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REVIEW ARTICLE

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Understanding the factors affecting the seal integrity in heat sealed flexible food packages: A review

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Abstract

Seal area represents the most problematic part in food packaging for controlling the moisture and gas ingress and preserving product quality. Understanding the mechanism of heat sealing, which is a widely used method in flexible packaging, is critical for maintaining product quality throughout the storage and preventing food waste. Likewise, understanding the factors causing the leak formation in the seal interface helps to avoid failures and increase integrity for all seal types produced by heat sealing methods. This review looks at heat sealing and its mechanisms in flexible food packaging materials and particularly focuses on the reasons behind the encountered seal integrity problems that have a detrimental effect on food quality and shelf life. Heat sealing mechanisms, form fill seal systems and seal types were analyzed. Then, factors affecting the leak formation have been grouped as process parameters, material properties, contaminants, and further processes, which is uncharted territory in open literature. Finally, the details of these groups and their interrelationships were examined and discussed. Revealed key factors as shown in this study are expected to guide future research for understanding leak formation mechanisms in flexible food packaging.

KEYWORDS

flexible packaging, heat sealing, leak formation, seal integrity, shelf life

1 | INTRODUCTION

Food packaging can be made from many materials, but most come from four material groups: glass, metal, plastic and paper/board. Plastics are often divided into rigid plastics like those used for caps, cups and trays and flexible plastics for lidding and wrapping films. Flexible plastics are one of the most widely used forms of food packaging, thanks to their cost competitiveness.¹ However, the flexible packaging must be securely sealed. A good sealing prevents leakage of content and its volatiles. It eliminates microbial contamination risk and also minimizes the changes in head space gases and moisture content. Therefore, the integrity of seal area has to be taken into consideration in flexible food packaging to prevent quality changes and ensure safety of food products. $^{1} \label{eq:safety}$

Sealing of flexible packaging film can be achieved by several methods. These methods can be based on melting and joining of sealant layers using heat sealing or on the application of a cold-seal adhesive coating to the inner surface of the packaging film.² Heat sealing is the most common method, which has been used for decades to shape and close the plastic packaging materials in the food industry. Common approaches to heat generation include seal bar (seal jaw) sealing, hot wire (hot knife) sealing, induction sealing, hot air sealing, pneumatic sealing and ultrasound sealing. In conduction-based heat sealing, heating can be supplied by a constant source or by impulse

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heating. With constant heating, heating pipes or wires and temperature sensors are integrated into the body of the contact equipment. With impulse heating, a resistance-based sealing band is placed on the surface of the contact equipment, which is also generally covered with a thin Teflon layer to obtain homogeneous heat distribution.^{3,4}

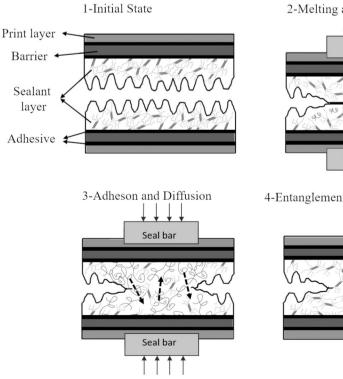
Heat sealing with contact equipment such as seal bars and seal bands is achieved by the combination of temperature, pressure and time. These parameters have a key role on the melting of the sealant film layer and the fusion of the molten surfaces. Seal quality is defined by the requirements of product sensitivity, design and easy usage requirements. Good sealing is a concept that indirectly measured via various methods such as Hot-tack, T-peel and leak detection tests. Seals having sufficient results from these tests generally meet the industrial requirements. Poor sealing, meaning the layers are not fused, and leakages, meaning small disturbances in the fused surface, are the main problems encountered with current heat sealing technologies used in flexible food packaging. Even small leakages may reduce the shelf life of products due to changes in the composition of headspace gases and moisture content. In this respect, developing better sealing techniques to maintain product quality has become a very important target for food industry.⁵

In packaging design, there are three different aspects related to sealing. These aspects are reducing material consumption, producing easy to open packages and lowering the sealing time. Each of given issue has several challenges. For example, in the first aspect, the material amount that is spent to form seals can be decreased in two ways: decreasing the seal width and decreasing the film thickness. The width of standard bar seals is around 10 mm; this can be decreased up to 1 mm with impulse sealing. However, decreasing seal width may

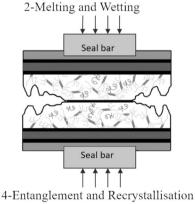
contribute to the leakage problem. Also, decreasing the film thickness to minimize material consumption may increases the risk of pinhole formation and breakages in the sealing area.^{6,7} For the second aspect. producing easy to open packages can be achieved by controlling process parameters or modifying sealant composition to increase the peelability of the sealing area. This especially offers convenient solutions for children or elderly people. Nevertheless, increased peelability is often associated with difficulty in obtaining hermetic seals.⁸ For the last aspect, lowering the dwell time increases the process output during manufacture by increasing the amount of bags produced per minute. However, providing sufficient time to supply the adequate diffusion of molecular segments across the sealing interface is critical during the sealing process.⁹ As a conclusion, all these challenging aspects require attention, and the solution of sealing problems lies in a better understanding of the sealing mechanism, good optimization of the parameters and development of new technologies or approaches.

In this paper, the mechanism of conduction-based heat sealing, its application in food packaging and the encountered problems related to the process that results with leak formation are examined to develop a comprehensive understanding of the heat sealing issues. As a method systematic review on scientific papers was conducted. Industrial guidelines and reports were also used as a secondary source for the necessary technical details.

2 | DYNAMICS OF HEAT SEALING



The parameters involved in a sealing process are basically: the melting temperature of the sealant interface, the chain diffusion rate



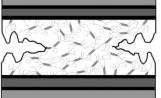


FIGURE 1 Mechanisms behind the heat sealing of flexible packages

(depending on the molecular weight and long chain branching), melt strength and crystallization rate.¹⁰ When melted and hot pressed, the two surfaces come in good contact (wetting) over the seal area, and in a fraction of a second, the chain's diffusion across the interface occurs, creating molecular entanglements mainly due to chain reputation.¹¹ After sealing, there is recrystallization of segments that holds the surfaces together and enhances the seal strength. Figure 1 visualizes each of these stages.

2.1 | Wetting and melting

Wetting is the key parameter for interface applications, and it can contribute to filling all the small gaps between two surfaces during the first milliseconds of the sealing process. The wetting behavior of a material is strongly related to its surface free energy. High surface free energy means higher interaction potential between two surfaces that come into contact. Surface free energy can be calculated based on the contact angle measurements that show the polar and dispersive interactions of a solid surface against the test liquids. When the contact angle decreases, the surface free energy and the interaction potential for the two materials increase. It is noteworthy that the contact angles of the materials are strongly dependent on their surface topography such as roughness, besides their hydrophilic, hydrophobicity properties. In addition to wetting, material immediately starts to melt during the sealing process. Even though the heat application and the melting behavior of sealants make the wetting dynamics more complex during the sealing process, calculating the wettabilities of the sealants gives a good idea on how much energy is needed to initiate the seal between two surfaces.¹² The equations that are necessary to estimate the wetting behavior of sealants are given below:

Young's equation to calculate surface free energy¹³;

$$\sigma_{\rm s} = \sigma_{\rm sl} + \sigma_{\rm l}.\cos\theta$$

Dupre's equation to calculate the amount of energy bound by wetting¹²;

$$W_a = \sigma_l + \sigma_s - \sigma_{sl}$$

 σ_s is the surface energy of the solid; σ_{sl} the interfacial tension between the solid and the liquid; σ_l the surface tension of the wetting liquid; θ the contact angle between the solid and the liquid; and W_a the work of adhesion.

Additionally, a certain amount of pressure is important during heat sealing to supply sufficient contact and enhance wetting at the interface.

2.2 | Diffusion

Diffusion is another critical step of the heat sealing process and the diffusivity of molecules along the sealant is derived by different

internal and external factors. For example, the diffusion rates of molecule chains depend mainly on the molecular weight and chain branching. Long chains are more effective than shorter chains in strength build-up at the interface.¹⁴ Additionally, the diffusion of low molecular weight (Mw) chains occurs in a shorter time than longer chains having higher Mw. The use of low Mw chains will therefore speed up the sealing process. However, these may not provide strong adhesion and entanglement.¹⁵ In the sealing of two different polymers, diffusion properties are strongly dependent on the compatibility of the two materials. When the packaging material is sealed to itself instead of different kinds of materials, interdiffusion rate will be equal to the self-diffusion rate.¹⁶ The rate of molecular self-diffusion is limited, and it stops when an equilibrium penetration is reached.¹⁷ The Rouse model for calculating the self-diffusion related to the melt viscosity is given below¹⁸;

$$D_{int} = D_{self}$$
$$D_{self} = \frac{RT}{N_a \zeta_o n}$$
$$\eta_o = \frac{\zeta_o n \rho R_g^2 N_a}{6Mw}$$
$$D_{self} = \frac{\rho RT}{6} \times \left(\frac{R_g^2}{Mw}\right)$$

 η_o

 D_{int} is the interdiffusion coefficient; D_{self} the self-diffusion coefficient; R the universal gas constant; T the temperature; η_o the zero-shear viscosity of the polymer melt; R_g the chain gyration; N_a the Avogadro number; ζ_o the monomeric friction coefficient; Mw the molecular weight of the polymer; n the number of the monomeric units per chain; and ρ the mass density of the undiluted melt polymer

Additionally, here it is important to mention from Rouse relaxation time (t_{Ro}) of whole chain and reptation time (t_r). When the dwell time (t) is in between t_{Ro} and t_r ($t_{Ro} < t < t_r$), segments start to diffuse through the interface, and when $t > t_r$, segment diffusion completed and seal region becomes one bulk. Moreover following equations gives idea on average interface distance¹⁹:

$$X_t = X_{\infty} \times (t/t_r)^{1/4} \text{ for } t < tr$$
$$X_t = X_{\infty} \times (t/t_r)^{1/2} \text{ for } t > tr$$

where X_{∞} is equilibrium interpenetration distance of monomer and (X_t) is average interpenetration distance which is half of the interface thickness.

2.3 | Adhesion

Adhesion has a main role in strength enhancement at the seal interface. Different interfacial self-adhesion mechanisms play roles in the 4 WILEY Packaging Technology and Science

adhesion of polymer surfaces: chain bonding, intermolecular bonding, wedge bonding, electrostatic bonding and vacuum bonding (Figure 2).⁵ In heat sealing of flexible packaging, adhesion takes place at a certain temperature and for a limited process time. For amorphous polymers, the adequate seal strength development depends solely on the self-adhesion and diffusion dynamics rather than the crystallization due to cooling. Also, for semicrystalline polymers, the peel strength measured immediately after the sealing via the use of hot tack test represents the adhesion strength. However, the measured peel strength will differ before and after cooling for each polymer due to forming crystals, changing toughness and rheological properties of the materials.^{15,20}

2.4 **Entanglement and recrystallization**

Entanglement in the sealing area is associated with the type, length, Mw and branch content of the molecules that diffuse through the seal thickness. After movement of molecules enhanced by the heat, attraction forces will appear between the molecules at the new interaction points. Entanglement starts from the initial stages of sealing and continues through the cooling process in the melt state until the molecular movement slows down. The density of the chain entanglement has a crucial effect on the final seal strength.²¹ Additionally, for semicrystalline polymers, the sealing process involves the recrystallization step in addition to the other main sealing mechanisms such as melting, wetting, interdiffusion, adhesion and entanglement. During recrystallization, the rate of cooling plays an important role on the crystal growth. With slow cooling, larger crystals occur in smaller amounts. In fast cooling larger amounts of smaller crystals appear through the sealed area, and the mechanical properties will be affected.²²

DEFINING THE SEAL TYPES 3

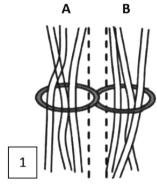
Different types of heat seals can be created using flexible packaging films. The choice of the seal type depends on the type of the packaging material and the requirements of the packaging design. Based on the peelability function of the sealant layer, seals can be grouped into two different types.

Lock seal 3.1

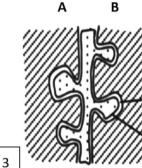
Lock seal contains very strong interface interactions. This type of seal is generally preferred in hermetic sealing. Vacuum vegetable packs in which the products are further processed or sausages that are boiled in the package can be given here as an example. To open the packaging must to be cut or torn.

3.2 Peel seal

Seals that are openable by a certain amount of pulling force after the heat sealing without causing any damage to the packaging material are referred as peel seals. By controlling the process temperature or



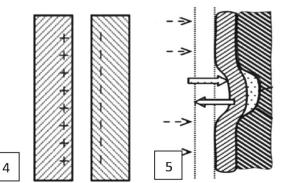




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FIGURE 2 Adhesion in microscale. (1) Chain bonding, (2) intermolecular bonding, (3) wedge bonding, (4) electrostatic bonding and (5) vacuum bonding⁵

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altering the material composition by adding incompatible substances, weakly bonded, easy to open seals can be obtained.⁸ A peel seal can be in either a bag or tray configuration. There are a number of technologies that can be employed to achieve an optimal peelable seal. The most commonly available three peelable technologies are controlled contamination, dissimilar resins and controlled delamination. Each has its own advantages and disadvantages. Gorny and Brandenburg²³ describe the basic attributes of the three technologies.

- 1. Controlled contamination: This technology utilizes a small amount of contaminant resin within the sealant layer to achieve a controlled weak seal or 'peel seal'. Common contaminant resins are various grades of polybutylene. This technology is used predominately for applications where the film is peelable to itself. Through engineered blending technology, the film can be designed to be peelable within a specific temperature range. This results in peelable areas and non-peelable areas within the same finished packaging material.
- 2. Dissimilar resins: This technology utilizes a dissimilar resin between the peelable lidstock and the bottom tray stock. This technology is most commonly used in lidstock applications where a peelable lid is sealed to a rigid or flexible thermoformed bottom web or rigid container. The choice of peelable sealant layers is directly dependent upon the sealant layer of the bottom material. Whenever possible, the top and bottom sealant layers should be designed together so the optimum match is made, and the desired peel seal is achieved.
- 3. Controlled delamination: This technology involves achieving an initial fusion seal between the sealant layers of the lidstock and the bottom material. The 'peel seal' is then achieved through the delamination of the coextruded sealant layer. The delamination within the coextrusion occurs between the sealant layer and the next inner layer (often the core layer). It is possible to achieve a frosty or 'tamper evident' seal, as well as a resealable structure. This technology is often found in medical packaging.

Seal types can also be categorized depending on the folding geometry as fin seal and lap seal. Offset fin seal and pinch seal are two different variance of standard fin seal. Those seal types were shown in Figure 3. Dashed line in the figure represents the midline of the packaging design in horizontal configuration;

3.3 | Fin seal (fold-over seam)

Both interior edges having the same properties of a flexible film are sealed together at the fin sealing. Then the seam is folded right over left or left over right. When the folding corner of seal overlaps with the midline of the packaging design, offset fin seal; when the seal area is left as unfolded, pitch seal will be obtained as different variants of fin seal.

3.4 | Lap seal (overlap seam)

In this sealing type, the external surface of the film is sealed onto the internal side of the same sheet. Although the film needs to have sealant on both sides for this sealing application, a lap seal uses less material surface than a fin seal.

In pouch production, two flexible packaging films can be sealed to each other in different positions and directions. Definitions of the sealing types according to their process direction can be found below.

3.5 | Transverse sealing

The transverse sealing, or cross sealing, the packaging materials are sealed in a transverse direction, which is perpendicular to the direction in which the packaging material is conveyed. The transverse sealing jaws operate as a pair in a horizontal direction for vertical form fill and seal (VFFS) systems and in a vertical direction for horizontal form

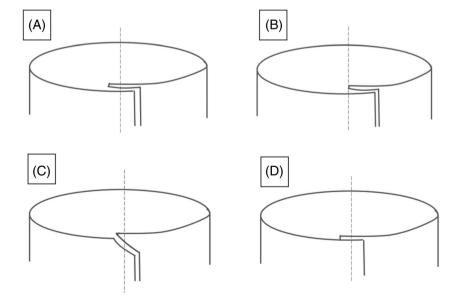


FIGURE 3 Examples of seal configurations. (A) Fin seal, (B) offset fin seal, (C) pinch seal and (D) lap seal

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fill and seal (HFFS) systems to shape the top and the bottom seals of plastic pouches. While the transverse seal jaws only open and close in the intermittent packaging systems, some jaws can also move along the machine direction to be able to achieve specific dwell time without slowing down the process in the continuous systems.²⁴

3.6 Longitudinal sealing

In this type of sealing, the sealed edge will be parallel to the direction of the film movement. The side seals are produced by longitudinal sealing. In this sealing method, a single, constant seal jaw allowing film movement by pressing the film against the forming tube with just enough pressure is generally used. Also, rotary wheels having equal angular velocity with the moving film speed in the continuous systems are popular. Additionally, in intermittent systems, timed, press release seal jaws are preferred to prevent the packaging material from overheating.²⁴

Additionally, flexible lidding material can be sealed to the top of the trays. A rigid tray and lid combination can be designed to create a peel or a lock seal. Therefore, compatibility between tray and sealant layer of the lidding film is critical. Also, one of the most important packaging parameters that must be considered is the selection of the sealant layer polymer and configuration. Choice of the correct sealing polymer and format is dependent upon operating parameters including package machine type, filling speed, package configuration, seal configuration and product type and its weight (mass). Potential sealant layer polymers exhibit a wide range of seal characteristics. Common choices of polyolefin sealant polymers include low-density polyethylene (LDPE), ethyl vinyl acetate, ultra-LDPE and plastomer metallocene. Each polymer has its own sealing characteristics including ultimate seal strength, hot tack strength, seal initiation temperature, ability to seal through contamination and oxygen transmission rate. Careful consideration should be given in order to optimize the specific polymer characteristics to the specific sealing requirements of the package.²⁵ To include all necessary properties other functions of the lid film such as giving a view of the product by transparency, providing a surface for labelling, or being printable for information provision may be considered.

PACKAGING SYSTEMS AND 4 | ENCOUNTERED HEAT SEALING PROBLEMS

Flexible packaging films can be shaped and filled with product and sealed by different packaging systems according to the specific design requirements. They can be sealed to itself or to the different materials such as on top of the plastic trays, plastic bottles, metal cans and glass containers. The different flexible packages produced by these packaging systems are given in Table 1. According to process direction, there are two main systems, namely, VFFS and HFFS systems. In these two techniques, packaging materials are formed from a web of film into pouches in different sizes and shapes. Then, they are filled with the product and sealed. For the sealing of other kinds of packaging such as trays and plastic bottles, thermoform fill seal (TFFS) system can be used. There are also systems only for applying direct sealing of the flexible top lid to glass and metal containers. All these systems supply different advantages depending on the package design and other production requirements. They can process based on either intermittent or continuous motion principles. The details about the form fill and seal (FFS) systems are given in the following sections.^{3,26} Also, Figure 4 demonstrates the single line VFFS, HFFS and TFFS systems.

4.1 VFFS system

In the VFFS system, the food content flows vertically down into the package during filling. Generally, powdery, granulated products or liquids are packaged with VFFS machines. These machines start packaging with varying sizes of film roll. The film web is usually pulled by the transport belts to the forming tube, which forms the film into a tube shape. In some VFFS models, the sealing jaws themselves grip and pull the film. Then the film is folded around the forming tube that was arranged to make a lap or fin seal at the longitudinal side of the package. During this process, the amount of film tension controlled by dancer arms and the film position is controlled by sensors and constantly corrected. VFFS machines can work with single line or multiline processing options and need less floor space than horizontal machines. However, certain package types can only be produced on horizontal machines. However, nowadays, doypacks can also be made on a vertical machine by filling them from the side. Offering the possibility to make pack types on a vertical machine has the advantage of less floor spacing, increasing the range of package design with VFFS with these small modifications.²⁶⁻²⁹

4.2 HFFS systems

In the HFFS process, the product is not dropped from above; instead, it is slid into the package horizontally or placed on the packaging film. This type of packaging is generally applied to the individually packaged granola bars, cookie trays and candies. A particular advantageous of HFFS machines is their applicability for irregular shaped products. A widely used horizontal process is flow wrapping. In this kind of process, either rotary jaws or horizontally moving long dwell sealing jaws are preferred for sealing and cutting of the top and the bottom edges to produce different variations of pillow bags. Additionally, the bottom gusseted doypacks and three sided sachets can also easily be formed using horizontal systems.^{26,30}

4.3 TFFS systems

In the TFFS technique, a package can consist of two separate materials. While one of these materials is shaped into a cup or tray by thermoforming the other flexible film is used for producing the top lid. In TFFS, after the package is filled with the content, the barrier lid film is placed on top by heat sealing.³

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TABLE 1 Types of flexible packages^{24,29}

Package type		Usage	Package type		Usage
	Twist wrap	Packaging of bread, chocolate, candy and so forth. It can be one or two sided.		Flat bottom bag	Packaging of beans, grains and flour suitable for heavy products
	Three- to four-sided sachet	One portion product such as seasonings salad dressings and soup powders		Quad seal flat bottom bag	Coffee bags give more stable design and more printing area than the flat bottom.
	Pillow bag	Chips, crackers and snack foods are usually packaged with pillow bags.		Brick pack	Used for long-life products, such as UHT beverages with spouted option
	Gusseted bag	Packaging of breakfast cereals, confectionaries, gives printing area.	• •	Doy pack	Widely preferred for beverages, granulate products and pastes with zipper option
	Tetrahedron bag	Used for UHT beverages, takes less space than the brick packs		Spouted pouch	Used for beverages, it can have a customized shape
	Lid film	Closing the yogurt, pudding cups, ready to eat meal trays		Square bottom bag	Packaging of powdery food mixtures and herbal teas
	Shrink label	Used for UV protection or labelling of beverages		Stand cup	Packaging of highly viscous liquids such as ketchup and honey. Cups take less storage space.

Abbreviations: UHT, ultrahigh temperature; UV, ultraviolet.

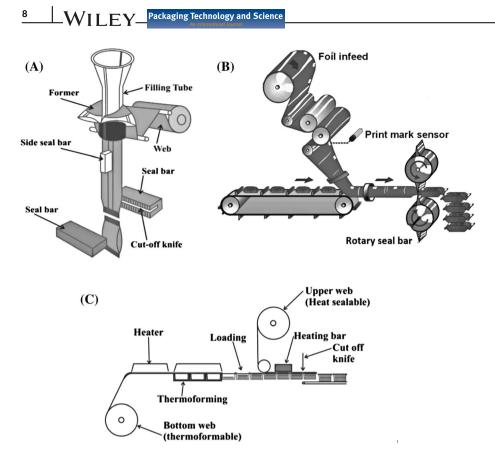


FIGURE 4 Single line (A) vertical form fill and seal (VFFS), (B) horizontal form fill and seal (HFFS) and (C) thermoform fill seal (TFFS) systems^{26,31}

A weak point of barrier packages are the sealed seams. O2 can permeate through the sealing material into the package. In the case of food packaging using barrier materials and modified atmosphere packaging (MAP), this cross diffusion is negligible. However, sealing defects such as damaged barrier layers at the sealing zone and pinholes in the sealing layer are more critical. These enable permeation and in the worst cases free diffusion of increased amounts of O₂ into the package.³² These defective packages need to be filtered out by quality control procedures to avoid consumer complaints due to oxidation-based food deterioration. Sampling-based destructive leak detection methods such as dye leak detection, immersion biotesting and electrolytic test are not sufficient to supply total process control.33 Non-destructive methods for leak detection are necessary for industrial packaging processes. Most relevant for the application in combination with filling machines are gas leak detection methods showing detection limits between 10 and 25 $\mu m.^{34}$ Other nondestructive methods are inspection-based systems such as ultrasonic imaging and infrared thermography, which are not widely used so far.^{35,36} Considering a machine output of 60 packages per minute³⁷ for the single line production, pinholes with a diameter less than 10 μ m are particularly critical because they cannot be detected at this rate with standard leak testers on the basis of CO₂, He or H₂ detection.³⁸ Package integrity can refer to seal quality and/or presence of pinholes in the packaging material. If the seal is not good and pinholes are present, moisture vapour can get in and out of the package. If there is moisture gain, the products like cereal can become stale and leathery. If there is moisture loss, fruit additives, for example, can dry out and become hard. The lining film must fulfil all other product protection requirements. It must be flexible enough to withstand the packaging line without puncturing but rigid enough to provide seals without wrinkles, which could allow moisture to penetrate, and without product crumbs, which can also affect seal integrity. It must resist the penetration of water vapour and the transport of gases or vapours in either direction.³⁹ Moreover, creases and wrinkles in the sealing surface caused by the manufacturing process can act as capillary channels that may cause leaks in the package. Previous studies have shown that the minimum sealing pressure of lidding film that results in a leak proof seal within the materials investigated was 1.8 N/mm² to fill the wrinkles in the sealing surface. The depth of crease that can be sealed in a leak proof manner was found to be up to 150 μ m.⁴⁰ Also, literature highlights that several types of multilayer films, including bioriented polyethylene terephthalate/polypropylene (PET/PP) bilayer films, aluminum foils and metalized layers, exhibit delamination phenomena.41 Delamination phenomena frequently occur when multi-layer flexible polymeric films are employed for high-pressure treatments of food packaging for pasteurization and sterilization purposes. The theoretical results highlight the crucial role played by the mismatch of Young's moduli and Poisson ratios of the laminated film sheets in promoting delamination.42

5 | FACTORS BEHIND THE HEAT SEALING PROBLEMS

During the heat sealing, multiple factors affect seal strength and integrity development. These factors can be grouped as process

parameters, material properties, contaminants and further processing. For example, sealing quality is strongly dependent on the main process parameters: temperature, time and pressure.^{5,9} By using the correlations between these parameters and precise controlling, different types of seals such as lock seal or peel seal can be obtained, energy consumption can be decreased or high-speed processing can be achieved without even changing the material type.⁷ However, poor optimization of these parameters may cause many problems, which eventually results in poor sealing and leakages. Even though the effects of some process parameters such as time, temperature and pressure on the seal quality are well studied, the roles of other parameters such as contaminants, seal bar design and cooling rate on the formation of gaps and channels at the sealing area are still unknown. As a result, understanding the factors/mechanisms behind the formation of gaps and channels that results in leakages is critical to prevent food waste.

Based on the literature, ontology has been prepared to indicate the various factors causing the leakages at the heat sealing area of plastic food packages. The factors that are more intensely researched and the areas that require more study were indicated with the changing color intensity in Figure 5, and each factor was explained briefly in the following sections.

5.1 | Process parameters

5.1.1 | Temperature

Plastic film materials start to show seal behavior after a certain temperature level called 'seal initiation temperature'. The temperature range between the seal initiation temperature and the maximum temperature that can be applied without causing any damage to films is named as the 'process window' or the optimum sealing temperature range. The process window is determined by a hot tack test, and it includes the plateau region in the temperature versus hot tack strength graph where the maximum and consistent seal strength is obtained. If the sealing temperature is low and outside the operating window, the sealant will not sufficiently melt and full seal integrity cannot be achieved.43,44 A certain amount of flow behavior in the sealing area is desirable during the sealing to fill all the voids and gaps and prevent the leakages and polymer molecules get more movable around their melting temperature. So, when the temperature is increased and becomes close to the melting temperature, the viscosity of the sealant layer decreases; thus, diffusion between sealing surfaces enhances.⁴⁵ On the other hand, when the seal temperature exceeds the operating temperature range or it goes to the high end of this window, the squeeze-out amount increases exponentially due to the exponential decrease in the melt viscosity of sealant. If the amount of squeeze-out increases, adequate sealing cannot be supplied and the defects occur in the sealing region.⁴⁶ That is why using sealants that have a wider process window or applying a precise temperature control is critical to prevent sealing failures.

5.1.2 | Dwell time

As a dominant sealing variable, time helps to increase the area of the sealed film from the first stage of sealing, until full sealing integrity is obtained. Because the processing time is a cost related factor,

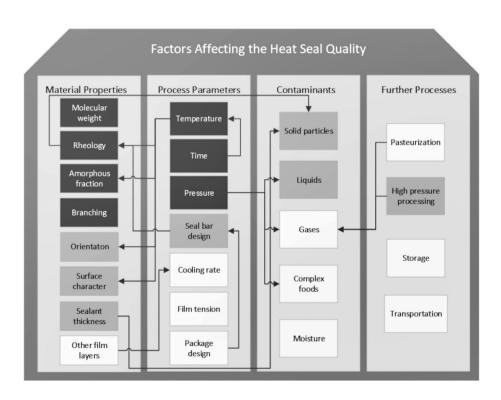


FIGURE 5 Ontology on the factors behind poor seal integrity

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determination of the minimum dwell time for the sealing process is an important issue. When the required interface temperature to obtain a good seal is known, heat transfer calculations can be made by considering the film thicknesses, different layers and thermal conductivities of the materials.⁴⁵ Also, within the optimum sealing temperature range, seal strength increases as dwell time and temperature increase because it enhances the formation of entanglements.⁴⁷ According to Najarzadeh et and Ajji,⁴³ there is a linear correlation between the strength of the seal and the square root of dwell time, and the slope of the $t^{1/2}$ versus seal strength graph will increase when the sealing is performed at the higher temperatures. This means the maximum seal strength is obtained faster at higher temperature levels. Inconsistently, an empirical model developed by Morris indicates that the temperature where the plateau seal strength is reached in hot tack tests will decrease when the dwell time is increased.⁴⁸

5.1.3 | Pressure

Many studies indicate that any increase above the required level of sealing pressure, which is adequate to supply full contact between layers does not affect the sealing temperature and sealing strength significantly.^{9,45,49} Also according to Najarzadeh and Ajji, increasing the seal pressure from at a very low level such as from 0.1 to 0.5 N/mm² will decrease the seal initiation temperature. In that range, increased pressure provides better contact between two film layers. However, above that level, seal initiation temperature does not show any chance. The reason behind it might be the decreasing free volume under compression that can block the chain movement. So, at the interdiffusion stage, increasing the pressure will not help the entanglement and mechanical strength development in sealing.⁴³ In addition, a research was done by Farris et al. based on response surface methodology (RSM), showing that seal strength is affected negatively by increasing the bar pressure and the positive effect of temperature will be disrupted at high-pressure levels.⁵⁰ Also, the elevated temperatures enhance this negative effect of high pressure by decreasing the polymer viscosity critically. As a result, squeeze-out increases linearly with increasing pressure because the extra force will push out the molten sealant easily from the sealing area.⁴⁶

5.1.4 | Seal bar design

The profile of sealing bars affects the seal quality based on bar width, segregation pattern, and type of material. As the seal bar width decreases under the constant jaw force, applied seal pressure per area will increase. In that case, smaller seal bar width leads to more squeeze-out problem. Also, serrated seal bar geometries can create additional shearing action compared to flat surfaces. This may help the sealant to fill all the gaps and voids in seal interface as a theory. However, sometimes there will be a relationship between the viscosity of molten polymer and shear rate. In that case, increasing the shear rate via changing seal bar design can cause a dramatic decrease

of polymer viscosity, and that may raise the squeeze-out problem at the seal interface.⁴⁶ Also, different crimp angles might alter the pressure distribution and create different stretched regions within the sealing area. Even if this factor has not been clearly understood yet, some studies are pointing to its importance. For example, according to Matthews et al., with seal bar designs having the crimp angle more than 90°, sealing starts from the flat sides of the serration pattern, but with bars having a smaller crimp angle such as 60°, sealing starts from the peak points of the crimps at the seal initiation temperature. The study also revealed that crimp angles smaller than 90° offer more potential to gain higher seal strengths for biopolymer films in a 25- to 45-µm thickness range. However, when the film thickness is outside of this range, higher crimp angles above 90° are more efficient in maintaining high seal strength.⁴⁷ Additionally, one of the earliest studies on the effect of seal bar geometry suggests that the direction of the serration pattern is also important in creating leak resistance. According to the results of the study, packages heat sealed with the seal bars having crosswise segregation pattern show better drop resistance than ones sealed with lengthwise or inclined segregated seal bars which are also superior to the packages sealed with flat bars.51

5.1.5 | Cooling rate

Crystallization always induces changes in the mechanical properties of the materials together with some of the characteristic properties such as concentration, density and thermal behavior. As a result, controlling the cooling rate that influences crystal growth may help to enhance seal strength.⁵² Supporting this, a recent study revealed that films showing a faster crystallization behavior had higher hot tack strength at temperatures lower than the temperature that supplies total melt of the crystals. This means that at the low temperatures, the seal strength will be enhanced by solidification due to quickly starting recrystallization. However, when the polymer shows a slower crystallization behavior or when the applied seal temperatures are much higher than the crystallization temperature, the hot tack seal strength will be more related to the interdiffusion rate than the solidification.⁵³ Also according to the study of Simanke et al., between two metallocene linear LLDPE fractions, it can be observed that the sample having a higher crystallization temperature shows better hot tack performance than the other one. This feature can be attributed to the immediate recrystallization of the m-LLDPE molecules after the sealing process, because of its higher crystallization temperature.54

5.1.6 | Film tension

According to process guidelines, nonuniform film tension along the film width is the main source of wrinkles and misalignments as indicators of poor seal quality.^{55,56} In the packaging process, pulling force is applied to the film web to draw it through the bagging system.

In some cases, such as in continuous systems sealing is performed when the packaging film is moving with a certain film tension. However, in intermittent motion systems, generally, only the longitudinal seal is performed when the system is still moving while the top and bottom seals are made when the system has stopped. Additionally, in VFFS systems, if the bottom bag is not supported during the sealing, the mass of the product filled into the package creates a tension that pulls the bag down under the effect of gravity. Moreover, it has been known that there is a small amount of shearing and stretching that occurs in the sealing area due to the patterns of grooved seal bars.^{46,47} Even though the film tension is an unavoidable part of the packaging process, its effects at molecular level on seal formation mechanisms are still unknown.

5.1.7 | Package design

The design of the food package is one of the factors affecting seal quality. During the shaping process, the packaging film is folded in different directions based on the design requirements. In flexible films, at the intersection of longitudinal and transverse sealing two layers before the T section can be folded up to four layers thick at the gusset areas depending on the packaging design. When the layer jump occurs, the rigid seal bars cannot apply equal pressure to the film through the folded and unfolded areas where the thickness change occurs. As a solution, flexible sealing jaw faces are used in the industry. They can accommodate the extra pressures formed by the side seal and gussets.⁵⁷ Another problem is the leakage risk at interaction points between two different seal direction (transverse and longitudinal). Two consecutive heat applications in these regions may weaken the seal by squeeze out and delamination.

5.2 | Material properties

5.2.1 | Molecular weight (Mw)

In food packaging, there are a variety of polymer types used in flexible packaging materials, and material properties such as Mw of each material, especially of each sealant layer, have a crucial effect on the sealing process. Based on the formulas given for self-diffusion, it can be concluded that there is a negative correlation between molecular weight and chain travel.¹⁸ However, as the Mw increases, higher adhesion and stronger seals are observed. This is because the high molecular weight chains are more effective in enhancing interfacial adhesion comparing to the low molecular weight chains. Additionally, the narrow molecular weight distribution supplies a more homogeneous structure. Between two versions of the same polymer, the one having a wider Mw distribution will have a lower seal initiation temperature. However, that kind of polymer may create difficulties in determining the optimum process parameters to achieve full seal strength and result in lower adhesion strength during sealing.15

5.2.2 | Rheology

The viscosity of a melt sealant decreases with increasing temperature. When the sealant becomes too fluid, squeeze-out occurs and the sealant will be pushed away from the seal area easily by the applied excessive pressure. Nonetheless, the rheology of melt polymer during the sealing can be arranged by changing the process temperature depending on the polymer characteristics.⁴⁶ As an example, Morris⁵⁸ indicated that ionomers show less squeeze-out than the plastomers at low sealing temperatures. This feature is coherent with the high viscosity of ionomers at low temperatures and low shear rates. However, the amount of squeeze-out of the ionomers rapidly increases with temperature. This is because the viscosity decreases in ionomers occur faster than the plastomers. Consequently, the characterization of melt viscosity or melt flow index of the sealant for the certain process conditions is decisive in preventing overheating and squeeze out. Moreover, another study that measures the melt toughness of sealants from areas under the stress strain curve at the sealing temperatures suggests that the materials having higher melt toughness will have a larger process temperature window. This is because they can maintain their strength in a wider temperature range. This supplies advantageous in processing by decreasing optimization failures.²⁰

5.2.3 | Amorphous fraction

In semicrystalline polymers, sealing will occur around the crystal melting temperature, because the mobility of macromolecules increases around the Tm level. For those polymers, the sealing temperature determines the exact crystalline and amorphous molecule ratios of sealant during the sealing process.54 Studies show that the seal strength and hot tack strength are strongly correlated with the amorphous molecule ratio of melt polymer at a certain sealing temperature because the unmelted crystal macromolecules create obstacles against the chain diffusion.⁴³ Also, knowing the melt distribution of sealant at a certain sealing temperature helps to predict the seal guality and understand the number of available chains for diffusion and the ratio of obstacles. For example, the heat seal initiation temperature (HSIT) is obtained when the surface temperature reaches 77% amorphous fraction for polyethylene and around 60% for PP homopolymers.¹⁴ Also as Stehling and Meka revealed, in various semicrystalline polyethylenes, the minimum required amorphous molecule ratio to achieve sufficient seal strength is between 75% and 80%. After reaching this amount, the seal strength increases with the increasing amorphous molecule ratio. The weight fraction of amorphous molecules $f_a(T)$ can be predicted via the heat of fusion of the materials from the equation below, where the Δ Hs heat of fusion of sample Δ Hu is the heat of fusion for a 100% crystalline polymer and Δ Ht is the total heat of fusion at a certain process temperature.¹¹

$$f_a(T) = 1 - \frac{\Delta Hs}{\Delta Hu} + \frac{\Delta Ht}{\Delta Hu}$$

5.2.4 | Branching

Results of earlier studies indicate that the type of molten chains is more important than the theory between amorphous fraction and seal strength. Because the chain branching is directly associated with the movement ability of molecules through the molten media, it affects the diffusion rate under certain process conditions. According to Najarzadeh and Ajji,¹⁵ LDPE samples containing highly branched molecules result in very weak seal bonding due to the inhibition of interdiffusion. On the other hand, small molecules having short chain branches can diffuse easier. Further, for the semicrystalline polymers long linear branches are part of the formation of crystals and determine the density of crystalline links that provide strong seal structures.²¹

5.2.5 | Orientation

Heat sealing studies conducted with oriented sealant materials indicate that, when the orientation degree is increased along the seal width by cutting the specimens parallel to the machine direction as it is shown in Figure 6A, the maximum seal strength of packaging materials will be affected positively. However, this makes the material more rigid and brittle at the seal area compared to the specimens having more molecular orientation along the seal length, as given in Figure 6B.⁵⁹ On the other hand, in oriented monolayer films such as oriented polypropylene (OPP), it can be observed that during sealing the molecular orientation ratio is high at low seal temperatures and drastically drops after the temperature reaches a certain level. Above that level, low tensile strength is observed. According to the literature, this phenomenon can be related to the relaxation behavior of oriented molecules with increasing temperature levels. This relaxation of orientation is accompanied by a decrease in length (shrinkage) and a simultaneous increase in the thickness of the sealant layer. That feature is not observed in cast polypropylene (CPP) films, which show only a decrease in sealant thickness during the sealing process at high temperatures.^{4,59,60} As a result, preserving the molecular orientation of oriented films during heat sealing is important to prevent relaxation and thickness change that may decrease problems in heat transfer efficiency in seal strength.

5.2.6 | Surface characteristic

There are two main property related to surface characteristic of plastic films: surface free energy and surface roughness. Surface free energy or in other words surface tension of the films directly affects the wetting stage of the heat sealing. Higher surface energy enhances the ability of plastic materials to adhere to other plastic surfaces, between the test liquid and the surface.⁶¹ However, most packaging films are hydrophobic and have low surface free energy. That is why some surface treatments such as corona discharge treatment have been applied to the surfaces of the packaging materials to increase their surface free energy, surface treatments will increase surface roughness, and high surface roughness enhances wettability and adhesion performance.⁶²

5.2.7 | Sealant thickness

According to a study by Guo and Fan,⁶³ the influence of sealant layer thickness on the heat seal strength is significant in the barrier films, because heat seal strength consistently increases with increasing sealant layer thickness above the seal initiation temperature. However, the lowest limit of sealing temperature range increases with increasing sealant layer thickness. On the other hand, higher sealant thickness leads to higher squeeze-out problems according to a model predicting the squeeze-out amount during heat sealing.⁴⁶ Additionally, for packages containing faultless aluminum barrier layers, only permeation pathway for moisture and gases will be the sealing layer. In that situation, gas and moisture transmission rate of sealant and the dimensions of the permeation area again gains importance.⁶⁴ It also needs to be considered that contaminated particles, joints and corners are the most critical points in sealing. Sealant thickness must be enough to fill the tiny gaps at these points.⁷

5.2.8 | Other film layers

Packaging films may contain many different layers that supply various features and the main layers are sealant, barrier, printing and adhesive layers. According to the studies, some food components can be largely

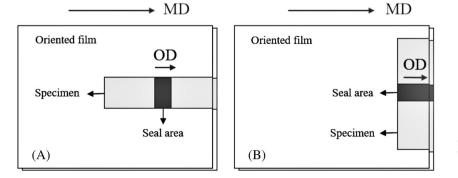


FIGURE 6 Changing the orientation direction at seal area (A) high orientation along the seal width and (B) high orientation along the seal length⁴⁹

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absorbed through these polymer layers. This can weaken the adhesive bond strength between layers and even cause delamination. For example, Nielsen et al. revealed that adhesion strength between the aluminum barrier layer and sealant polymer decreases 50% after a small amount of β -carotene absorption during 8 weeks of storage.^{65,66} Following this information, the quality in the seal area may also decrease in laminated films under certain circumstances. On the other hand, Planes et al. suggest that the seals obtained from multilayer films containing stiffer layers induce an improvement of the mechanical seal properties as compared to the seals obtained from sole monolayer flexible sealant material.⁶⁷ Additionally, when there are other film layers, cooling rate may dropdown depending on the layer thicknesses.

5.3 | Contamination at seal area

5.3.1 | Liquid contaminants

Liquid food contaminants in the sealing interface can cause a heat sink effect which decreases the heating efficiency. On the other hand, a study published in 2012 suggests that, when water replaces the air gaps in the sealing area, a heat sink effect is not observed, because the water molecules replace the air gaps and the heat conductivity of water is much higher that of air. As a theory, that property counterbalances the heat sink effect. Additionally, in the presence of liquid contaminants, increasing the seal bar pressure usually can push out the liquid between the layers and helps to form better contact at the sealing area.⁶⁸

5.3.2 | Solid particles

Solid particle contamination may cause microchannel formation and poor integrity throughout the sealing area. In case of solid contamination, melt viscosity of the sealant gains importance. Low melt viscosities help to prevent the formation of gaps around the particles by encircling them.⁵ Additionally, static charge or polarity at the packaging surface can attract the small solid particles to the sealing area. To prevent particle contamination, the static attraction between packaging material and food particle needs to be eliminated by changing the surface properties.⁶⁹

5.3.3 | Moisture and gas

According to the literature, gases present in the polymer may act as a plasticizer and result in changes in polymer crystallinity under pressure.⁷⁰ In MAP and in the packaging of carbonated beverages, gas molecules can be easily absorbed by sealants at different ratios than the normal atmospheric conditions. Additionally, it is expected that the moisture content in the headspace of packages will increase under certain conditions or processes such pasteurization. Under the effect

of temperature, more moisture can get absorbed through the sealant and seal quality might be affected. Humidity conditions and moisture content of the film during the sealing also have a role in sealing performance. A study by Suh et al. revealed that when starch-based films are pre-conditioned at different relative humidities, films with high moisture content show better adherence. Stronger bonding at high moisture content can be explained by the increased molecular mobility via water molecules showing the plasticization effect.⁷¹

5.3.4 | Complex food matrices

Food products generally consist of the combination of different type of molecules, such as proteins, fats, carbohydrates and water. During the sealing process, it has been reported that the denaturation of contaminated protein molecules or melting of fats can affect the interface temperature and seal integrity by decreasing the efficiency of heat transfer.⁶⁸ However different types of molecules may affect the sealing dynamics differently. Additionally, the effect of absorbed gases such as those in MAP, carbonated beverages and volatile compounds needs to be taken into account. To understand the overall effect of different molecules that contaminated through the seal area, there is a need for more real food contaminant testing.

5.4 | Further processes

5.4.1 | Pasteurization

Pasteurizing and cooking and retorting of the packaged and sealed products is a widely used process in the food industry. There are several types of food products preserved by this techniques called in-pack pasteurization or in-pack cooking. At the pasteurization stage, applied heat can affect the sealing quality. Also, during the pasteurization CO_2 concentration and moisture concentration at the headspace of the packages highly increase and the sealant layer can easily absorb that CO_2 and moisture. However, the effects of this situation on seal strength are unknown.

5.4.2 | High pressure processing

High pressure processing (HPP) is applied to the packaged and sealed products as a food preservation method that minimizes the sensory and nutritional quality losses. However, under high-pressure, delamination, the formation of wrinkles, holes or changes in the mechanical or barrier properties of the packaging material is expected. So, the strength of heat seal areas is critical in maintaining the package integrity during the HPP process because the sealing areas must resist processing pressure.⁷² The other aspect of the mechanism behind the irreversible deformations on packaging materials during the HPP is the quick expansion of absorbed gases within the layers under

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pressure.⁷³ Also, another issue with the HPP is the localized increase of crystallinity in polymers. Because the crystalline phase is denser than the amorphous phase, this phenomenon may lead the changes in barrier properties of polymeric materials.⁷⁴

Storage and transportation 5.4.3

There is a possibility that the time factor and environmental conditions change the seal quality. However, in the literature, there is not sufficient study on those factors. On the other hand, one study conducted by de Oliveira and Faria⁵¹ showed that drop resistance of 250-g ground coffee filled packages will change after a transport simulation. This change is also affected by the seal bar profile. For instance, the packages that sealed with the seal bars having a crosswise serration pattern had shown better drop resistance than those sealed with the flat or lengthwise serrated seal bars. However, transport simulation does not show any significant effect on seal quality independently from the seal bar geometry.⁷⁵

FUTURE PERSPECTIVES 6

In the literature, history of heat sealing studies starts in 1994 and the amount of studies stays limited with roughly 10 publications per year until today. The topics of those publications are also limited with characterization of sealing behavior of various commercial or treated flexible packaging materials and optimization of main process parameters. Based on the available information from the literature, future studies on seal quality might focus on three aspects:

- 1. Completing the information gaps in the literature
- 2. Characterization of sealing properties of newly developed packaging materials
- 3. Innovation of better sealing applications

As it presented in this study, the gap areas in the literature mentioned in the first aspect mainly consist of lack of knowledge on the influences of factors other than temperature, time, pressure and certain material characteristic on seal guality. Future researches on the influence of neglected parameters, such as contamination, seal bar design, stress on films, cooling rate, and post packaging processes can bring solutions to the question of how to prevent leak formation phenomenon at the interface.

The second aspect is related to the dynamic shift towards more environmentally friendly packaging solutions in food packaging industry. Current changes include innovations of better recyclable materials such as multilayers delaminating on-demand or thicker mono materials.⁷⁶ Additionally, to reduce the energy consumption in the process chain, oriented PE films are being developed to replace OPP films having a higher melting point. Especially high density biaxially oriented polyethylene (HD-BOPE) films show promising features also in production of recyclable, multi-layer, mono material packagings.⁷⁷ Besides, replacing petroleum-based packagings with bio-based and/or biodegradable alternatives such as bio-monomer, cellulose, or polylactic acid (PLA) based films as well as searching for alternative fillers to lower the CO_2 footprint is gaining priority.⁷⁸⁻⁸⁰ In the future, more study is expected on characterizing and optimizing their sealing behavior considering that the seal performances of some of these alternative packaging are lower than commercial materials. To facilitate the development of a better sealing mechanism on these new materials, the relationships between various factors affecting the seal quality needs to be clearly defined and heat sealing process needs to be modeled. Then, the behavior of the material can be characterized under the effects of the determined critical factors.

Finally, the third aspect is developing leak proof, efficient sealing systems that can also include in-line seal inspection technologies. Gaining better control over various parameters behind the leakage problems can open the doors to achieve less heat energy and material consumption during seal application. From that perspective, ultrasonic sealing can be improved as an alternative to conduction-based heat sealing technology, with its promising lower energy and material consumption.⁸¹ In food processing, after the required improvements and precautions were applied, it is still important to monitor and identify sealing defects to ensure the safety at critical control points. Simple imaging techniques identifying some seal defects like misalignment are generally insufficient to detect small leakages. On the other hand, advanced techniques such as thermal, ultrasonic, infrared or X-ray imaging require more development.^{82,83}

The main study directions to achieve and ensure sufficient sealing considering the industrial requirements were summarized in this section. In conclusion, there is a lot of room for improvement of sealing processes of flexible packagings.

7 CONCLUSION

In this review, heat sealing mechanisms, form fill seal systems and seal types in flexible packaging were reported. Then, the factors affecting leak formation were particularly examined. These factors have been grouped as process parameters, material properties, contaminants and further processes, which were not structured in the open literature before. With this grouping and evaluation of various factors from a broad perspective, a clear road map for future studies on the seal integrity problems was created in Section 6. As the main finding of this study, it is discovered that there are various other factors are affecting the sealing process in addition to main process parameters (temperature, time and pressure). Because supplying sufficient seal integrity is a prevalent issue in food packaging because leakages reduce product quality, those factors require more attention to prevent small leakages, which decrease the shelf life and create food safety issues.

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