Effects of process parameters and solid particle contaminants on the seal strength of low-density polyethylene-based flexible food packaging films

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Seal strength is a key indicator of heat seal quality in flexible packaging. In this study, the effect of seal bar geometry, material composition and food particle contamination on the seal strength of widely used low-density polyethylene (LDPE)-based compound films was examined. Additionally, the maximum level of allowable solid food particle contamination was determined for ground coffee particles and powdered sugar. The results showed that adding metallocene LLDPE compound decreases seal initiation temperature (SIT) and increases overall seal strength. Also, changing seal bar geometry from flat to grooved bar with 0.56-mm pitch height enhanced the seal strength significantly. Moreover, pressure mapping and T-peel tests at SIT pointed out that grooved bars alter the pressure distribution and first contact points through the seal surface. Contamination of ground coffee particles at the seal interface as occurs during the packaging process when a powdery product is dropped in a package did not affect the seal strength up to 10 g/m² at 0.5-s dwell time. Above that amount, seal strength dropped dramatically. In the case of powdered sugar, threshold contaminant level was 2 g/m² at 0.5-s dwell time. Consequently, it has been revealed that knowing the type and the amount of contaminant during the food packaging process is important to maintain the seal quality, to find optimum process values between dosing and filling and to choose the right seal bar design because they can have a critical influence, especially at seal initiation phase.

KEYWORDS
contaminants at seal interface, flexible food packaging, heat seal strength, metallocene compounds, seal bar geometry

1 INTRODUCTION

Seal integrity is an important requirement in food packaging in terms of preserving food quality and safety. Microbial integrity studies show that pathogens can diffuse into the package through small channels less than 5 μm.1 Moreover, small channels in the seal interface allow gas and moisture ingress that can cause detrimental changes to the quality of sensitive products throughout the shelf life.2 Oxidation and
rancidity in coffee products can be given as an example of this situation.

According to the literature, four main factors influence the seal integrity of heat-sealed flexible packaging: material properties, sealing parameters, contaminant and further processes. As well as seal integrity, there is a strong relationship between these factors and adequate-uniform seal strength development. Although scientists are developing non-destructive leak detection methods such as ultrasonic imaging to determine the defective packages, measuring and optimizing seal strength is still a priority for the prevention of partially bonded and weak seals.

Monolayer films from such as low-density polyethylene (LDPE)-based compounds have many applications thanks to their relatively easy recyclability. Additionally, studies suggest that metallocene plastomers show superior seal performance such as lower seal initiation temperatures (SITs). That is why LDPE-based compound films containing metallocene linear LDPE (mLLDPE) have advantages in heat sealing. However, there are many other factors playing roles in the bonding quality of two layers, such as seal bar geometry and contaminants at the seal interface. Applying the optimum process parameters on the right material alone is not sufficient to eliminate sealing issues and leakages. The formation mechanisms of small channels at the seals produced by different types of seal bars are not well understood and encounter poor sealing problems that result in product loss and economical damage. Even though metallocene plastomers are able to seal through certain food contaminants, there is no study on the amount of contamination limit that does not risk seal security. Understanding the relationship between contaminant amount, characteristics and seal behaviour and examining the influence of seal bar geometry are vital as along with studying the effects of main process parameters on the seal quality.

This research focuses on understanding the parameters that influence heat-sealing behaviour of LDPE-based flexible food packaging films to detect the disturbances especially at the seal initiation stage. For this purpose, flexible packaging films containing LLDPE and mLLDPE compounds were characterized and compared with pure LDPE films. To find out the factors that have the most influence on seal strength among seal bar design, contamination and material composition, T-peel testing was applied. Additionally, contamination limits for ground coffee particles and powdered sugar were determined to find out the effects of powdery products at the seal interface.

### Table 1. PE-based packaging film compositions and their mechanical–thermal properties

| Film code | Film composition | Thickness (μm) | E-modulus (N/mm²) | Tensile strength (N/mm²) | Elongation (%) | Tm peaks (°C) |
|-----------|------------------|---------------|-------------------|--------------------------|---------------|--------------|
| Film 1    | 100% LDPE        | 50            | 370               | 20.1                     | 318           | 114          |
| Film 2    | 70% LDPE + 30% LLDPE | 50        | 280               | 25.4                     | 505           | 113          |
| Film 3    | 70% LDPE + 30% mLLDPE | 50        | 227               | 26.4                     | 496           | 115          |
| Film 4    | 60% LDPE + 40% mLLDPE | 50        | 303               | 29.4                     | 564           | 113          |

Abbreviations: LDPE, low-density polyethylene; mLLDPE, metallocene linear low-density polyethylene.
of seal bar designs presented in Figure 2 were used in this test, and the dimensions of those seal bars are given below. Ultra Super Low Pressure Prescale Fujifilms were chosen for pressure mapping. That type of Prescale Fujifilms can indicate the small pressure differences between 0.5- and 7-bar range, which also covers the average sealing pressures in flexible packaging. For the test, compression has been applied at 5-bar pressure, 3-s time and 23°C room temperature. The results were visually compared with the help of a standard colour scale chart.

Dimensions of the seal bars:

- Flat: \( w = 0.5 \text{ cm} \)
- Grooved 1: \( w = 1 \text{ cm}, \ h = 0.48 \text{ mm} - d = 1.66 \text{ mm} - \alpha = 120^\circ \)
- Grooved 2: \( w = 1 \text{ cm}, \ h = 0.56 \text{ mm} - d = 1.94 \text{ mm} - \alpha = 120^\circ \)

### 2.2.3 T-peel test

In T-peel test, influences of material composition, seal bar design and food particle contamination were examined on PE-based films that were given in the material section. T-peel test was performed at the SITs of each film, which were identified by the hot-tack test. Additionally, three different seal bar configurations (Flat, Grooved1 and Grooved2), which had been mentioned in the pressure mapping section, were applied for the sealing. All of the seal bars were covered with a thin Teflon tape to prevent sticking and burning of flexible packaging films. As food contaminants, ground coffee and powdered sugar were chosen. The coffee particles used in the experiment were around 200 \( \mu \text{m} \) in diameter, and the sugar particles were around 30 \( \mu \text{m} \). In the T-peel test, samples were prepared in 25 \( \times \) 200 mm size. Contaminant particles were spread evenly through the seal area with a small brush; 0.004-g contaminant was used for 10 cm\(^2\) film area to achieve 4 g/m\(^2\) contaminant density. The area to be contaminated was premeasured and marked on the test samples. Ground coffee was filled into a small container having a certain volume and weighed as 0.004 g. The same container was used for each contamination procedure. The tests were performed at 1-s dwell time and 0.8-bar seal pressure. After the sealing, samples were conditioned for 24 h at 23°C, 50% relative humidity. Then the peel strengths were measured with seal strength tester (Kopp, Labormaster HCT 3000, Germany) at 0.2 m/min peeling rate in accordance with ASTM F88.13

### 2.2.4 Determination of contaminant threshold

To determine the contaminant threshold, the material showing the best reduction in SIT and giving sufficient seal strength was chosen based on the hot-tack test results. Then, the level of contaminant threshold was determined for ground coffee particles (\( \geq 200 \mu \text{m} \)) and powdered sugar (\( \leq 30 \mu \text{m} \)). Samples were prepared in the same way as the T-peel test; however, this time, sealing duration was decreased to 0.5 s. The changes in the seal strength under the effect of elevated contamination densities (4, 8, 12, 16 and 20 g/m\(^2\)) and elevated temperatures (107–111°C) were analysed after 1 day of conditioning at 23°C temperature and 50% relative humidity. Peel strength measurements were performed at 0.2 m/min peeling rate.

### 2.2.5 Image analysis

Image analysis of film samples contaminated with 0, 4, 8, 12, 16 and 20 g/m\(^2\) ground coffee particles and powdered sugar were performed using VHX 7000 Digital Microscope (Keyence International, Belgium). Before the imaging of seal cross sections, samples were prepared by epoxy embedding. Then, embedded samples were both mechanically and chemically polished using Struers Tegramin 30 after 8-h drying. Then, the contaminated seals that have been sealed at SIT were inspected with the digital microscope at \( \times 200 \) magnification level. Additionally, digital camera images of contaminated seal surfaces were analysed via CellProfiler software.14
2.2.6 Statistical analysis

The mean comparisons of hot-tack test results were done via Tukey’s test ($p < 0.05$). For the T-peel test, full factorial design of experiment (DOE) was applied to evaluate the effects of seal bar design, material type and contamination ($p < 0.05$). Additionally, in the contaminant threshold test, response surface methodology (RSM) was used to map the impacts of increasing contaminant density versus temperature.

3 RESULT AND DISCUSSION

3.1 Hot-tack test

Hot-tack test results of each film can be seen in Figure 3. According to the results, the composite films containing LLDPE (C4) and mLLDPE (C6) show better sealing behaviour than pure LDPE film. Besides the seal strength increase, the difference in SITs is important to evaluate. SIT is defined as the temperature where a sufficient level of seal strength is first achieved. In this experiment, 2.5-N/25-mm seal strength is accepted as the starting point of sufficient sealing. As a result, it can be seen that SITs were lower in all composites. Also, the film samples containing mLLDPE compound (Films 3 and 4) presented better SIT reduction than the compounds containing butane-based LLDPE (Film 2). The SIT of Films 3 and 4 is 107°C and 108°C, respectively, while it is 110°C for Film 2. Additionally, it had been revealed that the increase of mLLDPE ratio in Film 4 supplied higher seal strength compared with Film 3 ($p < 0.05$). On the other hand, there is no statistical difference between Films 2 and 4 at the optimum sealing temperatures in terms of hot-tack strength ($p < 0.05$). This implies that using a 30% LLDPE (C4) compound supplies similar enhancement in seal properties as using a 40% mLLDPE (C6) compound. These results are also consistent with earlier studies indicating that metallocene plastomers supply superior sealing properties such as low SIT’s and high hot-tack strengths.\(^8\) According to Simanke et al., this superiority can be explained by a more homogenous comonomer distribution of mLLDPE. Also, in their chemical composition distribution study, most of the mLLDPE fractions elute below 80°C, and this explains lower SIT temperatures achieved at the hot-tack test.\(^{15}\)

Because the sealing times are generally shorter than 1 s in a fully automated food manufacturing process, the hot-tack test was repeated at 0.5 s. As a result, elevation in SIT is observed. In Films 1–4, SITs increased 4°C, 1°C, 3°C and 3°C, respectively, compared with 1-s dwell time. Initiation temperature of sealing process was the least time sensitive in Film 2 compounds containing 30% LLDPE (C4).

3.2 Failure modes

During the hot-tack test, several failure modes and their combinations were observed. The results are shown in Figure 4 for each type of film. In the table, Mode I indicates the temperature range for standard adhesive peeling behaviour without any observable defect. Elongation failure (Mode II) and material breakage failure (Mode III) start earlier in metallocene compound films, Films 3 and 4. Moreover, Mode III failure alone takes up the largest temperature range among the other failure modes in metallocene compounds. Failure Modes II and III are observed generally if the sealing temperature is close to $T_m$ values. Because, in the hot-tack test, peeling was applied immediately after the sealing, film samples were still hot when the failures occurred. This shows that increased tensile strength with the addition of LLDPE or mLLDPE compounds does not prevent material breakage at high temperatures.

A study by Planes et al. reveals that heat seal strength and observed failure modes are highly related. Complete peel failure (Mode I) corresponds to lower seal strengths as this study supports.\(^{16}\) When the seal temperature is increased, molecular diffusion and entanglement at the seal interface become greater. Thus, the peeling becomes more difficult, and Mode II failure, elongation, occurs. Failure Mode III, which represents the breakage at the seal edge, can be explained by maximized interdiffusion and entanglement at the interface that makes the bonding force at the seal area stronger than the packaging film itself.\(^{17}\) As it is known from the literature, tensile strength will decrease at elevated temperatures.\(^{18}\) This can also have a role in the occurrence of Modes II and III failures, and it explains the decreasing hot-tack strengths beyond a certain operation window.

3.3 Pressure mapping

In industry, various types of seal bars are used depending on package design and process requirements.\(^9\) Pressure distributions throughout the interface of different seal bars were obtained by pressure mapping, and the results were given in Figure 5. When the pressure intensities of flat and grooved seal bars were examined via the standard colour density scale, it was found that grooved bars were dramatically...
altering the pressure distribution within the sealing area. This result is in line with the study conducted by Matthews et al. According to it, if the crimp angle is more than 90°, sealing starts from the flat sides of the serration pattern of the grooved bars while the peak points stay as gaps. The grooved bars that were used in this study had a 120° crimp angle. Accordingly, peak points were applying less pressure than the other regions while the flat sides of the serration pattern were giving much higher colour intensity. Based on observed pressure alteration, it can also be expected that changing the size of the serrations will influence the seal strength. Especially at the SITs, serration patterns can influence the homogeneity of adhesion due to altered pressure and additional shearing effect. This additional shearing might occur in higher amount at bigger grooved patterns. Higher T-peel strength results obtained at second grooved bar that will be given in the following section can also be explained by this stretching factor.

### 3.4 T-peel test

In this study, the influence of different factors such as seal bar configuration, solid food particle contamination was examined on pure LDPE films and compound materials containing LLDPE and mLLDPE. According to the results presented in Figure 6, it can be concluded that the seal bar design has a significant effect on seal strength development at SIT’s of each film. Grooved bar having a bigger serration pattern with higher pitch height and pitch distance gave better seal strength results. This can be explained by the enhanced stretching effect through the sealing area and elevated shear factor through the larger grooved pattern. On the other hand, small serration patterns do not create a significant difference when it is compared with the flat seal bars. Between four different film samples, Film 1, which is pure LDPE, showed the breakage failure at the seal edge. Due to that failure, its peeling force was higher than the compound films while the results of compound films were not statistically different. Lower tensile strength and elongation values of pure LDPE than LLDPE and mLLDPE compounds can be the main reason behind this breakage failure. Besides, contamination of ground coffee or powdered sugar at 4 g/m² did not create any significant effect on the seal strength at 1-s dwell time. It is worth to mention that in the literature, contamination tests had been performed with much higher contaminant densities such as 25 g/m². In those higher densities, seal surface will be totally covered by the contaminant, and the negative effect of contamination on seal quality can be observed directly. However, in the packaging process of powdery or granulated products which are dropped down directly into the package via a tube, contamination may occur partially and at lower intensities. In this T-peel test, lower
amount of contamination (4 g/m²) was applied to imitate the packaging process more realistically. As a result, the film samples achieved adequate bonding although there are particles in the seal interface. Since the sugar particles were smaller, they were able to cover the entire seal surface as a thin layer. Even so this causes a higher drop down in the seal strength, the difference was not statistically important ($p < 0.05$).

Additionally, peeling force graphs indicating how the interface structure differs based on the seal bar configuration were given in Figure 7. Graphs show the peel force fluctuations throughout the peeling test. In flat bars, adhesion occurs more homogenously through the interface. On the other hand, in grooved bars, there are some irregularities at the peeling force. As can be seen in the graphs, the peeling force profile of the samples that sealed with different types of seal bars is also matching with the pressure mapping images of those seal bar geometries. As a consequence, it can be predicted that in the samples sealed with grooved seal bars, weak regions correspond to peak points of the serration pattern that apply less pressure. On the other hand, the regions showing good seal strength correspond to flat sides of the pattern that create a shearing effect. Even if theoretically, the overall seal strength increases by enhancing this shearing effect, gap regions were seen in the peel force pattern grooved bars may lead to leak formation. Also, according to industry reports, grooved bars may not be suitable to make hermetic seals on thin, monolayer packaging films due to non-uniform pressure created by rigid seal bars across the seal.21

3.5 | Contaminant threshold

Threshold contaminant analysis was performed to understand the effects of elevated solid food particle contamination under different

![FIGURE 6 Main effect plot for seal strength at seal initiation temperatures (SITs) (* represents statistically different results at $p < 0.05$)](image)

![FIGURE 7 Peel force profiles based on changing seal bar configuration](image)
temperature levels. Ground coffee and powdered sugar were applied as contaminants. As a flexible material, Film 3 was chosen based on its lower SIT. The results were evaluated via RSM, and the contour plots are shown in Figure 8. According to the contour plots, there is no change in the seal strength until a certain amount of contamination value is reached. For ground coffee particles, after around 10 g/m² contamination at the sealing area, the seal strength starts to gradually decrease with the added contaminants. This decrease can be more clearly seen when the process temperature was increased to the optimum sealing temperatures from the SIT. On the other hand, for powdered sugar, this threshold level was 2 g/m², much lower than the ground coffee particles for 0.5-s dwell time. Different material characteristics and particle sizes of ground coffee and powdered sugar can explain this variation.

Powdered sugar consists of 100% orderly arranged sucrose crystals, while ground coffee contains approximately 24% fat, 21% protein and 9% carbohydrates in addition to vitamins and minerals. This influences the thermal properties of two products. According to the literature, thermal conductivity values of ground coffee vary depending on the type of coffee and roasting level, and it is 0.182 W/m°C on average, while it is 0.126 W/m°C for powdered sugar.22,23 Higher heat conductivity of ground coffee particles might lead to better polymer melting during the sealing process. This can supply good encircling around the particles and results in higher seal strength. Besides, the main difference between the two products is their particle sizes, which are around 200 μm for ground coffee and 30 μm for powdered sugar. That is why powdered sugar was able to spread around and cover the sealing surface while there were larger distances between individual coffee particles. Thus, well-distributed sugar crystals might create more obstacles for adhesion and diffusion in the seal interface and decreased seal strength sharply.

In food packaging, food contamination through the sealing area is one of the main problems with supplying good seal integrity, and it has been known that contamination at the seal interface negatively influences seal quality and hermeticity.4,10,24 However, the threshold contaminant level affecting the seal strength was an unexplored territory in the literature. As a result, finding the threshold contaminant level for a food product is a valuable step for further researches. The packaging materials can be designed considering the risk of contamination or the sealing parameters can be arranged based on the maximum observed contamination level at the normal process to improve sealing quality and integrity. Additionally, filling mechanisms can be improved to prevent contaminations that occur higher than the threshold level. Thus, the process efficiency can be maximized.
3.6 | Image analysis

After the contaminant threshold analysis, contaminated seals were examined under the optical, digital microscope. As a result, the microstructure of interfaces at different contamination levels was imaged at \( \times 200 \) magnification with 1600 \( \times \) 1200-pixel image resolution. The images of contaminated film samples with ground coffee and powdered sugar were given in Figures 9 and 10, respectively. Film samples sealed without contamination showed full seal integrity. On the other hand, in the presence of ground coffee, monolayer films were not able to totally encircle the particles totally at process settings. At 4 and 8 \( \text{g/m}^2 \) contaminant densities, continuous sealed regions were dominant, and gap regions were only observed around single particles. Above 12 \( \text{g/m}^2 \), coffee particles started to aggregate, and when the contaminant density was increased, they created large gaps at the interface.

In the presence of powdered sugar, smaller particles covered the surface evenly, and they create more obstacles at the seal interface than ground coffee particles. Consequently, available contact points between two surfaces decreased easily, and sufficient contact did not occur above 12 \( \text{g/m}^2 \). Also, powdered sugar particles stuck to the material surface as it can be seen in the image representing 12 \( \text{g/m}^2 \) contaminant density in Figure 10.

According to the analysis of digital images, the percentage area covered up by ground coffee contaminants shows a positive correlation (\( R^2 = 0.979 \)) with the contamination density, which is given as gram per metre square in Figure 11. When the contamination density is increased from 4 to 20 \( \text{g/m}^2 \), the percentage area taken up by particles raised from 10.7% to 33.7%. This non-linearity can be explained via particle aggregations and big particle sizes of ground coffee. Also, the relationship between seal strength and contamination area shows that when ground coffee particles cover more than 20% of the sealing surface, seal strength starts to drop gradually. Because the small particles of powdered sugar cover the surface tightly for each contamination levels (0, 4, 8, 12, 16 and 20 \( \text{g/m}^2 \)), the area taken up by particles can be accepted as 100%. It was not calculated additionally via digital image analysis.

4 | CONCLUSION

This study revealed that adding 30% LLDPE compound to LDPE films enhances hot-tack seal strength significantly as well as adding 30–40% mLLDPE compared with pure LDPE. It also indicated that seal bar design has a large impact on pressure distribution throughout the seal area and influences the initial seal strength development. On the other hand, contamination of ground coffee particles and powdered sugar in a small amount around 4 \( \text{g/m}^2 \) did not affect T-peel strength at SITs for 1-s dwell time. However, in the contaminant threshold test performed at 0.5-s dwell time, the impact of contamination density, particle size and the contaminant type became
clever. When the contaminant density increased, gap areas and channels at the interface become a major problem. Also, the smaller, large amount of contaminant particles blocked the adhesion and diffusion at the seal interface, resulting in lower seal quality. In that context, this study brings the attention of researchers to the importance of determining the threshold contaminant level of different types of products for optimizing the process parameters accordingly. Further research is required to reveal the influence of seal bar geometry and contamination of different types and amounts of food products on the integrity of seal area.

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