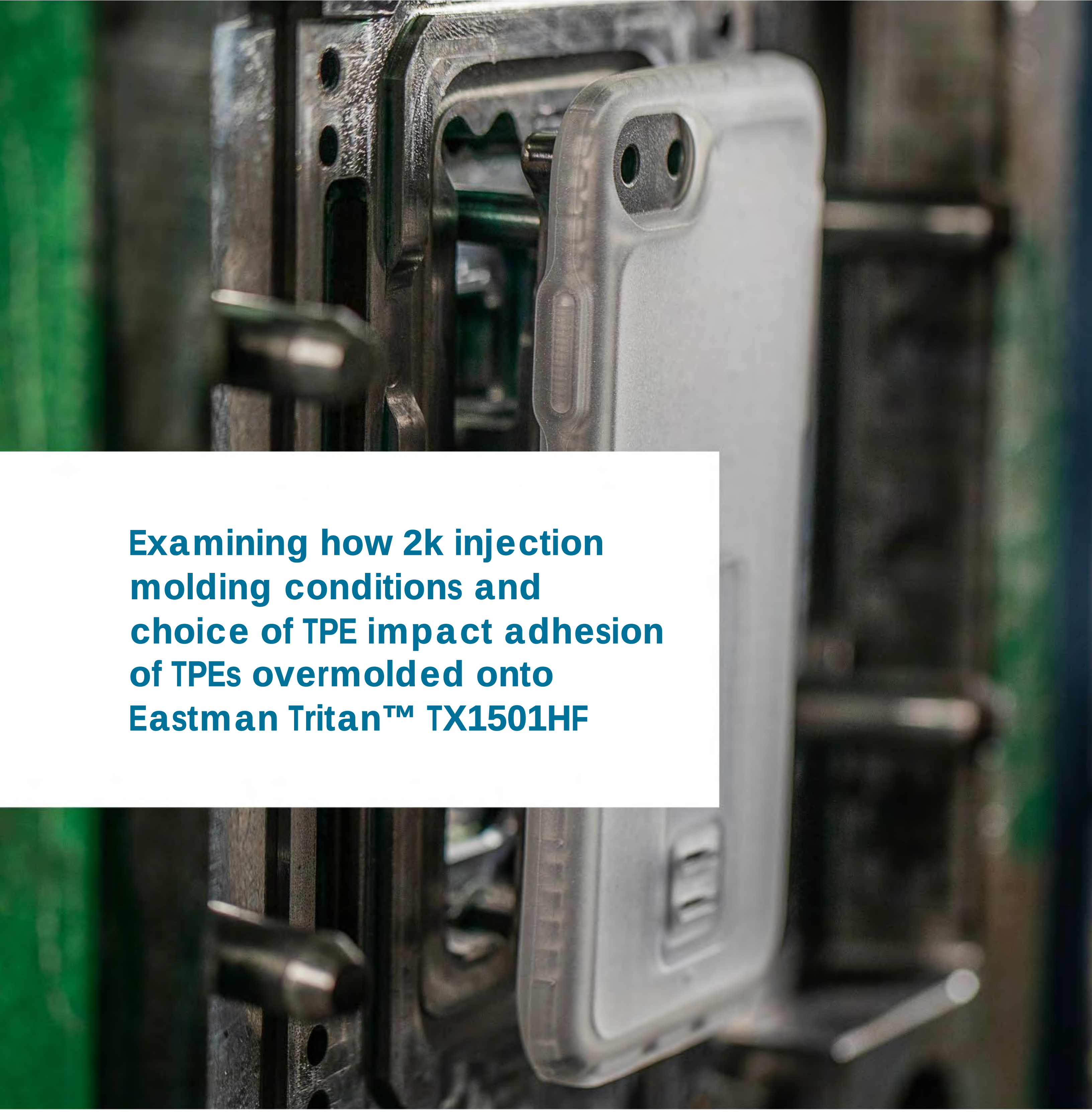

Examining how 2k injection molding conditions and choice of TPE impact adhesion of TPEs overmolded onto Eastman Tritan™ TX1501HF

Authors

Eastman: Stijn De Kimpe, Andreas Derweduwen, Mike Morrow, Doug Carico, Robbie Meul, Merryann Ledbetter, Emmett O'Brien, Dayton P. Street

Lubrizol: Mike Ramsay

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Abstract

The purpose of this work was to examine how the adhesion strength of thermoplastic elastomers (TPEs) overmolded onto Eastman Tritan™ TX1501HF, and Renew versions thereof, is impacted by changing specific processing variables of the examined 2k overmolding process. Holding the TPE constant, the first part of this study examined three easy-to-change variables (Mold Temperature, Pack and Hold Time after injection of the TPE, and Injection Speed of the TPE) and one hard-to-change variable (TPE Barrel Temperature). Utilizing a Split-Plot DoE, three variables (Mold Temperature, Pack and Hold Time, and TPE Barrel Temperature) were determined to be statistically significant. Moreover, by optimizing these conditions, the performance of the adhesion strength to TX1501HF could be maximized to an average adhesion strength value of ~294 N (minimum average adhesion strength values of ~240 N were observed for non-ideal conditions). Moreover, using these optimized processing conditions two commercially available grades, ESTANE® ECO TPU 12T80E and 16T85 from Lubrizol, showed even higher adhesion strength values (> 310 N) compared to the initial TPE used in the DoE and other screened TPEs utilizing similar optimized processing conditions. (Optimized according to the DoE conclusions and within the advised processing windows of the individual TPE.) These results highlight that both the processing conditions utilized during 2k overmolding and the choice of TPE have a significant impact on the adhesion strength between the rigid thermoplastics and thermoplastic elastomers.

Introduction

Injection molding is one of the most common and versatile fabrication techniques for generating polymeric components.[1,2] Over the last few decades, innovations to the traditional injection molding process have resulted in new processes, such as, in-mold labeling (IML), fusible core injection molding, and overmolding.[1,2] These new processes, which can result in improved part and material performance, have enabled engineers to push the known boundaries of material-property space resulting in new and complex product architectures.[1,2]

Of these processes, overmolding has seen utilization in myriad markets, such as automotive, consumer electronics, medical and packaging, due to its inherent ability to cohesively join two polymeric materials together.[1,3] Although the materials selected for an overmolding process will depend on the end-use application, in most cases, a soft thermoplastic elastomer (TPE) is molded onto a rigid thermoplastic polymer. Once the two materials are overmolded, adequate bonding between the hard and soft substrate is required to maintain performance in the end-use application. The adhesion strength of overmolded parts is dependent on both the mechanical and chemical adhesion between the two materials. While numerous theories describe interfacial interactions between two materials, in general, chemical adhesion is mainly impacted by three main variables: wetting of the substrate, interdiffusion of polymer chains, and solidification of melted material within the interphase of the interface.[1,4] By maximizing wetting of the substrate, interdiffusion of polymer chains across material interfaces, and ensuring solidification within the interphase one can achieve the best bond strengths between two materials. On the other hand, chemically incompatible materials will display low adhesion strengths. In either case, mechanical interlocking can be utilized to enhance the bond strength between two materials.[5] In this method, the rigid component will be designed to contain features and/or cavities in which the soft material can fill during injection molding that, upon solidification, will physically interlock the two materials together. Aside from tailoring the chemical and mechanical bonding, less information exists regarding how the processing conditions impact adhesion of overmolded solutions.[1] In situations in which mechanical interlocks are not an option and the materials being overmolded cannot be changed, the processing conditions are one of the final “levers” that can be manipulated to impact bond strength.

In the first part of this study, both the substrate (Eastman Tritan™ TX1501HF) and TPE (not identified) were held constant, while four processing conditions for the TPE (Mold Temperature, Barrel Temperature of the TPE, Pack and Hold Time after TPE injection, and Injection Speed of the TPE) were varied and the impact on peel strength was examined. The results from this study suggest that when the processing conditions are optimized for adhesion performance that the adhesion strength can be enhanced 125% compared to the adhesion strength observed when the least desirable processing conditions are utilized. The second part of the study examined how translatable the learnings identified from the DoE were for additional TPEs. The findings from this effort suggest that the processing conditions should follow the guidance established in the DoE (maximize Mold Temperature, TPE Barrel Temperature, and Pack and Hold Time) while staying within the TPE's suggested processing conditions. Once conditions were optimized for each TPE, a broad range of peel strength values were observed. Specifically, some TPEs exhibited lower adhesion strength values compared to the adhesion strength values observed in the DoE, while others – specifically two commercially available grades, ESTANE® ECO 12T80E and 16T85 from Lubrizol – displayed even higher adhesion strength values (> 310 N) compared to the adhesion strength values observed in the DoE. These results underscore the importance of chemical compatibility between the TPE and substrate.

Materials and methods

Materials and preparation of samples

Both the thermoplastic substrate (Tritan™ TX1501HF) and the TPEs were dried according to their respective Technical Data Sheets (TDS). In this work, overmolded samples were generated via a 2k injection molding process using an Engel E Victory 220, which is a Dual Barrel Molding Machine equipped with a clamping force of 2200 kN. The overmolded samples in this experiment had substrate dimensions of 50 mm x 90 mm and an overmolded area of 20 mm x 90 mm. When processing TX1501HF, the injection parameters, which are machine dependent, for a typical shot were as follows: the barrel temperature for TX1501HF was set to 270 ^{o}C, the injection speed set to 40 cm^3/s, and the holding pressure was set to 750 bar. As shown in Table 1, the first part of this study examined various TPU processing conditions. While the Mold Temperatures and TPE Barrel Temperatures are specifically stated, a baseline (DoE Designation "0") for the Injection Speed and Pack and Hold Time were established by varying the values until acceptable overmolded parts were generated. In our study, the baselines for the Injection Speed and Pack and Hold Time were found to be ~35 cm^3/s and ~550 bar, respectively.

Characterization

All overmolded samples were stored for at least 24 h at 23 ^{o}C and 50% RH prior to testing. After ageing and prior to testing, the four corners of the samples were removed to enable the samples to be inserted into the sample holder. Samples were then tested on a Zwick Roell Z010 Machine equipped with a 90^{o} sliding table peel test kit. This kit aligns with ASTM D6862 and ISO 913 standard test methods. Moreover, the carriage on this machine tracks at crosshead speed enabling exact observance of 90^{o} peel angle and ensures that the force required to advance carriage is not transmitted to the specimen. Samples were tested at 23 ^{o}C and 50% RH at a peeling speed of 100 mm/s. (A minimum of 10 samples were tested for each DoE Run.) During testing, all samples were tested until a sample exhibited cohesive substrate failure (tearing of the TPE) or the material was fully peeled off. Adhesion strength values for each sample are reported as the mean value of the curve over the set peel length. When calculating the mean value, the initial rise at the start of the test (first 20% of the peel test) and the end of the test (last 5% of the peel test curve) were excluded from the adhesion strength calculation. Finally, the values from ten samples were averaged and reported as the adhesion strength value for the sample set. (To calculate peel strength, one would simply divide the adhesion strength value by the width of the overmolded sample, in this case 20 mm.)

Results and discussion

Examination of TPE processing conditions on adhesion strength

To examine how the processing conditions impact the adhesion strength of TPEs overmolded onto Eastman Tritan™ TX1501HF, both the TPE (commercially available product not to be identified) and the thermoplastic resin (TX1501HF) were held constant. Moreover, the injection molding processing conditions for TX1501HF were held constant while the injection molding conditions for the TPE were varied. As seen in Table 1, four variables related to the TPE processing conditions were examined. Specifically, this study examined how three easy-to-change variables (Mold Temperature, Pack and Hold Time, and Injection Speed) and one hard-to-change variable (TPE Barrel Temperature) impact the adhesion strength of a TPE overmolded onto TX1501HF.

Table 1. Four parameters having a high (1), medium (0) and low (-1) DoE designation and a corresponding value were examined in this study.

Parameter	DoE Designation	Parameter Value
Mold Temp	-1	40 °C
	0	50 °C
	1	60 °C
TPE Barrel Temperature	-1	195 °C
	0	205 °C
	1	215 °C
Pack and Hold Time (TPE)	-1	Normal Pack and Hold Time
	0	Normal Pack and Hold Time Plus 10% longer
	1	Normal Pack and Hold Time Plus 20% longer
Injection Speed of TPE	-1	Normal Injection speed
	0	Normal Injection Speed + 10%
	1	Normal Injection Speed + 20%

Based on the parameters in Table 1, a DoE was designed and is displayed in the Appendix as Table A1. As shown in Table A1, 24 injection molding experiments utilizing various processing conditions were identified and completed to establish the main effects (linear terms), two-factor interactions and quadratic terms between the investigated variables and the adhesion strength. The individual average force values are plotted for each run in the DoE in Figure 1. The average force values calculated for each run are displayed by the connected blue line. Note, the impact of outliers can be eliminated by calculating the median force values and is displayed in Figure 1 as the middle line in box plot. The mean, standard deviation, lower 95% confidence interval, upper 95% confidence interval, and the median values for each DoE run are presented in Table A2.

As displayed in Figure 1, the average adhesion strength values (N) range from 240 N to 294 N. Utilizing the experimental data from Figure 1, we were able to generate a model between the significant variables and the adhesion strength. As shown in Figure A1, plotting the experimental values from Figure 1 vs the model's predicted value shows good agreement.

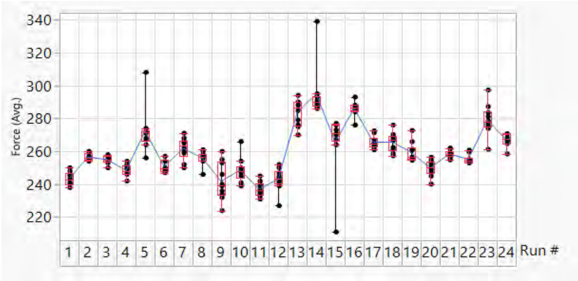

Figure 1. Average force values (blue line) plotted vs DoE run number. Median force values, which remove the effect of outliers, are displayed by the middle line in the box plot.

Specifically, the R-Square and Adjusted R-Squared statistics are 0.92 and 0.91, respectively. These values emphasize the model's agreement with experimental results. The parameter tests table shown in Figure A1 displays that TPE Barrel Temperature, Mold Temperature and Pack and Hold Time are all statistically significant at the 95% confidence level. (The effect of Injection Speed was not deemed significant within the investigated range and was removed from the model.) After the main variables that impact TPE adhesion strength were identified, next a profile plot was created to display the trends of the average force average with each process variable deemed statistically significant. Figure 2 shows that the average force value increases as each individual parameter (TPE Melt Temperature, Mold Temp, and Pack and Hold Time) increases.

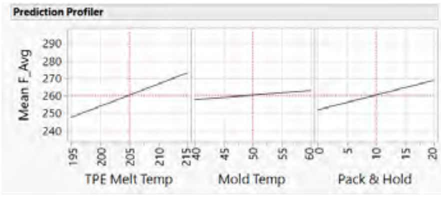

Figure 2. Average force average plotted against each parameter's respective values. Plot #1 displays an increase in average force as TPE Melt Temperature increases, Plot #2 displays an increase in average force as Mold Temperature increases, and Plot #3 displays an increase in average force as Pack and Hold Time Increases.

The final profiler, shown in Figure A2, displays that by adjusting the processing settings to generate the maximum adhesion force, a value of 284 N is predicted. More specifically, the final profiler suggests that in order to obtain the maximum peel force that 1) the TPE Melt Temperature should be set to the highest value (215 ^{O}C), 2) the Mold Temperature should be set to the highest value (60 ^{O}C); and 3) the Pack and Hold Time should be set at the highest level (normal time plus 20% longer pack and hold). Note, based on this study, the Injection Speed of the TPE was not found to have a significant impact on the peel force and thus could, in theory, be set at any level within the range studied (normal speed to normal speed +20%). Reflecting on the DoE, run 14 utilized the optimized settings noted in the final profiler to generate maximum adhesion strength and displays an average value of 294 N. This average force value was indeed the highest adhesion force observed during experimental testing (see Figure 1 and Table A2). To confirm performance outside of the DoE, additional samples were generated using the optimized settings identified by the DoE. Specifically, adhesion strength traces obtained for samples generated using the DoE optimized conditions are shown in Figure A3. As shown in Figure A3, the average adhesion strength value of samples generated using the optimized settings was found to be 274 N, which is within one standard deviation of the predicted maximum average adhesion force value from the model. In summary, results from the DoE suggest that TPE Barrel Temperature, Mold Temperature, and Pack and Hold Time should be maximized to maximize adhesion force for TPEs overmolded onto TX1501HF.

Examination of commercial TPEs

After completing the DoE, two questions remained: 1) How translatable are the optimized processing conditions to other TPEs, and 2) How does the choice of TPE impact adhesion performance? To address these questions, additional TPE grades were acquired and tested. A minimum of ten samples from each grade were generated via the same 2k overmolding process. During the generation of these additional overmolded samples, again utilizing different TPEs, many observations were noted. Specifically, and as expected, each TPE has a specified processing window. In addition to changes in the suggested processing conditions for each material, as materials change so do their inherent properties (viscosity, thermal stability, etc.,) which also impacts how the material can be injection molded. For example, some TPEs had lower maximum processing temperatures (up to 25 ^{O}C lower than the maximum temperature utilized in the DoE), lower acceptable mold temperatures, required longer cooling times for ejection, and required lower pack and hold pressures and times. Note, these processing changes were made while the injection molding machine remained constant. Changing machines is also expected to have a drastic impact on the processing conditions for each material. Coupling this insight, as materials and machinery change the processing conditions change, with the learnings from this study, one should adhere to the following guidance to maximize adhesion strength. Specifically, and in accordance with the processing conditions stated in a TPE's TDS, one should maximize the Mold Temperature, TPE Barrel Temperature, and Pack and Hold Time to obtain the best adhesion strength performance. Moreover, cooling time should also be maximized to ensure smooth ejection out of the mold. Note, these suggestions should always be considered and optimized within the limitations of cycles times.

Lastly, results from a preliminary screening of additional TPEs underscore the importance of the choice of TPE. Specifically, out of the TPEs examined, two commercial grades offered by Lubrizol displayed the best adhesion strength to Tritan™ TX1501HF. Compared to the TPE utilized for the DoE work, which displayed adhesion strength values ~ 294 N, two ESTANE® ECO TPUs from Lubrizol's portfolio displayed average adhesion strengths above 310 N, or up to a 5% increase in adhesion performance. Moreover, the mean difference between ESTANE® ECO 12T80E and the TPE utilized in the DoE is 22 N and is statistically significant at the 95% confidence level. Both ESTANE® ECO TPU grades utilized in this study are renewably sourced products that contain bio-based content according to ASTM D6866. These block copolymers based TPUs are made up of polyol soft segments and polyurethane hard segments. In our study, and as shown in Figure 3 and Figure 4, we observed ESTANE® ECO 16T85 and ESTANE® ECO 12T80 to have average adhesion values of 311 N and 316 N, respectively.

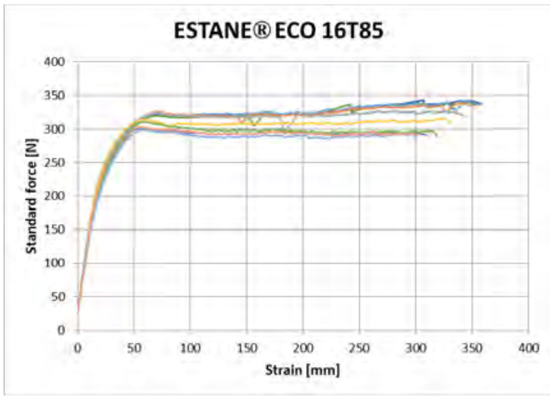

Figure 3. Adhesion strength traces of ESTANE® ECO 16T85 overmolded onto Tritan™ TX1501HF. The average adhesion strength value calculated from this data was found to be 311 N.

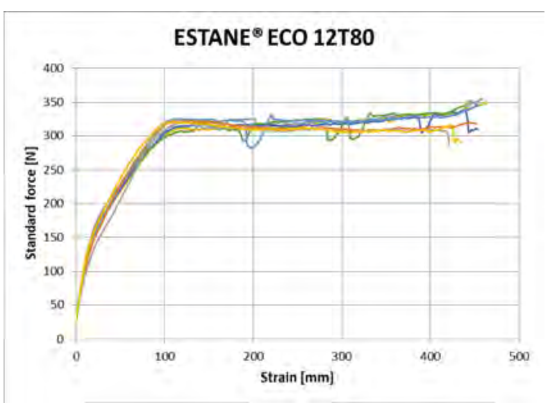

Figure 4. Adhesion strength traces of ESTANE® ECO 12T80 overmolded onto Tritan™ TX1501HF. The average adhesion strength value calculated from this data was found to be 316 N.

While these examples underscore the importance of the choice of TPE, they are by no means all encompassing. For instance, higher and/or lower peel force values could be obtained when overmolding other TPEs onto TX1501HF or when overmolding ESTANE® ECO TPU grades onto other rigid thermoplastic substrates. However, these results emphasize the importance of chemical compatibility between substrates. Which, as described in the introduction, can be impacted by the wetting of the substrate, interdiffusion of polymer chains across material interfaces, and ensuring solidification within the interphase.

In total, this study highlights how the adhesion strength of TPEs overmolded onto TX1501HF can be impacted by optimizing the processing conditions and increasing the compatibility between the TPE and overmolded substrate. As compared to samples generated in unfavorable conditions that displayed an average adhesion strength value ~240 N, optimizing the processing conditions and increasing, what is suspected to be, chemical compatibility led to a ~32% increase (up to 316 N) in adhesion strength.

Conclusion

While the exact processing conditions will be dependent on the guidance provided in a TPE's TDS and equipment utilized to generate the parts; in general, the results obtained from this DoE suggest that TPE Barrel Temperature, Mold Temperature, and Pack and Hold Time should be maximized to maximize the adhesion strength of overmolded samples. While production cycle times may limit one's ability to "max-out" certain variables, such as Pack and Hold Time, in general the variables highlighted above should be maximized within production time constraints to obtain the best overmolding performance. Moreover, by utilizing the optimized conditions established by the DoE, we found that the choice of TPE does influence adhesion strength. Specifically, two commercially available

TPU grades, ESTANE® ECO 16T85 and 12T80, displayed the highest adhesion strengths obtained in this study. These results underscore that in addition to the processing conditions, chemical compatibility between two substrates also has an impact on adhesion strength.

Acknowledgements

Sample support of ESTANE® ECO TPU grades from The Lubrizol Corporation is gratefully acknowledged.

Adhesion strength measurements were completed at Eastman's Technology Center in Ghent, Belgium.

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Appendix

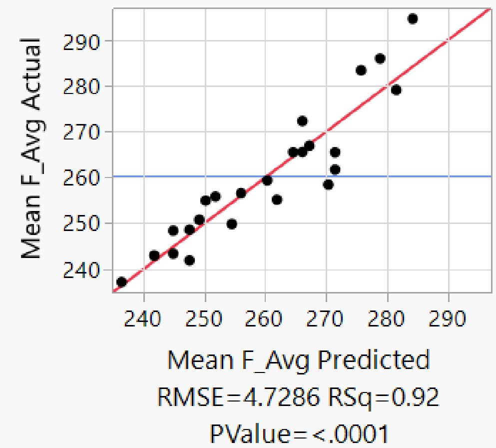
Table A1. The DoE designed to examine the impact four processing parameters (TPE Barrel Temp, Mold Temp, Pack and Hold Time, and Injection Speed) have on the adhesion performance of TX1501HF substrates overmolded with a TPE. This DoE will estimate the main effects (linear terms), two-factor interactions and quadratic terms.

Run#	Extrusion Day	Whole Plots	TPU BarrelTemp	Mold Temp	Pack & Hold	Injection Speed
1	1	1	-1	-1	0	-1
2	1	1	-1	0	1	0
3	1	1	-1	1	0	0
4	1	1	-1	0	0	1
5	2	2	0	-1	1	-1
6	2	2	0	-1	-1	0
7	2	2	0	1	1	1
8	2	2	0	0	-1	-1
9	3	3	-1	0	0	0
10	3	3	-1	0	0	0
11	3	3	-1	-1	-1	1
12	3	3	-1	1	-1	-1
13	4	4	1	1	0	-1
14	4	4	1	1	1	1
15	4	4	1	0	-1	1
16	4	4	1	-1	1	0
17	5	5	0	1	1	-1
18	5	5	0	-1	1	1
19	5	5	0	0	0	0
20	5	5	0	1	-1	1
21	6	6	1	-1	0	1
22	6	6	1	-1	-1	-1
23	6	6	1	0	1	-1
24	6	6	1	1	-1	0

Table A2. The Mean, Standard Deviation, Lower 95%, Upper 95% and Median values for each DoE run.

DoE Experiment	Mean	Std Dev	Lower 95%	Upper 95%	Median
Run #[1]	243.4	3.835507	240.6562	246.1438	243
Run #[2]	256.6	2.1187	255.0844	258.1156	256
Run #[3]	255	2.44949	253.2477	256.7523	255
Run #[4]	248.4	3.50238	245.8945	250.9055	248
Run #[5]	272.4	13.58267	262.6835	282.1165	271.5
Run #[6]	250.8	3.794733	248.0854	253.5146	249.5
Run #[7]	261.8	6.729702	256.9859	266.6141	263.5
Run #[8]	255.9	4.148628	252.9323	258.8677	256.5
Run #[9]	241.9	11.38664	233.7545	250.0455	239.5
Run #[10]	248.6	7.560129	243.1918	254.0082	248
Run #[11]	237.2	4.315347	234.113	240.287	236.5
Run #[12]	243	6.896054	238.0669	247.9331	243.5
Run #[13]	283.5	7.947746	277.8145	289.1855	286.5
Run #[14]	294.8	15.76071	283.5255	306.0745	290
Run #[15]	265.6	19.61972	251.5649	279.6351	271
Run #[16]	286.1	4.306326	283.0194	289.1806	286.5
Run #[17]	265.5914	3.707784	262.939	268.2438	264.8283
Run #[18]	265.668	5.77235	261.5387	269.7973	267.1333
Run #[19]	259.4342	5.895016	255.2171	263.6512	256.5069
Run #[20]	249.8719	4.834016	246.4138	253.3299	250.4251
Run #[21]	258.4683	2.237853	256.8674	260.0692	258.2774
Run #[22]	255.2045	2.741512	253.2433	257.1657	254.4993
Run #[23]	279.1933	9.386007	272.479	285.9077	277.9953
Run #[24]	266.9719	3.751265	264.2884	269.6554	267.5429
Force (Avg.)	260.2251	16.35498	258.1455	262.3048	257.1141

Actual by Predicted Plot



Summary of Fit

RSquare	0.924469
RSquare Adj	0.91314
Root Mean Square Error	4.728619
Mean of Response	260.2251
Observations (or Sum Wgts)	24

Parameter Estimates

Term	Estimate
Intercept	-23.66052
TPE Melt Temp	1.2785294
Mold Temp	0.2681375
Pack & Hold	0.8491985

Parameter Tests

Term	Prob>|t|
Intercept	<.0001*
TPE Melt Temp(195,215)	0.0056*
Mold Temp(40,60)	0.0387*
Pack & Hold(0,20)	<.0001*

Figure A1. The experimental values plotted vs the model's predicted value show good agreement. Specifically, the R-Square and Adjusted R-Squared statistics are 0.92 and 0.91, respectively. The parameter testes tables shows that TPE Melt Temperature, Mold Temperature, and Pack and Hold Time are statistically significant at the 95% confidence level. The effect of Injection Speed was not significant and was removed from the model. The parameter estimates can be used to predict the average of the average force within the ranges of the process factors studied.

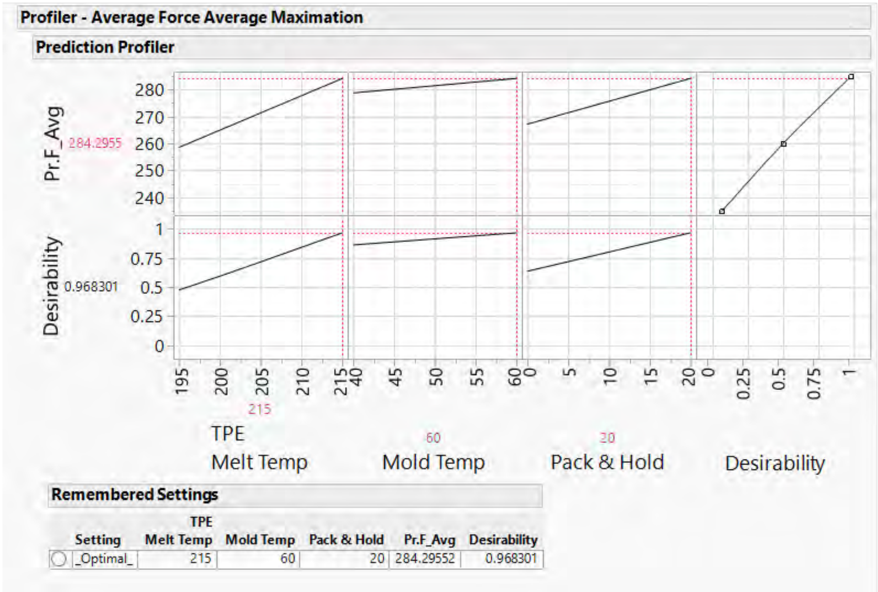


Figure A2. The Profiler shows that optimizing the processing settings to generate a maximum adhesion force predicts a value of 284 N. In order to obtain this value, all parameter (TPE Melt Temp, Mold Temp, and Pack and Hold Time) should be maximized.

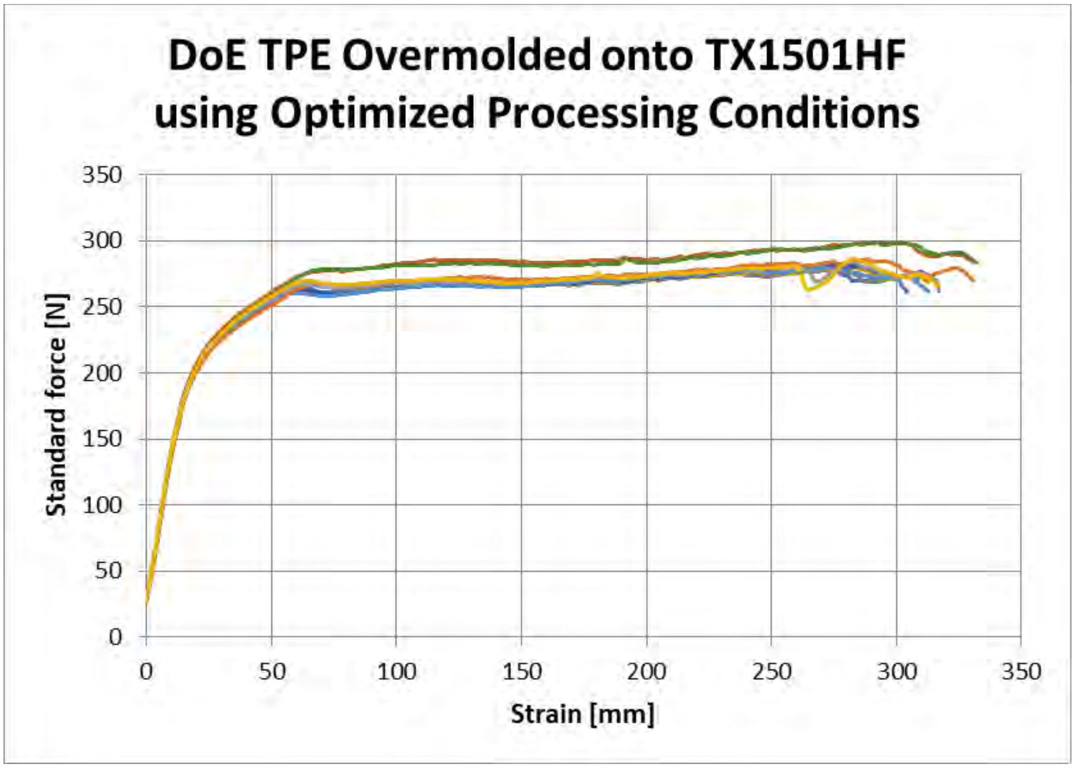


Figure A3. Adhesion strength traces of the DoE TPE (not identified) overmolded onto Tritan™ TX1501HF using the optimized settings identified by the DoE profiler. The average adhesion strength value calculated from this data was found to be 274 N, which is within one standard deviation of the adhesion strength value predicted by the profiler.