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Optimization of Injection Molding Parameters for Poly(Styrene-Isobutylene-Styrene) Block Copolymer

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Abstract. Poly(styrene-isobutylene-styrene) (SIBS) is a widely used thermoplastic elastomer in bioimplantable devices due to its inherent stability in vivo. However, the properties of the material are highly dependent on the fabrication conditions, molecular weight, and styrene content. An optimization method for injection molding is herein proposed which can be applied to varying SIBS formulations in order to maximize ultimate tensile strength, which is critical to certain load-bearing implantable applications. The number of injection molded samples required to ascertain the optimum conditions for maximum ultimate tensile strength is limited in order to minimize experimental time and effort. Injection molding parameters including nozzle temperature (three levels: 218, 246, and 274 °C), mold temperature (three levels: 50, 85, and 120 °C), injection speed (three levels: slow, medium and fast) and holding pressure time (three levels: 2, 6, and 10 seconds) were varied to fabricate dumbbell specimens for tensile testing. A three-level L9 Taguchi method utilizing orthogonal arrays was used in order to rank the importance of the different injection molding parameters and to find an optimal parameter setting to maximize the ultimate tensile strength of the thermoplastic elastomer. Based on the Taguchi design results, a Response Surface Methodology (RSM) was applied in order to build a model to predict the tensile strength of the material at different injection parameters. Finally, the model was optimized to find the injection molding parameters providing maximum ultimate tensile strength. Subsequently, the theoretically-optimum injection molding parameters were used to fabricate additional dumbbell specimens. The experimentally-determined ultimate tensile strength of these samples was found to be in close agreement (1.2%) with the theoretical results, successfully demonstrating the suitability of the Taguchi Method and RSM for optimizing injection molding parameters of SIBS.

Keywords: Taguchi Method, Response Surface Methodology, SIBS, Injection Molding Optimization.
PACS: 81.05.Lg

INTRODUCTION

Poly(Styrene-block-Isobutylene-block-Styrene) (SIBS) is a thermoplastic elastomer that has gained attention recently due to its high degree of biocompatibility [1-5]. This linear block copolymer has a triblock structure formed by a polyisobutylene (PIB) core sandwiched between blocks of polystyrene (PS). The formulation of SIBS can be tailored for different applications by changing the weight percentage of PS or by changing the molecular weight of the polymer chains. The hard PS blocks provide SIBS with a glassy microstructure that enhances mechanical strength and rigidity of the material, while the PIB has a soft microstructure with increased chain mobility that gives the polymer its elastomeric properties [4-8]. The possibility of tailoring mechanical properties, together with the high degree of biocompatibility, make SIBS an ideal material for use in biomedical devices. SIBS was approved for ultra-long term vascular device applications by the FDA and is currently used in the TAXUS® drug-eluting coronary stent [1, 4]. However, some studies have outlined the deficiencies of this material as prone to surface cracking and susceptible to viscoelastic creep [4, 9]. Therefore, it is extremely important to fully understand the mechanical properties of the material in order to take advantage of its excellent biocompatibility.

Block copolymers of similar composition might have diverse mechanical properties due to their composite nature. Parameters such as molecular weight, block weight percentage, and polymer chain structure are known to give rise to different microstructures that in turn lead to different material properties. Different grades of SIBS can have very different morphologies based on the ratio of hard phase to soft phase. At lower contents of PS, the hard phase forms spherical domains through the soft matrix. As the PS content increases, the spherical domains become double gyroid structures and as PS content is further increased, the structure of the hard phase becomes lamellar [7]. It is likely that the incompatibility of the soft and hard phases leads to micro phase separations and results in the different morphologies described [2]. It is well known that for composite systems, the interface between different phases play

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a major role in the performance of the material: A weakened interface might lead to premature cracking and failure. Additionally, the method of fabrication for SIBS might play a very important role due to the incompatibility of the different phases. Therefore, different methods may result in different qualities of the interface. For example, a linear SIBS with 30% PS by weight manufactured by KANEKA Corporation has a published ultimate tensile strength of 18 MPa, whereas in a study by Lim et al. [10], it is reported that the ultimate tensile strength of the same SIBS grade was measured experimentally at 12.7 MPa. It is likely that these differences are due to different fabrication processes.

One of the most common fabrication processes for thermoplastics is plastic injection molding (PIM). Although this fabrication process is quite complex, it has many advantages that make it an ideal candidate for fabrication of thermoplastics. One of the disadvantages of this manufacturing technique is the complex dependence of product quality on the multiple process parameters that drive PIM. Some of the process parameters are temperature of the injection nozzle, temperature of the mold, injection rates, and packing time of plastic inside the mold [11, 12]. The amount of process variables, together with the coupled relations among them, makes it a challenging problem. To be able to analyze such a complex system, it is necessary to make use of engineering optimization techniques and employ design of experiments to understand the effect that each parameter has on the outcome of PIM [13-20]. Altan utilized the Taguchi method, ANOVA, and an artificial neural network (ANN) approach in order to minimize shrinkage of polypropylene and polystyrene. Shrinkage was measured experimentally and the parameters that produced minimal shrinkage were found [21]. The use of a Response Surface Methodology (RSM) and genetic algorithms coupled with ANN in order to optimize PIM parameters for maximum strength and impact resistance was proposed by Tzeng et al. These methods proved to be effective tools for predicting and optimizing mechanical properties of a polycarbonate composite [22].

The tools for optimization of PIM parameters are readily available in the extensive literature and have been thoroughly studied for thermoplastics. However, not many of these studies have focused on thermoplastic elastomer block copolymers. The nature of these types of materials makes it a challenging issue that has yet to be adequately addressed in the open literature. In this work, a 30% PS by weight SIBS was studied by means of Taguchi orthogonal arrays and RSM in order to maximize ultimate tensile strength through PIM process parameters. Selected parameters for this study were nozzle temperature (T_{melt}), mold temperature (T_m), injection rate (Sp), and packing time (t). Predictions were validated experimentally in order to assess the accuracy of the methodology.

EXPERIMENTAL

Pellets of a 30% PS and a molecular weight of 132,000 g/mol (commercial name SIBSTAR 103T) were obtained free of charge from KANEKA Corporation. Pellets were injected using a bench top injection molder. Specimens were fabricated in a mold that complies with ASTM Standard D638 Type V. A linear actuator and a variable speed controller were retrofitted to the injection molding apparatus in order to precisely control injection speed. Before testing the samples, the injection molder was flushed to achieve steady state conditions and minimize process variability. Each parameter level consisted of the average of eight specimens in order to reduce measurement variation. All testing was performed using an Instron 5966 Tensile Testing Machine at a rate of 500 mm/min.

Taguchi Method

Taguchi method is an established and accepted technique used to analyze and optimize different processes [23]. Through the use of orthogonal arrays, it is possible to quantify effects of different parameters and predict outcomes without running costly, time-consuming experiments. This is achieved by using a specific Signal to Noise function (S/N) to characterize experimental quality. In this experiment, the selected function was “larger is better”:

$$\frac{S}{N} = -10\log_{10}\left[\frac{1}{n}\sum_{i=1}^{n}\frac{1}{y_i^2}\right] \quad (1)$$

Where y_i is the measured response of the i^{th} measurement and n is the number of measurements. The major drawback of this method is that it does not predict continuous variables and only works with categorical factors. As a result, this method is used for experimental setups in which factors are discrete or in order to reduce experimental complexity. In this work, an L_9 orthogonal array was used to analyze the four variables at three distinct levels.

Response Surface Methodology

RSM is a technique in which inputs and outputs are related through a second order polynomial [11]. By making use of statistical techniques to represent relationships between parameters and outputs, it is possible to predict outcomes of responses. This technique also considers the interaction between parameters, which is a key component of the PIM process. RSM is modeled through the following equation:

$$\eta = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{i=1}^{k} \beta_{ii} x_i^2 + \sum_{\substack{i=1 \\ i<j}}^{k} \sum_{j=1}^{k} \beta_{ij} x_i x_j \quad (2)$$

Where η is the response of a set of k design parameters $x_{1,} x_{2,} \ldots, x_{k,}$ and β indicates fitting coefficients. All analyses were performed using Minitab software.

RESULTS AND DISCUSSION

Parameter levels used for this study were: melt temperature of 218 °C, 246 °C, and 274 °C; mold temperature of 50 °C, 85 °C, and 120 °C; injection rate of 45 mm^3/s (1), 100 mm^3/s (2), and 120 mm^3/s (3); and packing time of 2 seconds, 6 seconds, and 10 seconds. These levels were based on previous experience with this material and suggestions made by KANEKA Corporation. The injection rate of 45 mm^3/s was the slowest injection rate possible that did not produce an incomplete mold. Table 1 shows the Signal to Noise ratios for the different samples tested and the different levels used for each sample.

TABLE 1. S/N ratios for SIBS 103T

T_{melt} (°C)	T_m (°C)	Sp	t (s)	S/N	σ_{uts} (MPa)
218	50	1	2	24.32	16.5
218	85	2	6	23.63	15.3
218	120	3	10	23.60	15.1
246	50	2	10	24.08	16.0
246	85	3	2	22.66	13.6
246	120	1	6	24.00	15.9
274	50	3	6	22.69	13.7
274	85	1	10	24.37	16.6
274	120	2	2	22.45	13.3

Table 2 shows the ranks for each of the tested parameters. As can be seen from the table, injection speed and holding time have the largest effect on the ultimate tensile strength of SIBS, while mold and nozzle temperature have the smallest effect.

TABLE 2. Ranks of injection molding parameters for SIBS 103T

Level	T_{melt} (°C)	Tm (°C)	Sp	t (s)
1	15.64	15.41	16.33	14.48
2	15.18	15.16	14.85	14.94
3	14.51	14.76	14.15	15.92
Delta	1.13	0.65	2.17	1.44
Rank	***3***	***4***	***1***	***2***

Figure 1 shows the curves for the main effects of each parameter level on the ultimate tensile strength of SIBS. Curves for packing time and injection speed are steeper than the nozzle temperature and mold temperature, which is an indication of the higher sensitivity of tensile strength to injection rate and packing times. Furthermore, in Figure 2 the levels that would produce the highest tensile strength as predicted by Taguchi method are circled. The predictions for Signal to Noise ratio and ultimate tensile strength are 25.19 and 17.96 MPa, respectively.

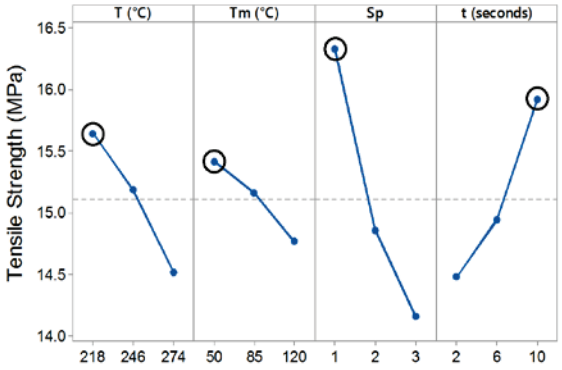

FIGURE 1. Taguchi main effects of injection molding parameters on the tensile strength of SIBS 103T

As shown in Table 2, mold temperature has the least effect on tensile strength. The standard deviation within samples at constant levels was 0.65 MPa on average, which is the same delta value for this parameter. This suggests that the mold temperature does not have a major effect on the ultimate tensile strength and that this deviation may be a result of variation during testing. Hence, mold temperature was removed from the RSM methodology in order to reduce experimental cost. A constant mold temperature of 85 °C was therefore used for all samples tested for RSM.

A Box-Behnken design was used for the RSM. Factors were spaced equally over the experimental domain and a total of 120 specimens were fabricated and tested in order to fit the second order polynomial shown in Eq. 2. Main effects of tested samples are shown in Figure 2. It can be seen that injection rate and holding time have the largest influence on tensile strength, as expected. Lower nozzle temperatures seem to be less optimal for tensile strength compared to results from Figure 1. This could be due to a coupled effect from mold temperature and nozzle temperature for which Taguchi method does not fully account.

Interaction plots are shown in Figure 3. As expected, curves for different injection rates are further apart, with slower rates at higher tensile strengths. The same applies for curves with different packing times, although the spread is not as large.

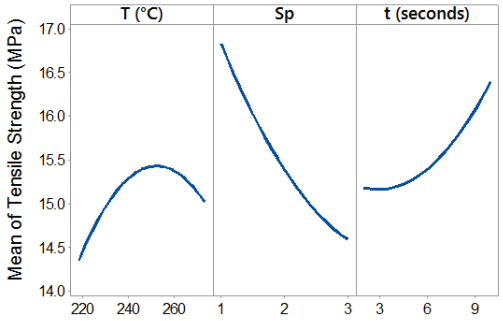

FIGURE 2. RSM main effects of injection molding parameters on the tensile strength of SIBS 103T

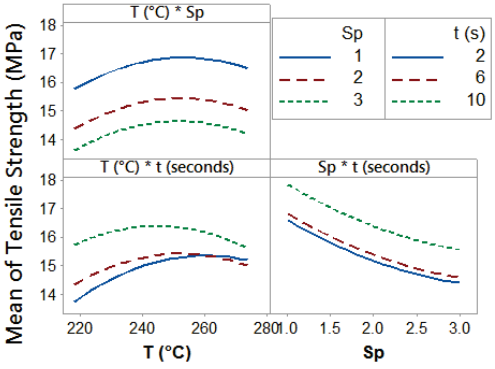

FIGURE 3. Interaction plots for different parameters for SIBS 103T

Using the built-in optimization function from Minitab for RSM, it is possible to search for the parameters that would produce the highest tensile strength. This method allows for locating maxima and minima that lie between levels, as it incorporates continuous instead of discrete variables. Ultimate tensile strength from the optimization function prediction was 17.9 MPa. Optimal parameter levels that resulted from the analysis were a nozzle temperature of 245 °C, 10 seconds packing time, and an injection rate of 45 mm^3/s. Experimental validation was carried out with a group of eight samples for testing. The resulting tensile strength for the optimized samples was 17.7 MPa. This value lies within the 95% confidence interval of the prediction by the RSM methodology. Furthermore, this result is an improvement of almost 40% compared to the reported value by Lim et al. [10].

CONCLUSION

In this study, the injection molding parameters of a biocompatible thermoplastic elastomer were analyzed and optimized to maximize ultimate tensile strength. The triblock copolymer used has a molecular weight of 132,000 g/mol and is composed of 30% polystyrene and 70% polyisobutylene by weight. Selected parameters for this study were nozzle temperature, mold temperature, injection rate, and packing time. A Taguchi orthogonal array was used to determine the rank of importance for the selected parameters. It was found that injection rate and packing time were the dominant parameters for tensile strength, while mold temperature had little effect. Consequently, a response surface methodology was performed for the three most prominent parameters while mold temperature was maintained constant at 85 °C. The RSM model was then optimized to find maximum tensile strength. Using the optimized parameters, validation samples were fabricated and tested in order to compare experimental results with theoretical results. Values are in close agreement, with an error of less than 1.2%. These results reinforce the usefulness of parameter optimization for tailoring mechanical properties in thermoplastic elastomers.

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