THERMOFORMING OF BIOPOLYMER-GREEN POLYETHYLENE SHEETS

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Abstract: Thermoforming is a forming technique in which petrochemical-based thermoplastics, biodegradable plastics and also plastic composites can be formed. Due to recent developments in the packaging industry, it was understood that biodegradable plastics could replace petrochemical-based engineering plastics. In this study, a biodegradable Green Polyethylene (HDPE SGF 4950, BRASKEM) was thermoformed using two different moulds. Heating temperature and time for Green PE, vacuum value, vacuum and cooling time were determined during the experimental procedure. Spherical and conical semi-finished thermoformed samples were obtained and these samples were cut in to four symmetrical pieces. Thickness measurements were achieved on some predetermined paths in quarter samples. Additionally, thickness distribution on the same paths was predicted by GEA (Geometric Element Analysis). Predicted and obtained thickness distribution results compared to each other. Finally, we have found out that GEA is not the most efficient method to obtain the thickness distribution precisely.

Keywords: biodegradable, green polyethylene, thermoforming, thickness distribution, geometric element analysis.

1. INTRODUCTION

Plastic materials are considered to be very important materials due to their extraordinary properties and performance compared to other materials such as metal and wood as shown in Azapagic et al. (2003) and Rosato (2003). Knowing that the plastic materials are of such importance in our society and these materials are a key determinant of sustainability, the replacement of petroleum-based plastics with bio-based plastics is seen as a promising alternative as plastics will reduce their dependence on fossil fuels given in Alvarez et al. (2012). Bioplastics, as a plastic material, are used in a wide variety of consumer products, such as food containers, and food packaging. Food packaging is becoming more and more important in the food industry, and functions such as convenience and portioning attract more attention. In the last decade, there has been an increasing interest in the development and application of bioplastics for food packaging. The biobased materials are generally used to packaging both the short shelf life and the long shelf life products shown in Peelman et al. (2013). The origins of the bioplastics industry date back to the mid-nineteenth century. However, academic studies on bioplastics have been concentrated in the last two decades. This kind of biodegradable materials such as Polylactic Acid (PLA), Green Polyethylene (HDPE), Polyhydroxyalkanoates (PHA) and Polyhydroxybutyrate (PHB) are known new polymers compared to classic petrochemical polymers such as polyethylene. Bioplastics can be classified into two main groups; biodegradable plastics and biobased plastics given in Cooper (2013). In recent years proteins from various sources such as whey, egg, soybean, gluten, pea, etc. and polysaccharides have been used as attractive raw materials for the production of bioplastics for a range of applications shown in Ribotta et al. (2012), Chao et al. (2013). Especially, since 2011, the market for environmentally friendly bioplastics has grown exponentially shown in Byun and Teck Kim (2012). Because of this, the use of bioplastics has an increasing trend in many industrial applications even their disadvantages such as cost, availability and strength. They are often reinforced with natural fibers to improve the weak properties. They were reinforced with flax, hemp, sisal, pineapple, cotton, oil palm fibres etc. Many researchers and scientists have made an effort on biopolymer composites and reinforcing natural fibres. Berthet et al. (2015) aimed to explore the potentialities of using lignocellulosic fibres derived from three different solid food by-products. They especially focused to understand the impact of fibre origin. Boronat et al. (2015) obtained fully biobased composite using a biobased polyethylene. They investigated the viability of a bio-composite material obtained of green polyethylene biopolymer filled with eggshell as a bio-filler. It has been reported that the use of natural fibres as reinforcing element develops other weak properties. According to the literature, it has been observed that natural fibres have developed bioplastics' some properties such as transparency, degradation in biological environment, biocompatibility, processability, mechanical and gas barrier features, crystallization and thermal stability as shown in Faruk et al. (2012), Bravo et al. (2015), Sehaqui et al. (2011), Leyva et al. (2013). Bioplastics may also be used for other engineering applications except food engineering, such as electronic and electrical housings and enclosures. In brief, bioplastics have been used in many different industrial fields such as automotive, electronics, food and beverage packaging, agricultural, textiles given in Gandini and Lacerda (2015). Song and Zheng (2008) focused on the thermo-molded bioplastics and their moisture absorption, weight loss (WL) in water and mechanical properties. Perez et al. (2016) investigated behaviour of pea protein isolate (PPI) as a potential candidate for the development of biobased plastic materials processed by injection moulding. Felix et al. (2014) carried out the injection molding as an alternative to produce biobased materials from blends. In the mentioned study viscoelastic measurements and DSC of protein/glycerol blends were used to select suitable processing conditions.

On the other hand, thermoforming is one of the most important production methods used in the packaging industry. The term "thermoforming" describes a number of related polymer processing techniques, in which thermoplastic sheets are softened by heat and subsequently formed by the application of vacuum, pressure, or a moving plug. The sheet may be stretched over a male mold or into a female mold. On contact with the mold heat is lost and the material regains stiffness as it cools. Thermoforming is one of the oldest industrial polymer processing techniques. Within packaging a large proportion of the market is concerned with the production of thin-walled polymer containers that are manufactured using industrial thermoforming processes. Thermoforming has a number of advantages and has innumerable applications and can be used by several different industries. Thermoforming of cut sheets is extensively employed in industry for the manufacture of lightweight, thin-walled products from pre-extruded plastic sheet and its largest application is in packaging shown in O'Connor et al. (2013), Sweeney et al. (2009), Nam et al. (2000). Typically, thermoformed products have uneven and undesirable wall thickness distributions as the original sheet material can locally experience very different levels of deformation to reach the extremities of the mold cavity, resulting in excessive thinning in some regions. As the depth of draw in the product increases it becomes impossible to successfully thermoform by air pressure alone and the need arises for a mechanical plug to locally capture material and draw it downward in order to create a more even, or more controlled wall thickness distribution. This process is known as Plug Assisted Thermoforming (PAT). PAT is a multistep process incorporating both a mechanical plug and air pressure and is the most common industrial thermoforming process given in McCool and Martin (2010).

In this study, a bioplastic material Green PE was thermoformed. Thermoforming parameters such as forming temperature, forming vacuum value were determined by trial and error method. Spherical and conical thermoformed samples were obtained after forming operation. Samples were cut into four pieces and thickness distribution was determined on the edges on each quarter sample. Thickness distribution on predetermined paths were predicted by GEA. Obtained and calculated thickness results were compared to each other. It results that GEA is not an appropriate method to obtain the thickness distribution precisely.

2. EXPERIMENTAL

2.1 Materials and procedures

In this study, BRASKEM SGF 4950 GREEN HDPE was used in experimental study. This grade is commonly used in producing bottles for household cleaning products and health, care products; food products; rigid containers for cosmetics and pharmaceutical applications. Green HDPE is a biodegradable polymer that has a biobased content of % 96 (ASTM D 6866). Some properties for Green HDPE are given as follows in table 1.

Table 1. Properties of BRASKEM SGF 4950 GREEN HDPE

Flow Rate (190°C/ 2.16 Kg)	Melt Flow Rate (190°C/ 21.6 Kg)	Density	Tensile Strength at Yield	Tensile Strength at Break	Flexural Modulus (1% secant)	Shore D Hardness	Notched Izod Impact Strength	Vicat Softening Temperature	Deflection Temperature Under load (0.45MPa)
D 1238	D 1238	D 1505/792	D 638	D 638	D 790	D 2240	D 256	D 1525	D 648
g/10min	g/10min	g/cm³	MPa	MPa	MPa	-	J/m	ºC	ºC
0.34	28	0.956	30	30	1350	63	150	129	75

Initially, Green PE granules were weighed using a precision scale (precision: 0.01 g) in order to form a sample in dimensions of 150 x 150 x 3 mm. Granules were placed in a sample preparation mould and heated to a proper temperature above the melting temperature. Sample preparation mould were closed to provide necessary pressure enough forming a sheet that has a thickness of 3 mm. Then the water at room temperature was passed through the cooling channels in the mould to cool the sample. There are two electric rod heaters in upper part, three electric rod heaters in lower part of the preparation mould. These heaters were adjusted as approximately 165-170° C. Heating and cooling process took about 5 minutes. Sample preparation mould and lab scale thermoforming machine used in experimental study can be seen in Figure 1.

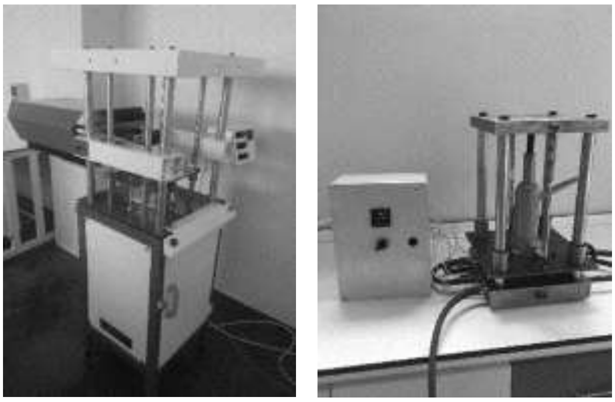
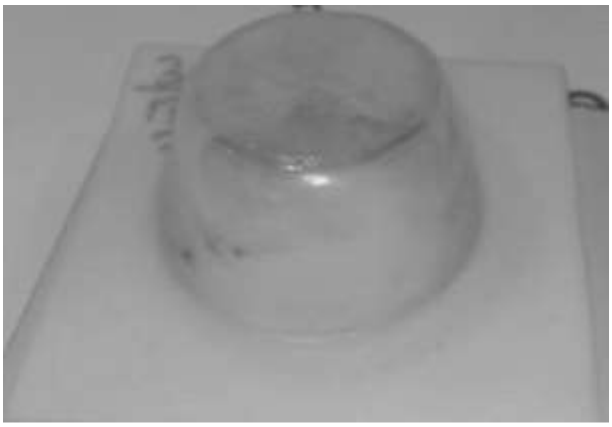


Fig.1. Lab-scale thermoforming machine and sample preparation mould respectively

At least 6 Green HDPE sheets were obtained from sample preparation mould. Half of the sheets were formed using spherical thermoforming mould. Rest of the sheets were thermoformed using conical mould. Heater temperature was chosen as 300°C. Heating time was 2.5 minutes. The quadrant of the thermoformed samples was cut and removed to make thickness measurements. Thickness values were measured using a digital caliper (precision: 0.01 mm). Thickness measurements were achieved at several points which are located from the centre of the base to the outer edge of the sample. Thicknesses were estimated for the same paths using Geometric Element Analysis (GEA). Estimated and measured thicknesses compared to each other by graphical method. Figure 2 shows thermoformed conical samples.



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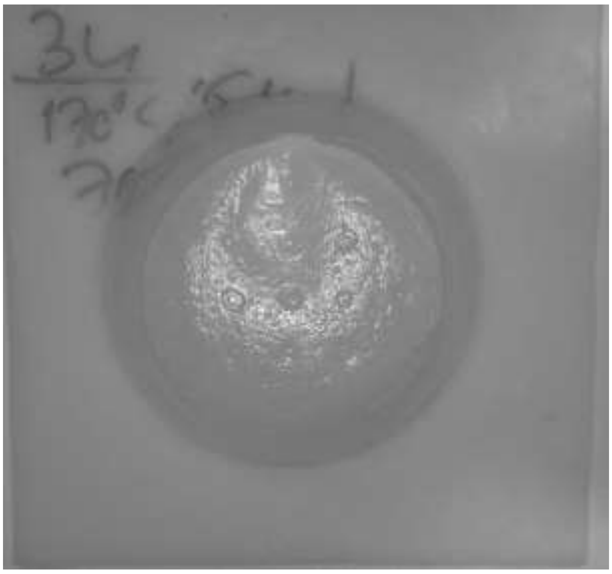


b)

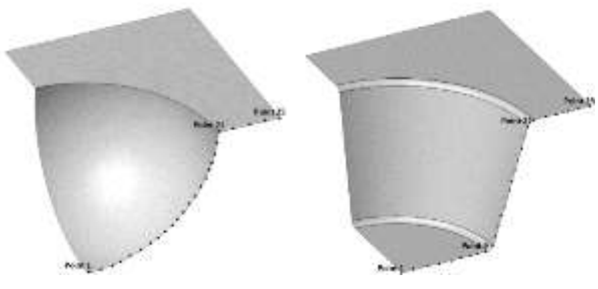
c)

Fig.2. a) Front view, b) Top view, c) Back view of a thermoformed conical sample respectively.

3. RESULTS AND DISCUSSION

Figure 3 shows dimensions for quadrant of a spherical and conical sample during thickness measuring.

Thickness was measured at 25 points in spherical thermoformed samples. However, conical samples had 28 points for thickness measuring. Thickness values were predicted by GEA for spherical and conical samples at the same points.



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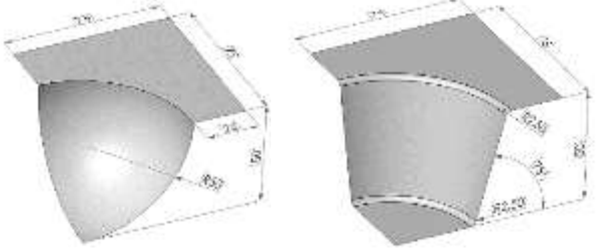


b)

c)

Fig. 3. a) Measuring points on spherical and conical samples b) Dimensions of spherical and conical thermoformed samples, c) Measuring points on a thermoformed quarter sample.

Figure 4 and 5 show the correlation between the estimated and measured thickness distributions for spherical and conical thermoformed samples respectively.

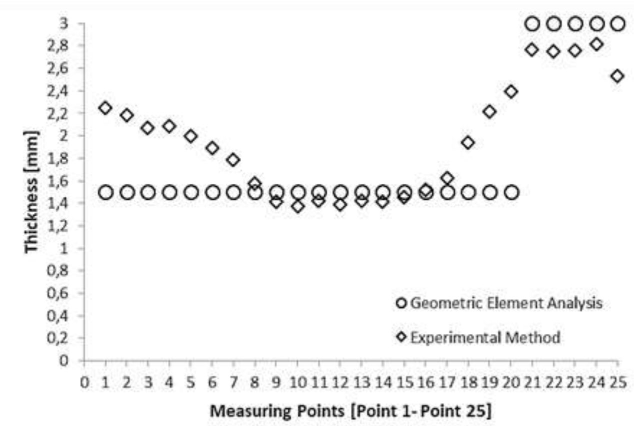


Fig. 4. Comparative thickness variation in spherical thermoformed sample

Taking into account Figure 4, from Point 1 to 7, there is a big difference between measured and calculated values. Measured and calculated thickness values are in agreement with each other from Point 8 to 17. But after Point 18, GEA has revealed different values than the measured thicknesses. One of the reasons is that GEA method does not require any material properties. It may create predictions only for simple shaped thermoforming moulds.

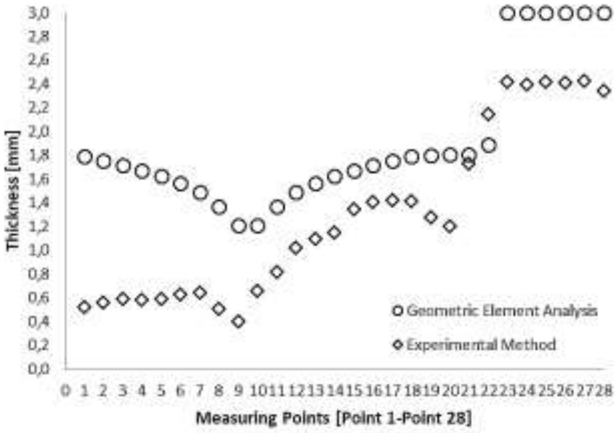
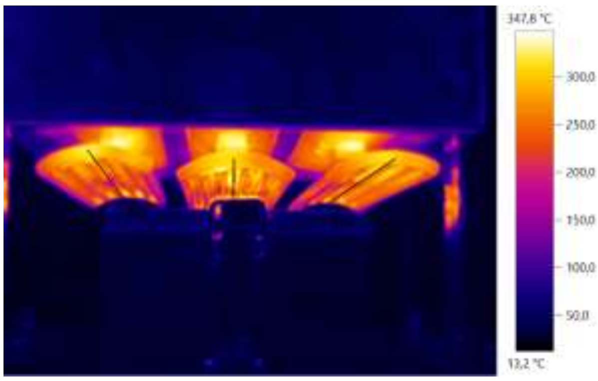


Fig. 5. Comparative thickness variation in conical thermoformed sample

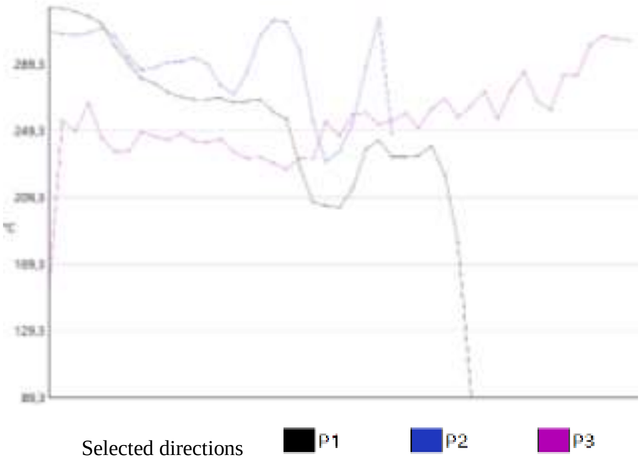
Looking at Figure 5, the first thing to notice is that all predicted values are almost greater than the measured ones. In conical sample, minimum thickness was measured as 0.39 mm at Point 9. It is said that GEA was able to predict only location, not the minimum thickness value. Compared to the spherical mould, the thickness values predicted by the GEA are closer to those measured in the conical mould.

The disadvantage is that the GEA method does not produce results according to the mechanical and flow properties of the plastic material. In addition to this, there are many reasons for non-uniform thickness distribution in thermoformed samples. Some of these reasons are caused by machinery. The most common problem is that the temperature changes over a heater which was adjusted to a proper temperature. Using a thermal imaging camera (Testo) some thermal images captured on heaters which was adjusted as 300° C as seen in Figure 6.

Temperature distribution is one of the leading parameters that affect the thickness distribution in thermoformed products. Providing unifom temperature distribution on heated sheet, facilitates controlling the thickness variation in the product. Because of this, more uniform the temperature on the heated sheet, more uniform the thickness variation after forming.



a)



b)

Fig.6. a) Thermal image of heaters at 300 ^{0}C,
b) Temperature variation along selected directions P1, P2 and P3 respectively

As can be clearly seen from the analysed thermal images, heaters could be considered as one of the main reasons for non- isothermal heating. Heating temperature was adjusted as 300° C but the temperature was above and below 300° C at many points on heaters, as seen in Figure 6. Three directions as P1, P2 and P3 were selected on each heater using a software from TESTO. Temperature variation along these directions have been investigated. It is said that heaters are not efficient and precise on heating on that lab-scale thermoforming machine. Also, this study is based on the results which was obtained from Scientific Research Projects (KLUBAP 55 and KLUBAP 68). Only GEA method was used for thickness prediction. Since it is necessary to produce extra samples to use new prediction methods, estimation methods such as Finite Element Analysis, Adaptive Network Based Fuzzy Inference System (ANFIS) and Image Processing can only be used in future studies.

4. CONCLUSIONS

Biopolymers are indispensable elements in packaging industry. The cost and availability are some disadvantages of biopolymers. In the near future, a large portion of the raw materials used in the packaging industry will be covered by biodegradable plastics. In this study, a biodegradable biopolymer BRASKEM SGF 4950 GREEN HDPE samples were produced and thermoformed. GEA prediction method was used for obtaining thickness distribution. It results that GEA is not an accurate method to estimate the thickness distribution. Thermoformable biopolymer composite production, using organic fibers will be investigated for future studies. Infrared heating may be applied for heating of biopolymer sheets additionally, number of original heaters may be increased and shape of them may be changed and more uniform heating could be provided as propose. Additionally, Finite Element Analysis, Image Processing and Adaptive Network Based Fuzzy Inference System (ANFIS) will be used as alternative methods for thickness prediction.

5. ACKNOWLEDGEMENTS

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6. REFERENCES

1. Azapagic, A., Emsley, A. and Hamerton, I., (2003). *Polymers: The Environmental and Sustainable Development*, pp. 219, John Wiley and Sons, England.
2. Rosato, D. V. and Rosato D.V., (2003). *Plastics Engineered Product Design*, pp. 1-2, 20-22, Elsevier, USA.
3. Alvarez, C.R., Edwards, S., Moure-Eraso, R. and Geiser, K., (2012). *Sustainability of bio-based plastics: general comparative analysis and recommendations for improvement,* Journal of Cleaner Production, 23, 47-56.
4. Peelman, N., Ragaert, P., Meulenaer, B.D., Adons, D., Peeters, R., Cardon, L., Impe, F.V. and Devlieghere, F., (2013). *Application of bioplastics for food packaging,* Trends in Food Science & Technology, 32, 128-141.
5. Cooper, T.A., (2013). *Developments in bioplastic materials for packaging food, beverages and other fast-moving consumer goods,* Trends in Packaging of Food, Beverages and Other Fast-Moving Consumer Goods (FMCG), Neil Farmer (Ed.), pp. 108-152, Woodhead Publishing Limited, Philadelphia.
6. Ribotta, P. D., Colombo, A. and Rosell, C.M., (2012). *Enzymatic modifications of pea protein and its application in protein cassava and corn starch gels*, Food Hydrocoll, 27, 185–190.
7. Chao, D., He, R., Jung, S. and Aluko, R.E., (2013). *Effect of pressure or temperature pretreatment of isolated pea protein on properties of the enzymatic hydrolysates.* Food Res. Int., 54, 1528–1534.
8. Byun, Y. and Teck Kim, Y., (2012). *Innovations in Food Packaging*, Jung Han (Ed.) 2nd ed, pp. 369-390, Elsevier, London, UK.
9. Berthet, M.A., Coussy, H.A., Machado, D., Hilliou, L., Staebler, A., Vicente, A. and Gontard, N., (2015). *Exploring the potentialities of using lignocellulosic fibres derived from three food by-products as constituents of biocomposites for food packaging*, Industrial Crops and Products, 69, 110–122.

10. Boronat, T., Fombuena, V., Sanoguera, D.G., Nacher, L.S. and Balart, R., (2015). *Development of a biocomposite based on green polyethylene biopolymer and eggshell*, Materials and Design, 68, 177–185.
11. Faruk, O., Bledzki, A.K., Fink, H.P. and Sain, M., (2012). *Biocomposites reinforced with natural fibers: 2000–2010*, Progress in Polymer Science, 37, 1552–1596.
12. Bravo, A., Toubal, L., Koffi, D. and Erchiqui, F., (2015). *Development of novel green and biocomposite materials: Tensile and flexural properties and damage analysis using acoustic emission*, Materials and Design, 66, 16–28.
13. Sehaqui, H., Allais, M., Zhou, Q. and Berglund, L.A., (2011). *Wood cellulose biocomposites with fibrous structures at micro- and nanoscale*, Composites Science and Technology, 71, 382–387.
14. Leyva, B.M., Silva, G.G. D., Gastaldia, E., Chávez, P.T., Gontarda, N. and Coussy, H.A., (2013). *Biocomposites from wheat proteins and fibers: Structure/mechanical properties relationships*, Industrial Crops and Products, 43, 545– 555.
15. Gandini, A. and Lacerda, T.M., (2015). *From monomers to polymers from renewable resources: Recent advances*, Progress in Polymer Science, 48, 1–39.
16. Song, Y. and Zheng, Q., (2008). *Preparation and properties of thermo-molded bioplastics of glutenin-rich fraction*, Journal of Cereal Science, 48, 77-82.
17. Perez, V., Felix, M., Romero, A. and Guerrero A., (2016). *Characterization of pea protein-based bioplastics processed by injection moulding*, Food and Bioproducts Processing, 97, 100-108.
18. Felix, M., Alfonso, J. E.M., Romero, A. and Guerrero, A., (2014). *Development of albumen/soy biobased plastic materials processed by injection molding*, Journal of Food Engineering, 125, 7-16.
19. O'Connor, C. P.J., Martin, P.J., Sweeney, J., Menary, G., Rose, P.C. and Spencer, P.E., (2013). *Simulation of the plug-assisted thermoforming of polypropylene using a large strain thermally coupled constitutive model*, Journal of Materials Processing Technology, 213, 1588-1600.
20. Sweeney, J., Spares, R. and Woodhead, M., (2009). *A Constitutive Model for Large Multiaxial Deformations of Solid Polypropylene at High Temperature*, Polymer Engineering and Science, 49, 1902-1908.
21. Nam, G.J., Ahn, K.H. and Lee, J.W., (2000). *Three Dimensional Simulation of Thermoforming Process and Its Comparison With Experiments*, Polymer Engineering and Science, 40(10), 2232-2240.
22. McCool, R. and Martin, P.J., (2010). *The Role of Process Parameters in Determining Wall Thickness Distribution in Plug-Assisted Thermoforming*, Polymer Engineering and Science, 50, 1923-1934.

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