

## HEAT-SEAL STRENGTH ANALYSIS OF LAMINATED FILMS WITH LLDPE AND LDPE SEALANT MATERIALS IN BAR SEALING APPLICATION

C.S. Yuan<sup>1</sup>, A. Hassan<sup>2</sup>, M.I. Ghazali<sup>3</sup> and A.F. Ismail<sup>4</sup>

<sup>1</sup>Department of Thermal-Fluid, Faculty of Mechanical Engineering, Kolej Universiti Teknikal Kebangsaan Malaysia, PO Box 1200, 75450 Ayer Keroh, Melaka, Malaysia.

<sup>2</sup>Department of Polymer Engineering, Universiti Teknologi Malaysia, Sekudai, Malaysia.

<sup>3</sup>Department of Mechanical Engineering, Kolej Universiti Teknologi Tun Hussein Onn, Johor, Malaysia.

<sup>4</sup>Department of Gas Engineering, Universiti Teknologi Malaysia, Sekudai, Malaysia.

Email: [cheng@kutkm.edu.my](mailto:cheng@kutkm.edu.my)

### ABSTRACT

*The heat-seal strength and failure modes of laminated films with Linear Low Density Polyethylene (LLDPE) and Low Density Polyethylene (LDPE) as the sealant materials were investigated. Heat-seal strength increases with platen temperature for all samples. The maximum achievable heat-seal strength for laminated film using LDPE and LLDPE as sealant material is 0.598 N/mm and 4.02 N/mm, respectively. Four failure modes were observed, namely, the peeling mode, tearing mode, delamination mode and combination of tearing and delamination mode failures. Strength of heat-seal and its failure modes are closely related. Heat-seal strength achieved under tearing mode failure is the highest, follow by heat-seal strength achieved under delamination mode failure, and the heat-seal strength achieved under peeling mode failure is the lowest. Corresponding platen temperature  $T_C$  and heat-seal strength  $SS_C$  where acceptable heat-seal begins to be made were 118, 114 and 108 °C, and 2.82, 2.38 and 0.48 N/mm for sample #1, #2 and #3, respectively.*

**Keywords:** heat-sealing; linear low density polyethylene (LLDPE film); low density polyethylene (LDPE film); seal strength, failure mode

### INTRODUCTION

Of all the sealing techniques used in packaging industry today, heat sealing is the predominant technique to fabricate flexible and semi-rigid packages of all shapes, sizes and types 1. Although a wide array of packaging equipment is employed to form heat-seal on a pouch, the basic sealing methodology is the same, which is bonding together of two polymer surfaces by forcing those surfaces into intimate contact while they are in at least a partially molten state 2. Therefore, proper control of platen temperature based on the requirement of laminated film is important in assuring that the pouches is leak free and with an acceptable heat-seal.

In packaging, a pouch made of a laminated film can be no stronger than the seal that holds it together 3. Therefore, acceptable heat-seal can be defined as a seal which has its strength greater than the strength of the packaging material. If this requirement is not fulfilled, heat-seal of a package will break open before the laminated film. Consequently, this would be a waste of using strong laminated film which is usually more expensive, as the package can easily failed at its seal when placed under stress. Based on this definition, an acceptable heat-seal is one that failed in delamination or tearing mode failure during peeling test. Thus qualitative study on the failure modes of heat-seals is equally important as quantitative measurement on the heat-seal strength; because this is how an acceptable heat-seal is identified. Note that this definition does not apply to peelable seal which is designed for easy opening packages 4.

Several studies concerning heat-sealing parameters and the characteristics of the sealant material have been reported in literature. Theller 5 was the pioneer researcher of this technique. He studied the heatsealability of plastic film in bar sealing applications. He reported that the interfacial temperature and dwell time are to be the primary factors in controlling heat-seal strength. Pressure normal to the seal surface had little effect above the level required to flatten the web for good contact. Stehling and Meka 26 had done a series of studies concerning heat-sealing process. The effect of heat-sealing process variables (seal bar temperature, dwell time and pressure) on seal properties (seal strength, seal elongation and seal energy) of polyethylene films was quantitatively determined. Their study also shows that to obtain the highest possible seal strength for a given semicrystalline polymer, the required platen temperature can be estimated with the given dwell time and interfacial temperature

by finite element model. Their results agree with Theller 5 where heat-seal strength is primarily control by sealing temperature and dwell time, rather than pressure. Tetsuya et al 7 had investigated the effect of heat-sealing temperature on the mechanical properties and morphology of oriented polypropylene (OPP)/cast polypropylene (CPP) laminated films. They reported that tensile strength of the seal was affected by the orientation of the films. While Hashimoto et al 8 had carried out investigation on the failure criteria of the heat-sealed part of OPP and CPP heat seals made by impulse type heat-sealing machine. They reported that seals were stronger in the transverse direction as compared with the machine direction. Studies of the effect of corona-discharge treatment (CDT) on heat-seal strength was done by Owens 9 and Farley and Meka 10. The former has reported that corona-treated polyethylene films exhibit strong self-adhesion when joined together under conditions of heat and pressure that give no adhesion with untreated films. Whist, the latter found that the primary effect of CDT on the heat-sealing behavior of LLDPE films is a transition in the failure mode of heat-seals from a normal tearing or inseparable bond to a peelable seal. They also found that CDT increases the seal initiation temperature by 5~17°C and decreases the plateau seal strength by 5~20% as the treat level, or wetting tension, increases from 31 to 56 dynes/cm. The effect of various sterilization methods and acceleration aging on heat-seal-adhesive-coated, spunbonded High Density Polyethylene (HDPE) film was studied by Jones 11. She reported that no practically significant decline in seal strength, transfer, sterilizer creep, or cold flexibility was observed after the film was being exposed to various sterilization methods and after undergoing up to 10 weeks of accelerated aging, regardless of the bottom web to which the coating was sealed or of the sterilization method employed. The effect of material type on heat-seal strength was reported by Falla 1 and Halle and Davis 12. The earlier reported that the coextruded films having a heat-seal layer comprised of the ultra low density polyethylene (ULDPE) resins in the sealing layer of a coextruded film significantly increase the heat-seal/hottack range. The ULDPE resin provides broader sealing range for pouch conversion and better physical properties in finished pouches. And the latter have shown that to obtain excellent heat-sealing performance from unlike seal layers is possible by utilizing a unique family of linear ethylene plastomers.

In this study, the heat-seal strength and failure modes of laminated film using LLDPE and LDPE as the sealant material are investigated. Platen temperature and heat-seal strength corresponds to the point at which an acceptable heat-seal begins to be made is quantified for each laminated film.

## MATERIALS AND METHODS

### Laminated Films

Three samples were used in this study, which are commercial laminated films of various constructions, thickness and sealant material use in flexible packaging application. In samples #1 and #2, LLDPE is used as the sealant material, while sample #3 using LDPE. Details on laminated film description are as shown in Table I.

Table 1: Description of laminated films for the study

Laminated Film Designation	Construction Description "Outer Layer/Inner Layer(s)/Sealant Material( $\mu\text{m}$ )"	Overall Thickness ( $\mu\text{m}$ )
1	PET/LLDPE(65)	79
2	PET/LDPE/F/PET/LLDPE(50)	99
3	OPP/LDPE/F/LDPE(20)	59

Samples #1 and #2 are manufactured from monolayer films using the dry-bond process. In this technique, a liquid adhesive (urethanes) is applied to one substrate. It is then dried with hot air. The dried surface is then adhered to a second substrate using heat and pressure. Sample #3 was manufactured using extrusion lamination process. In this process, a thin layer of LDPE was used to bond together two layers of film and foil.

### Melting Point Distribution

In this study, Perkin Elmer Differential Scanning Calorimeter (DSC), model DSC-7, was used to determine the melting point  $T_m$  of the sealing materials. The melting point of a polymer is generally taken as the point at which the maximum energy is absorbed. In DSC experiment, this corresponds to the peak maximum of the melting endotherm 13. Samples (aged at room temperature for at least 40 hours in accordance with ASTM Method D 685) of 6  $\pm$ 4 mg were prepared and placed in standard aluminum pans respectively. The samples were scanned at 10 °C/min scan rate starting at 25 to 300 °C under a helium purge gas. Results are summarized in Table 2.

Table 2: Melting point of sealant materials determined using DSC

Sealant Materials	Material Forms	Melting Point $T_m$ (°C)
LLDPE	Monolayer film	104.6
LDPE	Pellet	88.9

## Preparation of Heat-seals

In the preparation of heat-seal, laminated films are first cut into 15 mm wide strips. Clean-cut edges are produced to prevent premature failures in T-peel test. Heat-seals were made in the laboratory using a model HSG/ETK heat-sealer, made in Germany. This device clamps two pieces of filmstrips between flat, 10 mm wide heated metal bars. The temperature, pressure and dwell time of the sealing bars are adjustable. Microprocessor programmed controllers maintained and digitally indicated set temperature for each bar. Both bars were operated at the same temperature in these experiments, and keep closed between sealing to minimize heat loss and temperature fluctuations. The pressure was fixed at 2 bars while platen temperature and dwell time varies. After the heat-seal was made, the sandwich structure was allowed to cool at ambient condition. Dimensions of the heat-seal sample is as shown in Figure 1.

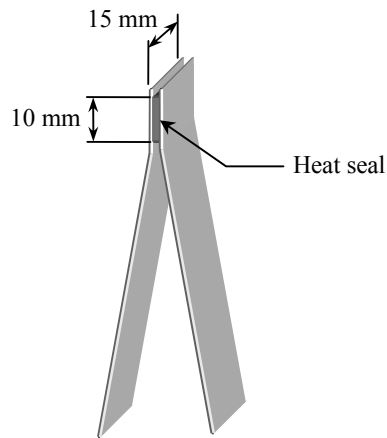


Figure 1: Heat-seal dimensions

## Testing of Heat-seals

After the heat-seals were produced, it were allowed to condition at room temperature for at least 40 hours to achieve chemical stabilization. Aging of heat-seal was necessary as the strength of seal may change in time, which may due to the memory of polymer, or thermophysical properties of polymer as the heat-seal samples undergo melting and cooling processes 10. In practice, packaging goods are aged through storage before being used by the consumer. Therefore, heat-seals made in this study were aged for at least 40 hours before tested.

The strength of heat-seal is often determined by measuring the force required to pull apart the pieces of which have been sealed together, ether in a dynamic or static load test 14. In this study, the sample was peeled apart at room temperature in tensile tester of model MICRO 350, using a 100 N load cell. Each leg of the test specimen was clamped in the tensile tester in such a way that the specimen is aligned in the clamps with its seal line perpendicular to the direction of pull (as shown in Figure 2). The heat-seal area of the specimen are placed at approximately equidistant between the clamps. The constant rate of loading 300 mm/min with initial jaws separation of 25 mm was chosen as recommended by ASTM F88 - 85. The maximum force required to tears apart the seal, and failure mode of each pull was recorded.

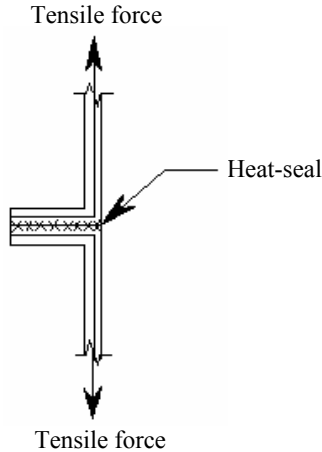


Figure 2: Direction of pull is 90° to the seal in peeling test.

## RESULTS AND DISCUSSIONS

### Heat-seal Strength and Failure Modes

The peeling test measures the force required to separate a heat-seal after it was allowed to cool. The maximum force per width value obtained in such a test is commonly defined to be the heat-seal strength  $SS$ . Hence, heat-seal strength  $SS$  is expressed as:

$$SS = \frac{F}{W} \quad (1)$$

Where  $F$  = Maximum force in peeling test measure (N)  
 $W$  = Width of the heat-seal (15 mm)

All representative values in this paper are the average of six. In addition, failure modes at each pulled was carefully examined and platen temperature at which the acceptable heat-seal begins to be made is recorded. Similar to the results reported by previous researchers [4678], the heat-seal failure of the laminated films was found greatly influenced by then platen temperature and were occurred in three stages as following: -

Heat-seal made at temperature substantially lower then the melting point of the sealant material was found to fail in peeling mode failure. Under this failure mode, disentangles and extricates chain ends from the opposite surface occurred, in which the heat-seal bond was peeled apart (Figure 3). This happened when the strength of the seal is lower than the strength of the laminate structures. Consequently, heat-seal strength of all samples reported under this failure mode are lower than the strengths reported in other failure modes. Hence, peeling mode failure occurred when

$$SS - SL < 0 \quad (2)$$

Where  $SS$  = Strength of heat-seal (N/mm)  
 $SL$  = Strength of laminated film (N/mm)

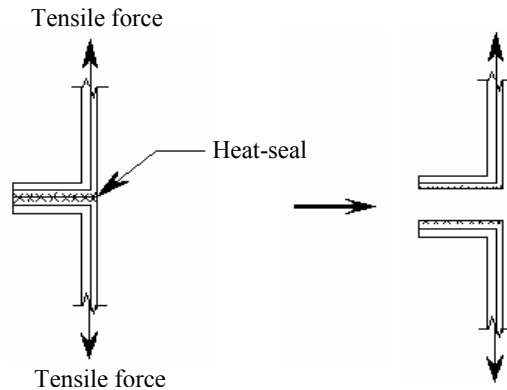


Figure 3: Sealing layer torn apart from each other

The strength of heat-seal increased gradually with sealing temperature (see Figure 4). This is attributed to the increased of thermal motion (micro-Brownian movement) of chain segments with temperature, and therefore, deepened the original zone of diffusion and causes a greater peeling force required to peel it apart.

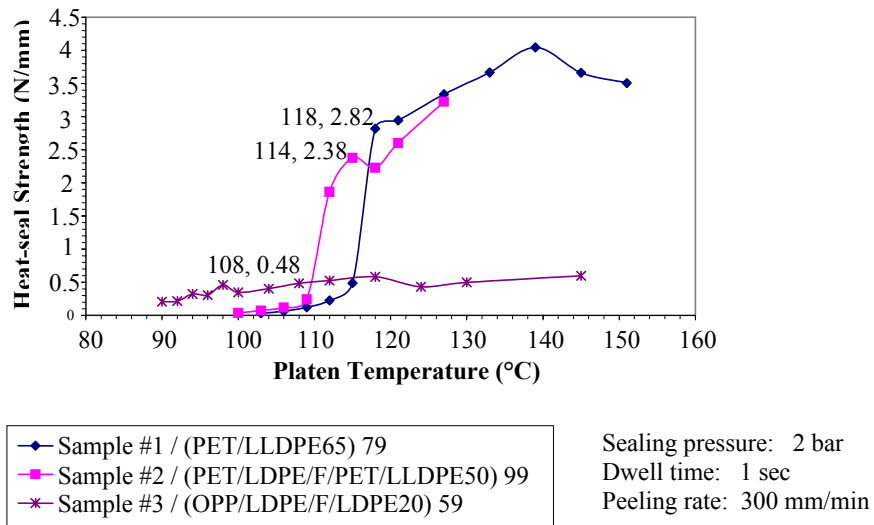


Figure 4: Graph of heat-seal strength versus platen temperature at dwell time of 1 sec.

At temperature a few degree Celsius before the final melting point of the sealant material, seal strength increased sharply for sample #1 and #2 (see Figure 4). However, this trend did not occurred on sample #1 which was due to its lower achievable heat-seal strength. This is the transition region in which failure mode was then changed from peeling to delamination (Figure 5) or tearing mode failure (Figure 6), or combination of both failure modes (Figure 7). Acceptable heat-seal strength was achieved at this stage.

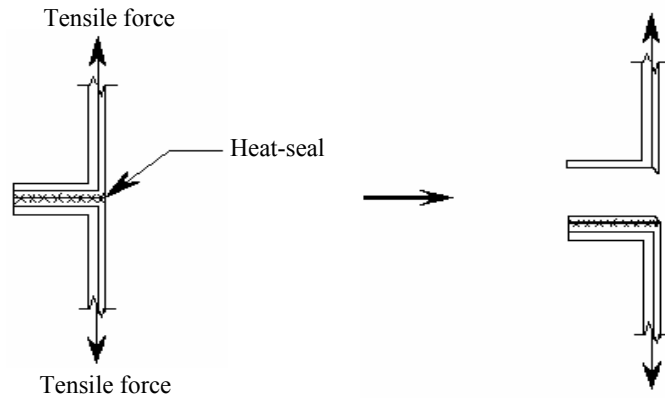


Figure 5: The laminated film separated from the sealing layer.

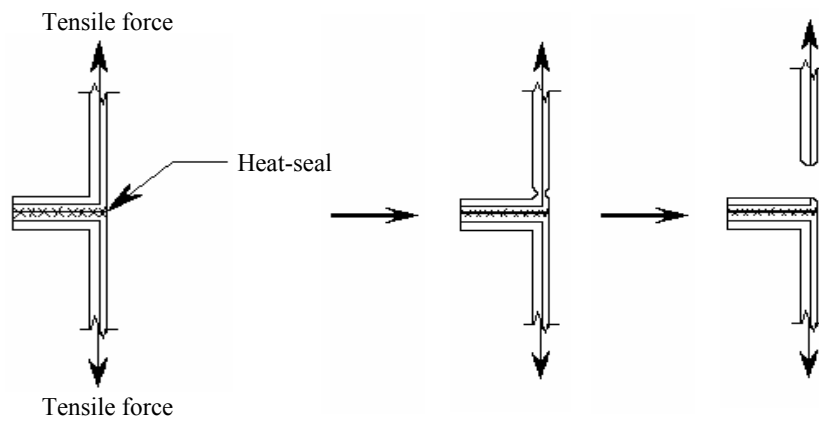


Figure 6: Breaking of the laminated film just at the edge of the seal.

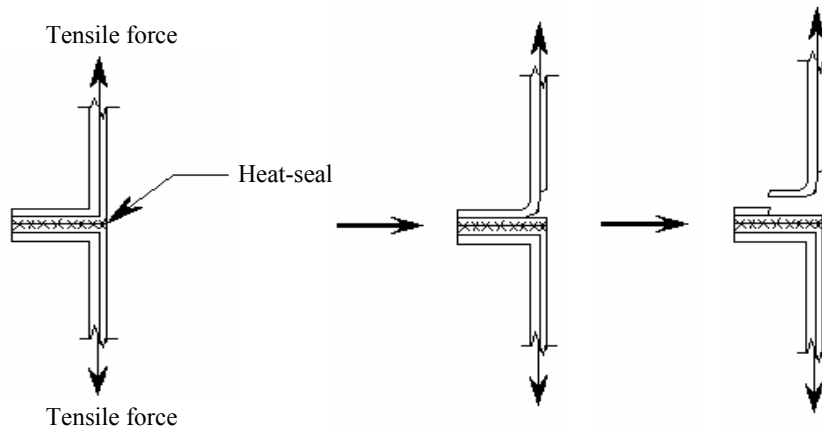


Figure 7: Combination of delamination and tearing modes.

Delamination mode failure involves tensile break of the heat-seal layer, which is thin and weak, followed by separation of the interlaminar bond. Whilst, tearing mode failure was due to the weakening of the seal at its edge as the material fused, or the weakening of the laminate structures as a result of interlaminar bond separation, plus the geometry of the peel test itself.

After the acceptable seal strength was achieved, no significant increased of seal strength with temperature were found for sample #3. In contrast, the strength of the seal for samples #1 and #2 continue to increase gradually with sealing temperature. The continuously increased of heat-seal strength in sample #1 and sample #2 may due

to the decreased of amorphousness in the LLDPE film. This is because, higher temperature setting deepen the molecular entanglement between polymer molecules 6. Consequently, LLDPE chain segments become closer packed, yielding a higher degree of crystallinity in the molecular structure. Therefore, higher strength is obtained since polymer with higher crystallinity offer greater physical strength 15.

Failure modes of heat-seal had a close relationship to its strength. As mentioned previously, heat-seal failed in peeling mode failure having relatively lower strength due to only a slight molecular entanglement between polymer molecules in the interfacial zone. Therefore, heat-seal can easily be peeled apart before any failure occur to the laminate structures which are relatively stronger than the heat-seal with only slight molecular entanglement.

As platen temperature increased, deeper molecular entanglement between polymer molecules in the interfacial zone occurred. Thus, the heat-seal strength increased subsequently as the depth of molecular entanglement increased. Up to a certain level, when the strength of the heat-seal exceeded the strength of the interlamina bond, the laminate structures separated from the sealing layer instead of peeling of the heat-seal, and leaving that portion and the other laminated film intact (Figure 5). Hence, heat-seal strength reported in delamination mode failure was relatively higher than the heat-seal strength reported in peeling mode failure.

The increased of platen temperature not only causes increased in heat-seal strength, but also increased the strength of interlamina bond of the laminated films. However, under marginal condition, uneven strengthening of interlamina bond on heat-seal area occurred. Laminated films tested failed in the combination of delamination and tearing modes failure. In these failure modes, the laminated film was first separated into monolayer structures at the peel line, then when come to the area in which uneven strengthening of interlamina bond occurred, the monolayer structure broke simultaneously after delamination, due to the uneven tensile stress distribution (Figure 7) at the heat-seal area.

Heat-seals made at a higher platen temperature after obtaining the combine failure modes had its interlamina bond strength being strengthened evenly at the heat-seal area. Laminated film tested was then failed in tearing mode failure. This is because the strength of the laminate structures (monolayer films) are now relatively lower then the strength of the interlamina bond and the heat-seal. Under this failure mode, the laminated film broke just at the edge of the heat-seal (Figure 6). Heat-seal strength achieved under this failure mode is the highest compared to heat-seal strength reported under other failure modes. The maximum achievable heat-seal strength for laminated film using LDPE and LLDPE as sealant material is 0.598 N/mm and 4.02 N/mm, respectively.

### Corresponding Heat-seal Strength and Platen Temperature for Acceptable Heat-seal

The corresponding heat-seal strength  $SS_C$  and platen temperature  $T_C$  where acceptable heat-seal begins to be made were identified based upon the failure modes of the T-peel test samples. Results are summarized in Table 3.

Table III Corresponding heat-seal strength  $SS_C$  and platen temperature  $T_C$  where acceptable heat-seal begins to be made

Laminated Film's Designation	$SS_C$ (N/mm)	$T_C$ (°C)
1	2.82	118
2	2.38	114
3	0.48	108

### CONCLUSIONS

The main conclusions that can be derived from the study are as follows:

1. Strength of heat-seal increases with platen temperature. The maximum achievable heat-seal strength for laminated film using LDPE and LLDPE as sealant material is 0.598 N/mm and 4.02 N/mm, respectively.
2. Corresponding platen temperature where acceptable heat-seal begins to be made are 118, 114 and 108 °C for Sample #1, #2 and #3, respectively.
3. Corresponding heat-seal strength where acceptable heat-seal begins to be made are 2.82, 2.38 and 0.48 N/mm for Sample #1, #2 and #3, respectively.

4. Strength of heat-seal and its failure modes are closely related. Heat-seal strength achieved under tearing mode failure is the highest, follow by heat-seal strength achieved under delamination mode failure, and the heat-seal strength achieved under peeling mode failure is the lowest.

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## ABBREVIATIONS AND NOTATIONS

CDT	-	Corona-discharge Treatment
CPP	-	Cast Polypropylene
DSC	-	Differential Scanning Calorimeter
<i>F</i>	-	Maximum force in peeling test
HDPE	-	High Density Polyethylene
<i>LDPE</i>	-	Low Density Polyethylene
<i>LLDPE</i>	-	Linear Low Density Polyethylene
OPP	-	Oriented Polypropylene
<i>T<sub>C</sub></i>	-	Corresponding platen temperature where acceptable heat-seal begins
<i>T<sub>m</sub></i>	-	Melting Point
ULDPE	-	Ultra Low Density Polyethylene
<i>W</i>	-	Width of heat-seal
<i>SS<sub>C</sub></i>	-	Corresponding heat-seal strength where acceptable heat-seal begins
<i>SL</i>	-	Strength of laminated film
SS	-	Heat-seal Strength