

## Numerical analysis on foam reaction injection molding of polyurethane, Part A: Considering re-condensation of physical foam agent

HyukSu Han, Hyun Nam Nam<sup>\*</sup>, Youngkee Eun, Su Yeon Lee, Jeongho Nam<sup>\*\*</sup>, Jeong Ho Ryu<sup>\*\*\*</sup>, Sung Yoon Lee<sup>\*</sup> and Jungin Kim<sup>†</sup>

*Korea Institute of Industrial Technology, Korea Creative Industry & Engineering Center, Ansan 15588, Korea*

*\*S&WISE Co. Ltd., Seoul 08389, Korea*

*\*\*Korea Institute of Industrial Technology, Incheon 21999, Korea*

*\*\*\*Department of Materials Science and Engineering, Korea National University of Transportation, Chungju 27909, Korea*

(Received October 6, 2016)

(Revised October 14, 2016)

(Accepted October 17, 2016)

**Abstract** Foam reaction injection molding (FRIM) is a widely used process for manufacturing polyurethane foam with complex shapes. Numerical model for polyurethane foam forming reaction during FRIM process has been intensively investigated by a number of researchers to precisely predict final shapes of polyurethane foams. In this study, we have identified a problem related with a previous theoretical model for polyurethane foam forming reaction. Thus, previous theoretical model was modified based on experimental and computational results.

**Key words** Reaction injection molding, Polyurethane foam, Numerical simulation

### 1. Introduction

Foam reaction injection molding (FRIM) is a widely investigated technique for producing polyurethane materials with complicated shapes which can be used for the applications such as vehicle, home appliance, and construction etc [1-3]. In FRIM process, products are fabricated by chemical reactions among different materials injected into a mold. Raw materials in reservoirs are sent to a mixing head with high pressure and high speed. The mixed materials are injected into a mold where polymerization begins to take place. Micro bubbles are generated by chemical and physical foam agents during polymerization, and subsequently saturation reaction is occurred which leads to growth of micro bubbles.

Polyurethane foam is light, rigid and cost effective with excellent thermal and electrical insulating properties, which can be synthesized by foam forming reaction between isocyanate and polyol. Temperature and viscosity of the foam are usually increased during polymerization due to strong exothermic reaction of isocyanate and polyol. At the beginning stage of polymerization, polyol and isocyanate exist as monomers mixed with

additives such as catalysts, interfacial activators, and foam agents. Foam forming reaction is consisted of two major reactions: polymerization reaction to secure mechanical strength and saturation reaction to form cellular structure. The later can be achieved by either evaporation of physical foam agents which has low boiling point (i.e., pentane) or gas generation from chemical foam agents such as water. The former is related with exothermic reaction of isocyanate and polyol.

A variety of chemical and physical processes, such as nucleation and growth of micro bubbles, mass transfer, mold filling, and thermal flow etc., should be considered in order to understand polyurethane foam forming process. Although a number of researches have devoted to study each chemical and physical process, it is extremely difficult to consider all the process at the same time to predict FRIM process, which makes practical studies of foam forming more difficult.

In this work, based on the both of experimental and computational results for I-shape polyurethane foam forming process, we have identified problems associated with the chemical model for the evaporation of physical foam agent during FRIM process. Therefore, a modified model for the evaporation of physical foam agent during FRIM process was proposed in order to improve accuracy of simulation for polyurethane foam forming process.

---

<sup>†</sup>Corresponding author  
E-mail: [swjik@kitech.re.kr](mailto:swjik@kitech.re.kr)

## 2. Theoretical Model and Numerical Method

### 2.1. Assumptions

Foam forming reactions between water and physical blowing agent depend on the reaction rate of chemical/physical blowing. Reactions of polyol-isocyanate (gelling reaction) and water-isocyanate (blowing reaction) are assumed to be independent of each other. In addition, polyurethane foam is assumed to have homogeneous single phase. Energy and mass conservation laws combined with chemical reaction and thermodynamic equations are employed to calculate the transient density and temperature. In this model, it is assumed that the evaporation of physical blowing agent is dominated by heat, and the generation of carbon dioxide (CO<sub>2</sub>) is controlled by the reaction between water and isocyanate. In addition, secondary allophanate and biuret reactions are not individually considered, since they are already involved in the basic reactions [4-6].

The volumetric expansion of polyurethane forming can be predicted by the aforementioned theoretical equations such as thermal energy balance, gelling reaction, blowing reaction, and evaporation of physical blowing agent. It is assumed that individual chemicals are ideally mixed and the nucleation rate by chemical reaction and evaporation is sufficiently fast. In addition, density, viscosity, and thermal properties of an arbitrary cell is treated as a mixture of each chemical species and phases based on continuum model theory. It is also assumed here that the continuum is a generalized Newtonian fluid that the rheology of foam is governed by constitutive equation. In order to calculate distribution of pressure, semi-implicit method for pressure-linked equation (SIMPLE) algorithm is adopted for the numerical analysis [7]. This method can give precise solution for the fluid flow and heat transfer by chemical reaction [8]. Also, volume of fluid (VOF) method was applied to trace the free surface of foam [9]. At beginning of the reaction, pressure is uniformly distributed in the mold, and the initial velocity is set as zero at every point. Additionally, slip at the mold boundary is not considered.

### 2.2. Evaporation of Physical Foam Agent

According to Baser and Khakhar, the rate for the evaporation of physical foam agent is controlled by the heat generated from exothermic chemical reactions [4, 5]. Therefore, it is assumed that as temperature increases

over the boiling point of physical foam agent, the reaction occurs via the first order kinetics. In this case, the evaporation of physical foam agent is irreversible process which can be written as,



$$\frac{dr_{\text{BL}}}{dt} = -A_L \exp\left(-\frac{E_{\text{BL}}}{RT}\right) r_{\text{BL}} \quad (2)$$

where, BL and BG indicate physical blowing agent in the gas and liquid phases, respectively,  $A_L$  is the pre-exponential factor,  $r$  is the mass of each component per unit mass of unreacted polyol and isocyanate (i.e., polymerizing mixture), and  $E_{\text{BL}}$  is the activation energy for the evaporation of physical foam agent.

### 2.3. Governing Equations

If surface tension at interface between the foam and the air is negligible, four representative governing equations for compressible Newtonian fluid with Stokes's hypothesis can be used for numerical analysis. The continuity, momentum, energy, and mass transfer equations are listed below.

Continuity equation:

$$\nabla \cdot \mathbf{v} = -\frac{1}{\rho_F} \left[ \frac{\partial \rho_F}{\partial t} + \mathbf{v} \cdot (\nabla \rho_F) \right] \quad (3)$$

Momentum equation:

$$\begin{aligned} \frac{\partial}{\partial t}(\rho_F \mathbf{v}) + \nabla \cdot (\rho_F \mathbf{v} \mathbf{v}) = & -\nabla p + \mu_F \nabla^2 \mathbf{v} \\ & + \frac{1}{3} \mu_F \nabla (\nabla \cdot \mathbf{v}) + \rho_F \mathbf{g} \end{aligned} \quad (4)$$

Energy equation:

$$\rho_F C_F \left( \frac{\partial T}{\partial t} + \mathbf{v} \cdot (\nabla T) \right) = \nabla \cdot (k_F \nabla T) + \rho_F H_g \quad (5)$$

Mass transfer equation:

$$\frac{\partial}{\partial t}(\rho_F m_i) + \Delta(\rho_F \mathbf{v} m_i) = R_i \quad (6)$$

, where  $\rho_F$ ,  $\mathbf{v}$  and  $\mathbf{g}$  are the density of foam, the velocity vector and the acceleration of gravity.  $R_i$  represents the reaction rate between polyol and water which follows Arrhenius equation.  $k_F$  and  $H_g$  are the thermal conductivity of foam and the heat generated by chemical reaction during the polyurethane foam formation, which is equal to the right side of Equation (1).

### 2.4. Volume of Fluid (VOF) Method

As mentioned above, the VOF method is adopted to track the free surface of expanding foam. The fractional volume function,  $F(x, y, t)$ , is given by,

$$\frac{\partial F}{\partial t} + u_j \frac{\partial F}{\partial x_j} = S_F,$$

$$F(x, y, t) = \begin{cases} 1, & \text{cell filled with foam} \\ 0 < F < 1, & \text{cell at the free surface} \\ 0, & \text{empty cell} \end{cases} \quad (7)$$

In Equation (7), the numerical cell is classified as fluid cell, empty cell, and free surface cell, when  $F$  is 1, 0, and value between 0 and 1, respectively. The fractional volume function is governed by the scalar advection equation (Equation (8)).

### 3. Results and Discussion

Fig. 1 shows a configuration of FRIM process for I-shape polyurethane foam. In order to demonstrate validity of chemical reactions and theory proposed in this study, computational calculations were performed for predicting density of I-shape polyurethane foam. Density of the foam was calculated by using meshes divided by 40 (width)  $\times$  100 (height)  $\times$  9 (thickness) pieces, and the results were compared with experimentally measured densities. For brief experimental procedure, the foam was divided into small pieces with the size of 7.5 cm (width)  $\times$  6.0 cm (height)  $\times$  1.3 cm (thickness) to

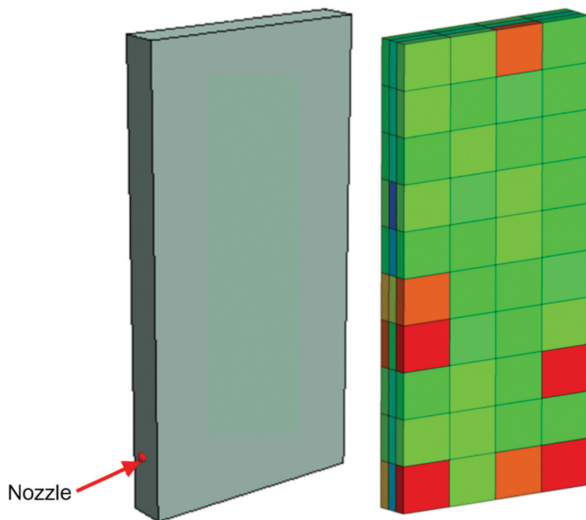


Fig. 1. Configuration of FRIM process for I-shape polyurethane foam.

measure density of the foam at different parts. It is important to note that for experimental density measurements, average density was used in order to minimize error which can be arisen due to different measurement time and method.

Fig. 2 shows the average density of foam with respect to the height. As can be seen in Fig. 2, smaller discrepancy was observed for densities between surface and core parts of the foam from the simulation results (dashed lines) compared to experimental results (solid lines). There was no significant change in density with respect to the height of mold for both of experimental and computational results. In addition, it is clearly shown in Fig. 2 that for both of experimental and computational results, foams formed at closer to the surface of mold (i.e., blue and black lines) have higher density compared to foams located at the middle part of mold (i.e., red line). For foams at the surface of mold, the experimental densities were higher than the densities obtained from numerical analysis. However, simulation predicted higher density compared to experimental density for foams at the middle parts of mold. These results might be due to the fact that for real experiments, the gas generated from the blowing agents might be able to diffuse into the outside of the mold at surfaces, which is not considered for numerical calculations. Thus, expansion of the foam might be suppressed at the surface of mold compared to the core parts, resulting in higher density of foam which formed at closer to the surface.

At the beginning stage of FRIM process, temperature

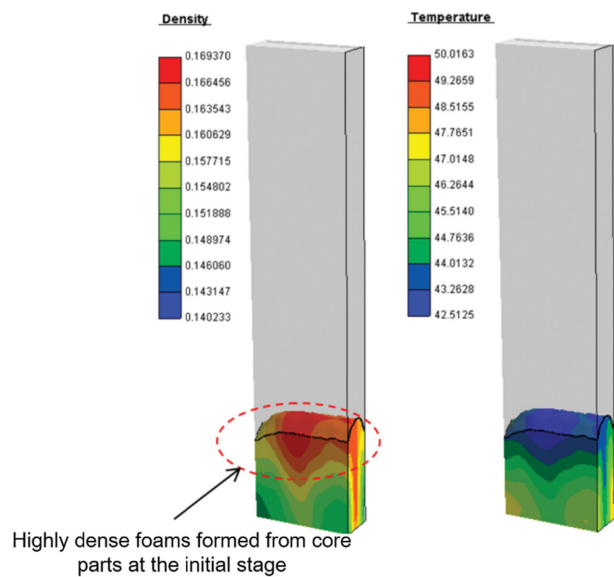


Fig. 2. Average density of polyurethane foam with respect to height of mold. Solid lines and dotted lines are experimental and computational results, respectively.

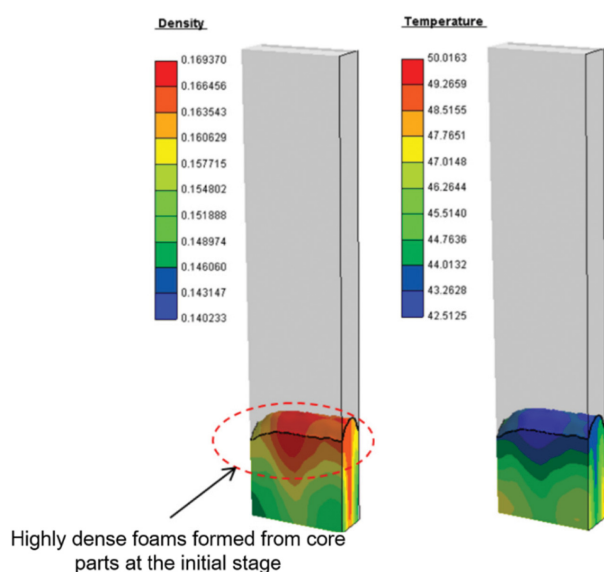


Fig. 3. Density and temperature distributions of polyurethane foam at initial stage of the reaction.

at the surface of mold is higher than that of core part, since raw materials are injected with approximately  $18^{\circ}\text{C}$  and temperature of the mold is kept at  $40^{\circ}\text{C}$ . Therefore, polymerizing mixture at the surface of mold might have lower density than materials in the core part of mold at initial stage of the reaction (Fig. 3). However, as foam forming reaction occurs, the core part of mold starts to be heated up due to exothermic reaction of chemical agents, leading to higher temperature of

core part than surface. Thus, density of foams at surface might be higher than that of foams at core part after foam forming reaction is occurred.

### 3.1. Re-condensation of Physical Foam Agent

Surface of mold can suppress flow of raw materials in the mold, resulting in velocity distribution as represented in Fig. 4. As can be seen in Fig. 4, it is clear that foams at the surface of upper part is originated from the raw materials at the core part which has low density after the reaction is occurred. Therefore, density of foams at the surface of upper part should be lower than that of foams at the surface of lower part. However, both of experimental and computational data contradict to this hypothesis indicating that there are no significant difference in densities between the surface of upper and lower parts (Fig. 2). This might be related with re-condensation of gas generated during foam forming reaction. Since  $\text{CO}_2$  gas generated by chemical foam agent has no possibility for re-condensation due to sufficiently low boiling point ( $-78.5^{\circ}\text{C}$ ), evaporated physical foam agent at initial stage of foam forming process might be responsible for the re-condensation. However, this reaction has not been considered in previous numerical methods for polyurethane foam forming reaction. Therefore, we have included phase transition of physical foam agent into polyurethane foam forming reac-

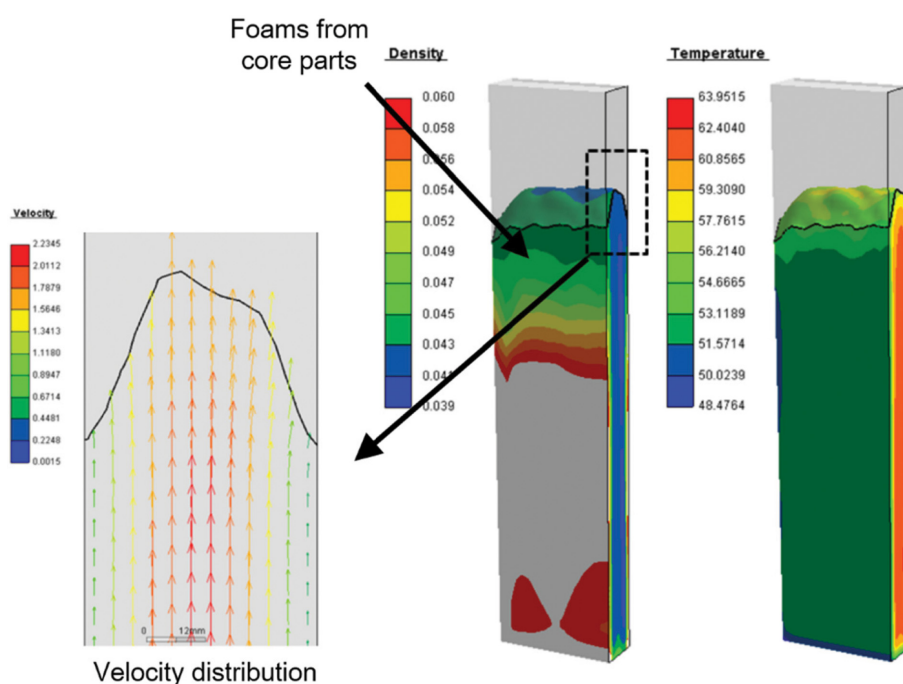


Fig. 4. Density and temperature distributions of polyurethane foam after reaction occurs for sufficient time. Left figure shows velocity distribution of foam at marked position.

tions to build new numerical methods with improved accuracy. This will be discussed further below.

### 3.2. Reversible Process for the Evaporation of Physical Foam Agent

Equilibrium constant for the evaporation of physical foam agent can be described by a temperature dependent Arrhenius equation. However, in this case, reversible phase transformation of physical foam agent can't be considered. R. Tesser *et al.* [10], have measured the weight fraction of physical foam agent (c-pentane) as a function of temperature, and the result is presented in Fig. 5. Based on the result, the evaporation of physical foam agent during FRIM process (i.e., Equation (1) and (2)) can be modified as,

$$BL \leftrightarrow BG \quad (9)$$

$$\frac{dr_{BL}}{dt} = -A_0 \exp\left(-\frac{R_0}{T}\right) \cdot \frac{dT}{dt} \quad (10)$$

where  $R_0$  is the ideal gas constant. Fig. 6 shows densities of polyurethane foam calculated by employing the modified numerical methods. For the purpose of comparison, densities calculated by using the previous model and experimental results are also presented. As can be seen in Fig. 6, density of foams at both of surface and core parts calculated by using the modified model shows much better consistency with experimental results compared to density of foams based on the previous model. To be specific, the average accuracy for predicting densities of the foam was improved from 87 % to 95 % when the modified model was applied. Furthermore, the modified model predicts that density of foams at the surface of upper part is lower than that of

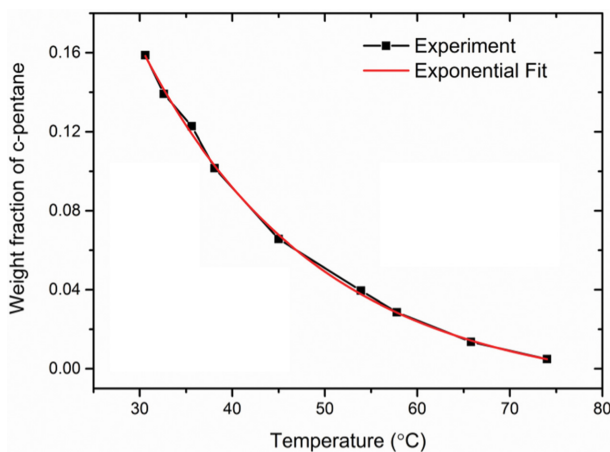


Fig. 5. Weight fraction of c-pentane as a function of temperature.

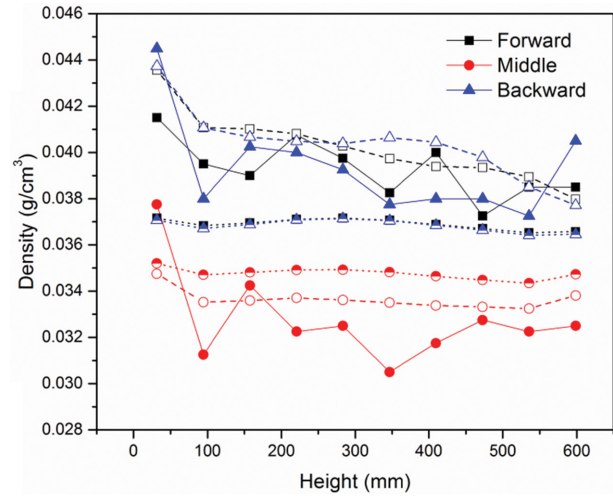


Fig. 6. Density of polyurethane foam calculated by using the modified theoretical model. Solid lines and dotted lines are experimental and computational results, respectively. For dotted lines, open and half-open circles represent results calculated by using the modified and the previous models, respectively.

foams at the surface of bottom part, which is consistent with the results from the velocity distribution (Fig. 4).

## 4. Conclusion

In polyurethane FRIM process, we have considered the evaporation of physical agent as reversible thermodynamic reaction. The theoretical model considering re-condensation of physical foam agent was developed based on experimental results previously reported. Numerical analysis was carried out to predict density and temperature distribution of polyurethane foam during FRIM process by using the simulation model developed in this study. It was verified by comparing simulation and experimental results that the modified model can predict density of polyurethane foam with better accuracy compared to the previous theoretical model.

## Acknowledgements

This research was supported by the Ministry of Trade, Industry and Energy (MOTIE), Korea, through “Upgrading the Engineering Industry Promotion” grant (No. N0000819).

## References

- [1] C. Kim and J.R. Youn, “Environmentally friendly pro-

- cessing of polyurethane foam for thermal insulation", *Polymer-Plastics Technology and Engineering* 39 (2000) 163.
- [ 2 ] M.S. Koo, K. Chung and J.R. Youn, "Reaction injection molding of polyurethane foam for improved thermal insulation", *Polymer Engineering and Science* 41 (2001) 1177.
- [ 3 ] W.H. Lee, S.W. Lee, T.J. Kang, K. Chung and J.R. Youn, "Processing of polyurethane/polystyrene hybrid foam and numerical simulation", *Fibers and Polymers* 3 (2002) 159.
- [ 4 ] S.A. Baser and D.V. Khakhar, "Modeling of the dynamics of R-11 blown polyurethane foam formation", *Polymer Engineering & Science* 34 (1994) 632.
- [ 5 ] S.A. Baser and D.V. Khakhar, "Modeling of the dynamics of water and R-11 blown polyurethane foam formation" *Polymer Engineering & Science* 34 (1994) 642.
- [ 6 ] D. Seo and J.R. Youn, "Numerical analysis on reaction injection molding of polyurethane foam by using a finite volume method", *Polymer* 46 (2005) 6482.
- [ 7 ] S.V. Patankar, "Numerical heat transfer and fluid flow", Hemisphere, Washington D.C. (1980).
- [ 8 ] S.R. Mathur and J.Y. Murthy, "A pressure-based method for unstructured meshes" *Numerical Heat Transfer Part B-Fundamentals* 31 (1997) 195.
- [ 9 ] C.W. Hirt and B.D. Nichols, "Volume of fluid (VOF) method for the dynamics of free boundaries", *Journal of Computational Physics* 39 (1981) 201.
- [10] R. Tesser, M. Di Serio, A. Scalfani and E. Santacesaria, "Modeling of polyurethane foam formation" *Journal of Applied Polymer Science* 92 (2004) 1875.