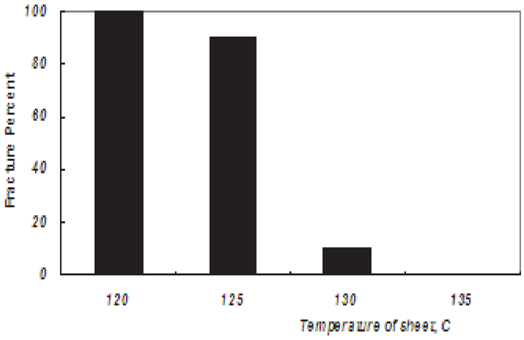

Figure 2. Fracture percent of HIPS sheets during vacuum thermoforming.

CONCLUSION

The gained experimental results confirm our presented model closely. Analysis of the results show that the occurrence of fracture in plug-assist vacuum thermoforming process is due to processing reasons as follow:

- as reduction in deformation temperature results in increasing of polymer relaxation time, and consequently there is a high level of accumulation on elastic deformation during plug-assist stage.
- increasing of plug velocity results in accumulation of elastic deformation to a high level.

REFERENCES

1. Hosseini, H., Berdyshev, B.V. and Mehrabani, A. (2009), *Polym. Eng. Sci,* **49**, 240-245.

2. Hosseini, H., Berdyshev, B.V. and Mehrabani, A. (2006), *Euro. Polym. J.*, **42,** 1836-1841.

3. Throne,J.L. (1987), *Thermoforming*, Munich, Hanser Publisher, pp.24-238.

4. Lee, J.K., Virkler, T.L. and C.E. Scott, (2001), *Polym. Eng. Sci.,* **41**, 1830-1836.

5. Ayhan, Z. and Zhang, Q.H. (2000), *Polym. Eng. Sci.* , **40,** 1-10.

6. Leonov, A.I. (1976), *Rheo..Acta*, **5**, 85-90.

7. Williams, J.G. (1970), *J. Strain Anal.*, **5**, 49.

8. Hosseini, H. (2006), Modeling of deformation processes in plug-assisted thermoforming, *PhD Thesis*, Moscow State University of Environmental Engineering, In Russian, pp.20-68.

9. Berdyshev, B.V. (1999), Methods and calculation of production of hollow polymeric articles, *PhD Thesis*, Moscow State University of Environmental Engineering, in Russian, pp.67-123.

10. Kim, V.S. (2005), *Theory and Practice of Extrusion*, Khimia, In Russian, pp.12-89.