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REVIEW ARTICLE

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Seal materials in flexible plastic food packaging: A review

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Abstract

Flexible packaging has many advantages in the food industry, arising from low weight, formability, multilayer complexity and cost. Heat sealing is a very efficient technique to close flexible food packaging. Currently, many thermoplastic materials are used in seal layers. A seal can be formed when these materials are heated and brought into contact; thereafter, polymer chains diffuse across the seal interface and entangle. Hydrogen bonds, polar and ionic interactions are molecular forces that can come into play, depending on the thermoplastic materials that are used in the seal layer. Bonds between identical polymers, referred to as autohesion, are formed in pouch applications (e.g., horizontal and vertical form-fill-seal packages). In lidding applications, the flexible film is sealed to a rigid cup, tray or bottle, whereby bonds can be formed between non-identical polymers because the materials are often provided by different suppliers. All heat seal technologies imply heating of seal layers but differ in the heating principle. In the food industry and in most scientific seal studies, the seals of mono- and multilayered packaging are mainly formed by conductive heating. Recently, the use of emerging technologies, such as ultrasonic and laser heating, is increasingly described in recent papers. Applied seals are characterized by strength after a specified cooling time. Immediately after heating, this strength is referred to as hot tack. A good seal performance is crucial to guarantee food safety and quality. Besides strength, tightness is important to prevent food degradation, caused by microorganisms and external gases, and to keep aromatic gases inside the package. This review aims to give a literature overview that can support stakeholders in the food industry to improve and optimize the material selection in flexible packaging, in order to obtain seals with desired tightness and strength. Heat seal studies on materials and seal technology of flexible food packaging, such as pouches and lidding films, are considered. Scientific data are categorized from a materials' perspective, based on chemical structure, which is revealed by chemical and thermal analysis. A majority of the seal studies is categorized in a first section on polyolefins as seal layers. The following sections describe the seal functionality of (i) ethylene copolymers, such as ionomers; and (ii) polyesters, such as poly (ethylene terephthalate), pol (lactic acid) and poly (butylene succinate). The role of plasticizers, fillers and other additives in the seal performance is also described. Finally, material properties, such as chain length and melting temperature (Tm), as underlying causes of seal performance, are summarized.

KEYWORDS
flexible food packaging, heat seal materials, heat sealing, hot tack, seal strength

1 | INTRODUCTION

Food packaging must protect its content from environmental factors, such as microorganisms, light and external gases, to prevent and/or inhibit microbial and biochemical degradation. Aromatic gases must be kept inside the package to preserve flavour during the life span of the packed product. Therefore, packages should remain tight until the consumer opens it for consumption.

Flexible packaging is often preferred over rigid packaging because of the low weight and low cost.[1] Lightweight materials such as paper, plastic and composites are most relevant for flexible food packaging.[2] Because of the light weight, less energy is consumed during transportation. The global market share of flexible packaging ranges around 30%, and the majority of flexible packaging has food packaging as primary end use. Flexible packaging must be sealed, because a mechanical fix is usually not possible.[1] Therefore, films are used with thermoplastics at the seal side to heat seal.[3] Thermoplastics are materials that become mouldable at elevated temperature and solidify when cooled down. In a heat seal process, temperature is increased when two seal sides of packaging materials come into contact. Figure 1 shows an example with hot jaws, whereby thermal resistant materials are used in outer layers to prevent contamination of seal jaws. During contact and heating, seal materials soften and/or melt, followed by wetting of the seal interface so diffusion and entanglement can take place. Recrystallization can occur when the seal is cooled down.[3,4] The formation of bonds at elevated temperatures of identical polymers is referred to as autohesion,[5] whereas bonds can also be formed between non-identical polymers, for example, lidding film, sealed to a tray or cup. The objective of the sealing process is to attain a leak tight seal with the desired strength. Several factors are influential in achieving this outcome.[1,3]

1.1 | Sealable materials

As thermoplastics need to be heated to allow diffusion and seal, thermal properties are of high importance, in particular the glass transition (T_g) and melting temperature (T_m). At T_g, the amorphous phases of the material change from a glassy state into a rubbery state. At T_m, all present crystalline phases start to melt, and the material changes from a solid to a viscous state, and the material starts to flow. Depending on the material, glass transition and melting are gradual processes over a specific temperature range. Amorphous polymers have no crystalline regions; for these polymers, T_m is not applicable. For semicrystalline polymers, both thermal transitions are important.[6] Decomposition temperature (T_d) is also relevant if there is an overlap with temperature during sealing. Table 1 gives an overview of the chemical structure and thermal properties of the most used commercial thermoplastics, which are applied in heat sealing and described in further detail in this review.

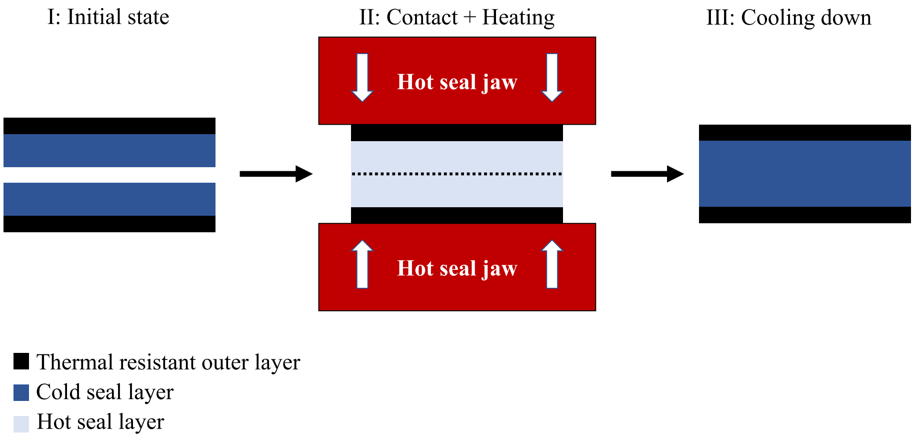
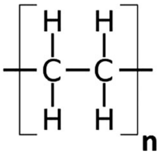
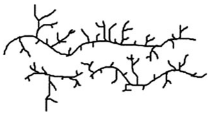
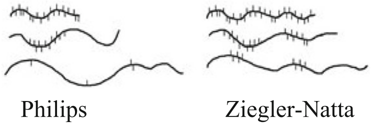
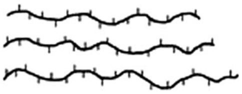
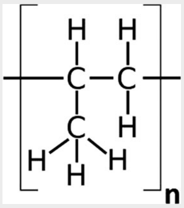
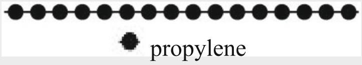
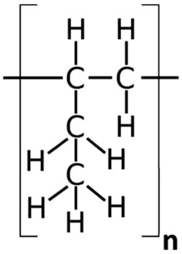


FIGURE 1 Three basic steps of a heat seal process with hot jaws; figure adapted with permission from Ilhan (2021).[4]

TABLE 1 Chemical structure and thermal properties of materials (polyolefins, ethylene copolymers, polyesters and other heat sealable polymers) used in commercial seal layers.[6–18]

Polyolefins			
Polymer		**T_g (°C)**	**T_m (°C)**
Poly(ethylene) 	Low-density PE (LDPE) 	−130 → −100	98 → 115
	High-density PE (HDPE) 		130 → 140
	Conventional linear LDPE (LLDPE)  Philips Ziegler-Natta		118 → 125
	Metallocene LLDPE (mLLDPE)  Kaminsky		60 → 119
Poly(propylene) 	Homopolymer PP  	−20 → −3	165 → 175
	Random copolymer PP  	−40 → −10	145 → 155
Poly(1-butene) (PB) adhesive resin 		−35 → −27	124 → 135

(Continues)

TABLE 1 (Continued)

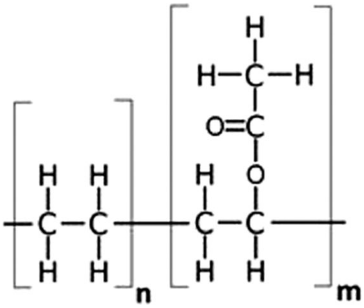
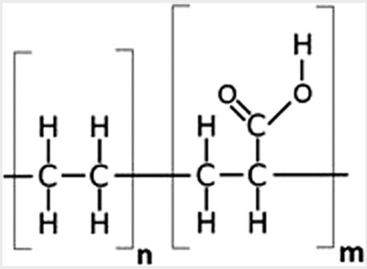
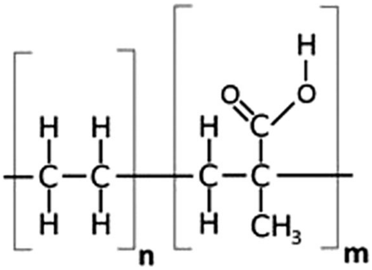
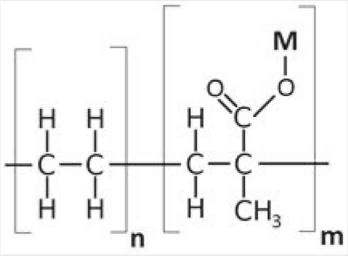
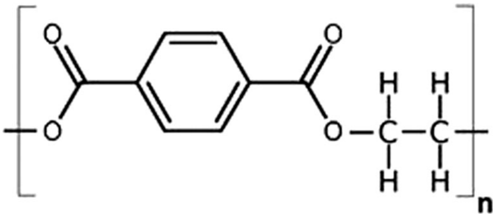
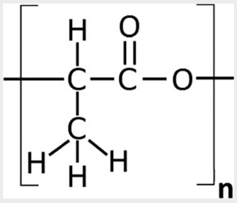
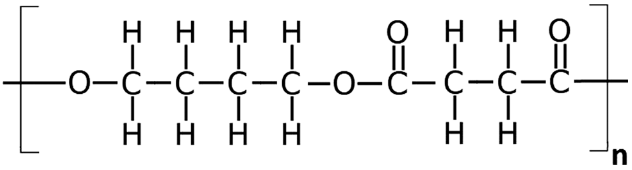
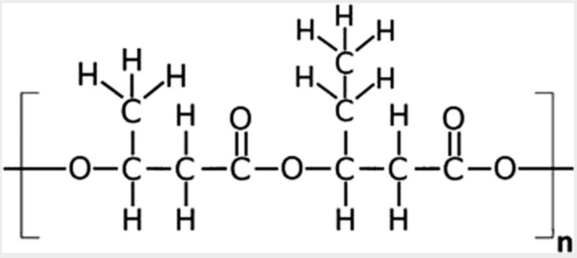
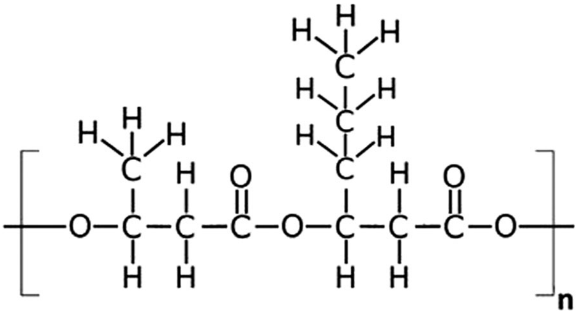
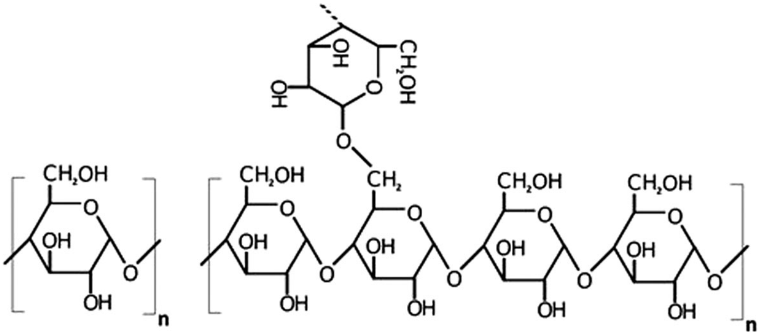
Ethylene copolymers		
Polymer	**T_g (°C)**	**T_m (°C)**
Poly(ethylene-co-vinyl acetate) (EVA) 	−45 → 20	30 → 100
Poly(ethylene-co-acrylic acid) (EAA) 	30 → 50	97 →102
Poly(ethylene-co-methacrylic acid) (EMA) 	30 → 50	97 → 109
Ionomer  M = Na, Zn	−120 → 50	88 → 100

TABLE 1 (Continued)

Polyesters		
Polymer	**T_g (°C)**	**T_m (°C)**
Semicrystalline poly(ethylene terephthalate) (CPET) 	70 → 80	245 → 265
Amorphous PET (APET)	70 → 80	Not applicable
Poly(ethylene glycol-co-1,4-cyclohexanedimethanol terephthalate) (PETG)	81	Not applicable
Poly(lactic acid) (PLA) 	55 → 65	120 → 178
Poly(butylene succinate) (PBS) 	−32	115
Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) 	−9 → 2	97 → 157
Poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) 	−4	52

(Continues)

TABLE 1 (Continued)

Other seal polymers		
Polymer	**T_g (°C)**	**T_m (°C)**
Thermoplastic starch	−50 → 110	220 (overlap with T_d)
		
Cellulose derivatives	53 → 180	260 (overlap T_d)
Poly(hexano-6-lactam) (PA6)	40 → 62	220
Poly(styrene) (PS)	90 → 110	Not applicable

1.2 | Seal technologies

There are several technologies available to heat materials.[19] A first group of technologies uses a heat source. Hot tools are pressed to the outer side of packaging films to conduct heat through the material to reach the seal interface, and tools can be heated constantly or with an impulse.[20] Heat can also be transferred by convection in hot air sealers.[21,22] Another seal technology uses infrared radiation to heat packaging materials.[23] In ultrasonic sealing, mechanical vibrations in the ultrasonic range are applied to generate heat in packaging materials.[24] A last group of technologies uses electromagnetic energy. In induction sealing, heat is generated when packages with metal layers, such as aluminium, approach a rapidly changing magnetic field.[25] In dielectric sealing, heat is generated when packages with polar polymers approach a rapidly changing electric field.[26] This paper considers all heat seal technologies. As the majority of studies uses conductive technology, the seal technology will only be mentioned if an alternative technology is used.

1.3 | Seal performance

Heat seal performance, shortened to seal performance, is used as an umbrella term, covering seal strength and leak tightness. The strength of seals can be measured immediately after heating,[27] which is referred to as ‘hot tack strength’, a relevant property in automatic production. Strength can also be measured after a cool down period of several hours or days, which is referred to as ‘seal strength’,[28] which is more relevant for transportation, storage and opening by consumers. Leak tightness, also referred to as ‘seal integrity’, can be evaluated with different methods. Method selection depends mainly on the desired output and test destructiveness. Inflating the package or putting it in a vacuum chamber while measuring pressure difference and using a penetrating dye solution are examples of evaluation methods.[29–32] The output of ASTM standards to assess seal performance is summarized in Table 2.

Process parameters, such as jaw temperature and seal time, determine the interfacial temperature, which is of critical importance for obtaining a seal with sufficient strength.[20] A recent review identified general factors that impact heat seal performance in flexible food packaging, such as material properties, process parameters, contaminants and further processes.[4] Our study gives a literature overview of seal performance from a seal materials perspective. Numerical values are shared to illustrate orders of magnitude of temperature and/or strength. The reader is referred to the individual studies to obtain more details. All information is categorized, based on the chemical structure of thermoplastics: polyolefins, ethylene copolymers, polyesters and other heat seal polymers, shown in Table 1. Poly(styrene) is only briefly mentioned as cup substrate, heat sealable to specific polymer types, because of its minor role as seal material in flexible food packaging. A separate section is added to describe the influence of additives in seal layers. At the end of the paper, the influence of material properties on seal performance is summarized. This review aims to support stakeholders in the food industry to improve and optimize the material selection in flexible packaging.

TABLE 2 Output of ASTM standards to assess seal performance.

Title of document	Designation code	Output-
Standard Test Method for Seal Strength of Flexible Barrier Materials	ASTM F88/F88M-21	Seal strength (N.mm^{-1}) and seal failure modes (e.g., adhesive peel, material break, …)
Standard Test Methods for Hot Seal Strength (Hot Tack) of Thermoplastic Polymers and Blends Comprising the Sealing Surfaces of Flexible Webs	ASTM F1921/F1921M-12	Hot tack strength (N.mm^{-1}) and seal failure modes (e.g., adhesive peel, material break, …)
Standard Test Method for Detecting Gross Leaks in Packaging by Internal Pressurization (Bubble Test)	ASTM F2096-11	Leak tightness: yes/no (250-µm defect size) + location of leak
Standard Test Methods for Pressure Decay Test for Flexible Packages With and Without Restraining Plates in Packaging by Internal Pressurization	ASTM F2095-07	Leak tightness: yes/no + Pressure decay (N.mm^{-2})
American Society for Testing and Materials. Standard Test Method for Nondestructive Detection of Leaks in Packages by Vacuum Decay Method	ASTM F2338-09	Leak tightness: yes/no (125-µm defect size) + Vacuum decay (N.mm^{-2})
Standard Test Method for Detecting Leaks in Nonporous Packaging or Flexible Barrier Materials by Dye Penetration	ASTM F3039-15	Leak tightness: yes/no (50-µm defect size)

2 | POLYOLEFINS

2.1 | Poly(ethylene) (PE)

2.1.1 | Density and branching morphology

PE is a polyolefin that is produced by the polymerization of ethylene monomers. It is the most common plastic in general and an important seal material in packaging.[33] Different structures with long and short-chain branches are shown in Table 1. This differentiation in PE is related to the difference in density, as mentioned in the name. Very low, low, medium and high-density PE are commercially available as VLDPE, LDPE, MDPE and HDPE subtypes. LDPE was developed during the 1930s, HDPE in the 1950s and linear low-density PE (LLDPE) was developed in the 1970s.[10]

LDPE has a density of 0.915–0.940 g.cm^{-3}. Table 1 illustrates the branching morphology of LDPE and HDPE, and it shows that polymer chains cannot be packed as dense as HDPE. This is caused by the number and length of branches. With decreasing density, it becomes less crystalline and more flexible. LDPE has short and long-chain branches and has molecules of low and high molecular weight (MW). It can form strong network structures, resulting in relative high melt strength, which is the strength to stretch the melt, compared to LLDPE.[34] MW distribution is given by the polydispersity index, of which values of 3.93 and 4.53 are reported in literature for LDPE.[35]

LLDPE has a density of 0.915–0.926 g.cm^{-3}. This material has a similar density but a more linear structure as LDPE. In LLDPE, short-chain branches are distributed over long linear main chains, as shown in Table 1. Side branching and MW are affected by comonomer content and process settings during production. The length of the branches is dependent on the comonomer used during production (e.g., but-1-ene, hex-1-ene and oct-1-ene). Commercial LLDPE has branches of 4, 6 or 8 carbon atoms for each branch (abbreviated as LLDPE-C4, -C6 or -C8). The branching morphology of LLDPE is related to the catalyst, as illustrated in Table 1. Philips (chromium-based, 1950s) and Ziegler-Natta catalysts (titanium-based, 1950s) produce LLDPEs with a broad MW distribution, having a high number of short-chain branches on short main chains. Another type of catalysts emerged in the 1970s. So-called Kaminsky catalysts are metallocene-based. Metallocenes are chemical structures with positively charged metal ions, such as cations of Zirconium (Zr), Hafnium (Hf) or Titanium (Ti), sandwiched between two cyclopentadienyl derivatives. The catalysts are used to produce LLDPEs with a narrower MW distribution, exemplified by a polydispersity index of 2.8[36] and a high number of short-chain branches, distributed on long main chains. However, without additional long-chain branches, mLLDPE is difficult to process in blown extrusion. The first generation of commercial mLLDPE in the 1990s had limited market success because of this disadvantage. This could be overcome by incorporating sparse long-chain branches (e.g., Affinity™), which decreases the viscosity and improves the formation of strong network structures, resulting in increased melt strength.[10,36–40] The presence of long-chain branches interferes with the forming of crystalline structures and thus facilitates a strong network because of higher intermolecular entanglements.[40]

If a high amount of comonomer is added, metallocene-catalysed PE grades are referred to as **VLDPE** (very low-density PE) because of the very low density (0.89–0.91 g.cm^{-3}), caused by a high number of side branches and/or as **polyolefin plastomers** (POP), because of the introduction of rubbery properties in combination with thermoplastic properties. These materials have a lower viscosity when heated and can fill up gaps more easily in pouches or flow around contamination, thus preventing leaks.[6,41]

Many variations in the LLDPE production process (process parameters, comonomer selection, catalyst) can affect the molecular structure. The length and distribution of short- and long-chain branches, MW, MW distribution and crystallinity are important properties for heat sealing, affecting seal/hot tack strength and initiation temperature, which is the minimum temperature at which a threshold strength value is exceeded.[42] Molecular structure also affects general film properties, such as processability, optical and mechanical performance.

HDPE has a density of 0.94–0.97 g.cm^{-3} as a result of tightly packed linear chains, as illustrated in Table 1. It is a rigid and highly crystalline polymer that is not considered in seal layers because of the high melting point, compared to better PE alternatives, such as LDPE and LLDPE.

2.1.2 | General seal characteristics

LDPE and LLDPE are both commonly used seal materials. These semicrystalline materials are flexible and can be sealed at relatively low temperatures, as the respective Tm of LDPE and LLDPE range between 98–115 and 118–125°C. Both materials are used in seal layers of multilayers, laminated to thermal resistant outer layers, which are composed of polymers with increased T_m. Recent studies have identified several molecular parameters that influence the thermo-rheological and processing properties such as the amount, length and distribution of branches, the MW and the distribution of molecular MW.[39,40,43–47]

In packaging applications, different grades of PE are blended to obtain desired properties.[48] A staggering number of grades are available with specific characteristics (seal temperature, clarity, puncture resistance, …) and costs.

Because of the similarities in seal behaviour of LDPE and LLDPE, general seal characteristics of PE are described first. PE is a semicrystalline material that requires melting to mobilize the polymer chains in both amorphous and crystal structures, which is influenced by the MW and branching morphology.

Typically, T_m, determined with differential scanning calorimetry (DSC), is correlated with the initiation temperature (T_{ini}) in a seal strength curve.[49,50] Seal initiation of PE occurs when the amorphous fraction increases by heating as crystalline regions dissolve and polymer chains become more mobile. For LDPE and LLDPE films, seal initiation occurs when the amorphous fraction increases to 77%. With rising temperature, diffusion and entanglement is facilitated, and there is a fast increase in seal strength, until a plateau value is achieved around T_m. The typical sigmoidal shaped seal strength dependence with interfacial temperature is illustrated in Figure 2. The plateau strength value is correlated with the yield stress, a tensile characteristic that marks a transition between elastic and plastic behaviour. Yield stress is a function of the amorphous fraction of PE. A lower fraction of the amorphous content at room temperature leads to higher seal strength.[42] The plateau value decreases when PE decomposes, is pushed away by seal jaws and/or if outer layers are molten or decomposed.

Another study relates melting and interdiffusion of heat sealed LLDPE films. The importance of the amorphous fraction is nuanced in their discussion. The type of chains that are able to melt and to diffuse across the interface are more important than the amorphous fraction. Only at temperatures at which high-molecular-weight chains with a low number of short-chain branches become mobile and are able to diffuse across the interface, high seal strength can be achieved. Low-molecular-weight chains with a high number of short-chain branches diffuse easily at lower temperatures but contribute much less to seal strength.[51]

The distribution of short-chain branches also plays an important role: If more short-chain branches are present on medium to long MW chains, T_m is decreased, and crystals with longer chains are created. These longer chains are released in the seal interface at T_m so diffusion can take place and seal performance is increased. With larger crystal sizes, yield strength is also increased,[36] which corresponds with the finding of an earlier study that correlates high yield strength with high plateau seal strength.[42]

Another study focusses on the correlation between hot tack related temperature dependence and DSC results of mLLDPE/LDPE blends. Seal materials with a high number of small-sized crystals melt faster and more complete during sealing, resulting in stronger bond formation caused by more interdiffusion. Hot tack strength is increased at temperatures close to recrystallization point in DSC.[52]

Besides conductive sealing, a much lower number of papers demonstrate compliance of PE as seal material in other seal technologies, such as impulse, ultrasonic and induction sealing.[24,53,54] Also, in these technologies, PE is heated to melt and to diffuse across the interface. Therefore, the above described molecular and crystalline properties

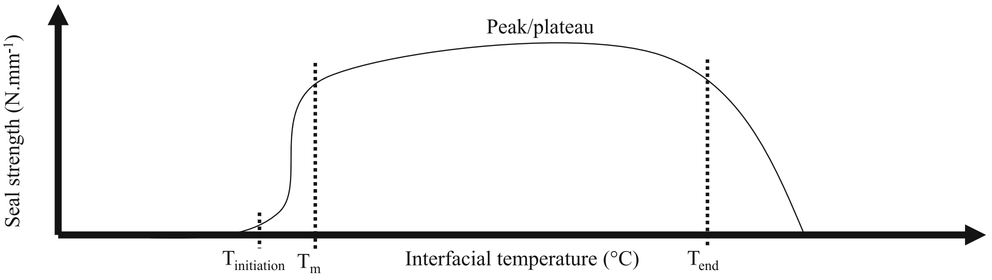


FIGURE 2 Temperature dependence of seal strength of PE.

impact the heat seal performance in general, not only in the case of conductive sealing.

Although polyethylene appears to consist of simple -CH_2-CH_2- bindings, this amazing polymer exhibits a multitude of properties depending on its molecular structure. This is also reflected in differences in seal performance between LDPE and LLDPE, which are highlighted in the following subsections.

2.1.3 | Insights from LDPE

At similar MW, LDPE shows a higher degree of shear thinning than LLDPE making it better processable for blown extrusion, caused by the formation of strong network structures, which is beneficial in blown seal film production.[43] The main disadvantage of the molecular structure of LDPE is the mechanical properties and, related to this, seal performance.

LDPE has a broad MW distribution compared to LLDPE. Low-molecular-weight molecules in LDPE decrease the mechanical properties such as strength and toughness compared to high-molecular-weight molecules in LLDPE.[46,55] LDPE is a weak subtype of PE for **seal strength**, mainly because of the presence of low-molecular-weight molecules and the highly branched structure that disrupts the crystalline structure. However, there are applications where a low seal strength is sufficient or desirable (e.g., in peel films where peel component is added to decrease seal strength).

Another important seal characteristic is the **initiation temperature**. LDPE is not the best option to increase the speed of the sealing process because of its relatively high T_m (98–110°C), compared to mLLDPE-subtypes.

The **hot tack performance** of LDPE is relatively bad, due to several causes. First, the presence of long-chain branches causes slow diffusion restricting molecules to entangle across the seal interface. In addition, LDPE, but also LLDPE, has no additional molecular forces, such as hydrogen bonds and ionic interactions, besides dispersion forces. Melt strengths, and resulting hot tack strengths at high temperatures, of LDPE and LLDPE, are relatively low, compared to other polymers with those molecular forces, such as ionomers, described later in this paper.[6]

In conclusion, LDPE is a commonly used seal material as it combines good processability with mediocre seal performance. The seal performance is increased by blending other PE subtypes.[56]

2.1.4 | Insights from LLDPE

By placing more short-chain branches on medium to long main PE chains, the seal performance is improved by decreasing T_m, which is beneficial in high-speed sealing operations, and by increasing hot tack strength, which is beneficial in automatic seal processes, such as vertical and horizontal form-fill-sealing. T_m is negatively correlated with the amount of comonomer added. Hot tack strength is positively correlated with entanglement depth, related with the length of the main chains that participate during entangling. The branching morphologies of LLDPE and mLLDPE structures in Table 1 illustrate the narrower MW distribution and the more homogeneous distribution of comonomers in mLLDPE. As a result for mLLDPE, medium to long chains participate during entangling. In conventional LLDPE, medium to long chains remain stuck in crystallite structures while short main chains are entangling. Because of this, mLLDPE is regarded as better performing seal material, compared to conventional LLDPE.[36,39,40]

There are many LLDPE types available with low T_m (e.g., POP, which are alpha olefin copolymers having rubber-like properties) that are more appropriate than LDPE for high speed packaging lines.[6] A selection of commercially available LLDPE grades with different properties is shown in Table 3. Three properties highlight the differences between these grades: (i) the melt flow index (MFI), which is a measure of the ease of flow through a capillary, (ii) density and (iii) MW.

In one study, a seal initiation temperature of 84–92°C is reported for Dow's resin Affinity™ 1140, a mLLDPE-C8 of medium MW with sparse long-chain branching. Seal initiation temperature values of other mLLDPEs (Affinity™ 1450 of Dow: mLLDPE C8 of low MW with sparse long-chain branching; Exact™ 3132 of ExxonMobil: mLLDPE-C6 of medium MW) are between 84°C and 92°C and the seal initiation temperature of LLDPE (TF-Y534-IP of Nova Chemicals: LLDPE-C6 of medium MW) occurs above 100°C.[36] The variations in seal initiation temperature are related with density.

The amount of detail, provided in Table 3, is often not available of other LLDPE materials, used in seal studies. The following section describes other LLDPE materials that were subject to studies investigating how the seal performance is related to rheology and squeeze-out behaviour, the effect of surface modification using corona treatment, the alternative use of ultrasonic sealing and the impact of contamination during sealing.

TABLE 3 Properties of commercial LLDPE grades.[36,55]

Resin (code)	Description	Supplier	MFI (190°C. 2.16 kg^{-1})	Density ($g.cm^{-3}$)	MW ($kg.mol^{-1}$)	T_{ini} (°C)
Affinity 1140	mLLDPE-C8	Dow Chemical	1	0.895	105	84–92
Exact 3132	mLLDPE-C6	ExxonMobil	1.2	0.900	NA	84–92
Affinity 1450	mLLDPE-C8	Dow Chemical	7.5	0.902	NA	92
Dowlex 2045	LLDPE-C8	Dow Chemical	1	0.920	102	110
(TF-Y534-IP)	LLDPE-C6	Nova Chemicals	0.75	0.934	118	120
(FPI 20)	LLDPE-C8	Nova Chemicals	1	0.920	105	120–122

The **squeezing-out** of seal materials is modelled in one study. A small amount of flow is desired to fill up channel leaks at wrinkles, with solid contamination and/or at gusset areas, where horizontal and vertical seals are crossing. A high flow can decrease the seal performance if seal materials are squeezed out. This study is carried out on a film with a polyolefin plastomer (mLLDPE) of 0.906 $g.cm^{-3}$. Important rheological properties are identified. The viscosity at low shear rate is a good indicator, while the MFI, which is often used in polymer processing, is not. The authors point out that caution is advised in the particular practical cases where a thick film with LLDPE seal layer is switched to a thin film in a packaging line and when profiled seal jaws are used with serrations (crimpers).[57] This squeeze-out behaviour is also subject of another study with mLLDPE seal layer. At low jaw temperatures (105°C), squeeze out flow (SOF) and its dependency with seal time are reduced. SOF also decreases with thin seal layers (seal layers of 50 and 130 μm are considered). With thin seal layers, SOF is only increased at high seal pressure and long seal time if viscosity of seal material is reduced. A 10-fold seal pressure increases SOF only with thick seal layers.[58]

Few papers study the influence of **surface modification**. The influence of corona discharge treatment on seal performance is subject of two studies on LLDPE-based seal layers. In both papers, a decrease of seal strength is observed with treated samples, with a change from tear to peel failure at a seal strength test. Crosslinking of surface molecules is suggested by the authors as underlying cause.[59,60] Cascaded and standard dielectric barrier discharge (DBD) treatments are compared on films with PE seal layer with the objective to sterilize polymer films and maintain seal performance. The cascaded treatment decreases seal strength to a much lesser extent, compared to standard DBD. The production of low MW oxidized materials at the surface is suggested as underlying cause of decreased seal strength.[61]

Ultrasonic sealing is subject of other papers. A seal optimization procedure of a commercial flexible film with LLDPE-C4 seal layer is described. A seal window is developed that shows the broad area, covering 39% of the seal amplitude × force design space, that considered extreme values of 18- to 36-μm seal amplitude and 2–6 $N.mm^{-1}$ seal strength, where 90% of the optimum strength can be achieved.[62] Another study compares seal strengths with ultrasonic and conductive sealing of LLDPE seal layers. A decreased seal strength is observed with ultrasonic technology, and this effect is more pronounced for materials with a narrow melting range and high viscosity slope at increasing temperatures. Besides a high relative maximum seal strength, compared to heat conduction, a broad parameter window in which strong seals are produced and bond formation at low seal forces are described as good ultrasonic sealing performance. Different types of contamination, solid and liquid, are evaluated in the same study, comparing seal strengths of ultrasonic and conduction sealing. In the comparison, ultrasonic sealing is optimal with wheat flour and coffee powder contamination, while conduction sealing is the better choice with grated cheese, olive oil and salad dressing.[24,63]

Studies with **contamination** are often performed on seal layers of LLDPE because of the required high seal performance. These studies highlight the importance of optimization of process parameters, depending on the applied contamination to produce strong and leak tight seals. In most cases, especially if high contamination densities are used, a decreased seal strength is observed. One study optimizes seal settings with liquid contamination, water and oil, of an LLDPE film.[64] Another study uses vegetable oil and a salt water solution as liquid contaminants.[65] Another study evaluates the influence of solid contamination, using coffee or blood powder, on seal performance and hot tack strength. Two commercial flexible film structures with a variation of mLLDPE composition in the seal layer, of which one with plastomeric properties, are compared. The film with plastomer-based seal layer achieves a hot tack initiation temperature of 90°C and leak tight seals at all evaluated cases with solid contamination. The other film achieves an initiation temperature of 105°C and a minority fraction of leaking seals at optimal settings with contamination.[66] Another study evaluates the seal performance of a topfilm with LLDPE seal layer, sealed to a non-woven filter with polyolefin seal layer for coffee capsule application, in terms of conductive seal parameters and coffee contamination. Differences in heat conduction are revealed with thermal imaging.[67] Another study uses coffee or sugar powder as solid contamination. Three films with blends of LDPE and LLDPE are compared with an LDPE reference: one film with 30% conventional LLDPE-C4, another film with 30% mLLDPE-C6 and a third film with 40% mLLDPE-C6. Seal strength is not affected at low contamination levels, up to 10 $g.m^{-2}$ for coffee and up to 2 $N.mm^{-2}$ for sugar. Additional hot tack results illustrate that LLDPE blends outperformed the LDPE reference film with decreased hot tack initiation temperatures (3–4°C), increased hot tack peak strengths (approx. 125–150%) and increased hot tack windows.[68]

In general, LLDPEs, especially metallocene catalysed, are regarded as better performing seal materials in food packaging because of lower initiation temperature, high seal/hot tack strength and good seal-through-contamination performance, compared to LDPE.

2.2 | Poly(propylene) (PP)

2.2.1 | Tacticity and copolymerization

PP is a polyolefin that is produced by the polymerization of propylene monomers. The structure of PP is shown in Table 1. For packaging, it is the second most used polymer, after PE. It was first developed in the 1950s.[10]

A first differentiation of PP is made by the position of the methyl heads (CH_3 branches) in the polymer chain. In isotactic PP, all heads stick out at the same side as shown in Figure 3. Isotactic PP is a rather stiff material that can crystallize, and it has a good chemical and heat resistance.[69] In syndiotactic PP, the heads repeatingly stick out at both sides. Syndiotactic PP can also crystallize.[70] In atactic PP, all heads stick out randomly, and the resulting polymer will not crystallize and is amorphous. Standard commercial PP used in packaging is semicrystalline. It contains over 90% isotactic polymer and has a small amount of atactic polymer. Commercial PP has a narrow density range of 0.898–0.908 $g.cm^{-3}$, and no extensive differentiation based on density is made, unlike PE.

A second differentiation of PP is made between homopolymers with a repeating sequence of propylene units, and copolymers, produced by adding small amounts of comonomer, usually ethylene and/or butene. Copolymers are more transparent and have higher impact strength, flexibility and/or a lower and broader T_m. The randomness and the amount of incorporated comonomers are determining factors. There is a broad range of copolymer grades commercially available. Block and random copolymers can be differentiated, based on the distribution of comonomers as shown in Table 1.[10]

PP is commonly used for its rigidity as a tray or cup and as a seal material in topfilm for sealing these trays or cups. In contrast to PE, PP has a relatively high T_m, which ranges between 145–155°C for random copolymer and 165–175°C for homopolymer PP. Nevertheless, this can be beneficial if the package is heat treated by pasteurization or sterilization after sealing. If a decreased seal temperature,[71] high transparency and flexibility are needed, random and terpolymer PP (copolymer with three different monomers) can be used. If high impact resistance and high flexibility at freezing conditions are desired, block copolymers can be used. Block copolymers have a decreased T_g.[72] In freezing applications, PE is often preferred over PP as seal film because of the low T_g.[73,74] Blends and multilayer structures can be made of homopolymer and copolymer(s) to combine properties to get a suitable material for the desired application.

A third differentiation is made between cast and oriented PP. Cast PP (CPP) can be used as seal material while oriented PP is used as substrate in flexible packaging, typically for snack packages.[7]

2.2.2 | Seal characteristics

The number of available papers on PP seal layers is much lower compared to those with PE. Similar to PE and other olefinic polymers, it needs to be (partially) melted to mobilize the chains so that diffusion can take place at the interface.

In a study with an impulse sealer, jaw temperature is maintained for 0.1 s, followed by a decrease to room temperature in 5 s. Seal strength of a film with CPP seal layer increases sharply from 110°C to 120°C. After this temperature, a plateau strength is reached.[75] Melting of the CPP, determined with DSC, initiates at 80°C and increases gradually into a broad peak at 146°C.[76] Another paper studies conductive sealing of a film with metalized CPP. At seal times of 0.1 and 1.0 s, seal initiation and plateau initiation occur below the T_m of the seal material, determined with DSC. Pressure is also varied, but there is no significant change of seal strength after a threshold value of 1.25 bar.[77] The influence of cyclic loading, as treatment to enhance seal strength, is evaluated on films with CPP seal layer. The increase of seal strength is related with molecular orientation, caused by cyclic deformations and relaxation of the heat-sealed edge. In that study, seal strengths, between 1 and 1.5 $N.mm^{-1}$ can be obtained at jaw temperatures of 120–180°C, seal time of 0.1 s and seal pressure of 0.42 $N.mm^{-2}$.[78] The optimization of seal temperature, time and pressure is subject of another study. It is performed on a film with ethylene-propylene copolymer seal layer. Seal temperature, time and their interaction impact seal strength significantly. High seal strength is obtained at 120°C and 0.9 s.[79] Another study evaluates correlation between results of hot tack and seal strength with results of successive self-nucleation and annealing thermal analysis. Films with ethylene-propylene copolymers and terpolymers of ethylene-propylene-butylene are studied. Seal and hot tack curves are in accordance with melting curves. Temperatures of seal initiation and 40% molten fraction are linear correlated.[80] Seal initiation in conductive experiments occurs at an amorphous fraction of 60% with homopolymers. The correlation between plateau seal strength and yield strength differs from the model obtained for PE.[42]

2.3 | Isotactic poly(1-butene) (PB)

PB is a polyolefin that is produced by the polymerization of 1-butene. The structure is shown in Table 1. It is available as homopolymer and copolymer with ethylene or propylene. PB is commonly used as component in easy-opening packages that peel cohesive, by rupturing weak bonds between PB and the matrix component, which is often PE.[81–84] It can be blended as peel component with PE, PP and ethylene copolymers such as poly(ethylene-co-vinylacetate) (EVA), poly(ethylene-co-methacrylic acid) (EMA), poly(ethylene-co-acrylic acid) and ionomers.[85–87] PB forms incompatible mixtures in blends with one of these polymers, resulting in controlled peel failure of the seal. Besides peelability, addition of PB in the seal layer can decrease seal initiation temperature.[84] The most common peel system in practice is the blend of PE and PB. PB is dispersed in low amounts (e.g., 15%) in a PE matrix. The PB–PE ratio, the chemical composition of the used PB and PE grades, the solid-state mechanical properties of both polymers and the dispersion can determine the peel performance of the overall film. PB is present as small islands in the PE matrix. These islands behave like small microperforations after sealing as PE and PB. This seal however is still hermetic and safe. Because of the weak spots, a smooth cohesive peel failure is obtained during opening of the seal.[81,88] The quality of the seal is assessed negatively if there is a large amount of hazy elongated parts of the blend, referred to as ‘angel hair’, present at the peeled surface.[81]

FIGURE 3 Tacticity of PP.

3 | ETHYLENE COPOLYMERS

3.1 | EVA

EVA is produced by copolymerizing ethylene and vinylacetate. The structure of EVA is shown in Table 1. This polymer can be differentiated by the amount of vinyl acetate. Polymers with low vinyl acetate content are referred to as vinyl acetate modified PE. With higher vinyl acetate content (4–40%), the polymers are referred to as thermoplastic EVA. EVA is used in blends with PE in seal layers. In these blends, addition of EVA changes the seal (decreases the seal initiation temperature, broaden the seal plateau temperature range), mechanical (increase toughness) and/or optical (increase clarity and gloss) performance.[89,90] Besides chain entanglements, EVA exhibits additional polar interactions.[6]

Peel performance of films with EVA seal layer, sealed to a biaxial oriented poly(ethylene terephthalate) (BOPET), is subject of a seal study. Without surface treatment, seal strengths are very low, increasing with rising VA content, up to 0.06 $N.mm^{-1}$ with 24% VA. When films are treated with DBD treatment, seal strengths of 0.21 $N.mm^{-1}$ are achieved. The underlying cause of this increase is the presence of more reactive polar groups. Lower thickness of EVA seal layers increases seal strength slightly, caused by a decreased crystallinity during sealing with thin seal layers.[91] In another study on peel performance of EVA seal layer, heat sealed to a PET substrate, seal strengths around 0.25 $N.mm^{-1}$ are achieved.[92] EVA can also be used in seal layers of peelable lidding applications, sealed to PS or PP substrates.[6]

Another paper studies blends of EVA and PE. Yield strength decreases gradually with increasing EVA concentration, which can result in a decreased seal strength. The high mobility of EVA in the blend leads to higher diffusion and better surface adhesion that can increase the seal strength. These counteracting phenomena are more or less pronounced, depending on the amount of EVA in the PE/EVA blend. Interdiffusion is more pronounced with 20% and 40% EVA, resulting in increased seal strength. At 60% or higher, yield strength decreases significantly, resulting in decreased seal strength. Seal strengths of PE/EVA blends, considering blend ratios from 0% to 100% EVA content, variate between 0.5 and 0.7 $N.mm^{-1}$. Seal initiation temperatures of these materials range between 75°C with 100% EVA and 110°C with 0% EVA.[93]

3.2 | Acrylic acid copolymer

Two common acrylic acid copolymers are poly(ethylene-co-acrylic acid) or ethylene acrylic acid copolymer (EAA) and poly(ethylene-co-methacrylic acid) or ethylene methacrylic acid copolymer (EMA). EAA is the copolymer of ethylene and acrylic acid. EMA is the copolymer of ethylene and methacrylic acid. In literature, this group of polymers is often referred to as acid copolymer resin (ACR). Both structures are shown in Table 1.

Besides their uses in seal layers, these copolymers are widely used as adhesives in laminated structures. EAA and EMA adhere well to polar substrates such as PET, aluminium and paper.[94] A differentiation with these types of polymers can be made by acrylic acid content and composition. EMA is the starting substance in the production process of ionomers, a material group that will be discussed in the next section. EAA and EMA have the ability to make hydrogen bonds that can enhance its strength properties, such as melt strength and resulting hot tack strength.[6,95] EAA and EMA are often used in seal layers of peelable lidding applications, like EVA, because of their ability to seal to a broad range of materials, such as PET and PS and PP.[6]

A seal study shows low seal and hot tack initiation temperatures of 100% ethylene acrylic acid film, below 100°C. Hot tack and seal strengths achieve respective approximate peak values of 0.7 and 1.1 $N.mm^{-1}$.[85] Another study compares hot tack performance of papers with EAA and EMA seal layers with similar acid content, respectively, 8.4 and 8.7%. Hot tack initiates well below 100°C, and peak values are almost identical, achieving 0.65 $N.mm^{-1}$ at 100°C.[96]

Next to ethylene, styrene can be used as comonomer in acrylic resins. This combination is often used in water soluble dispersions such as Joncryl® of BASF. It is commonly used as heat seal lacquer in flexible packages.[97–99] Sealable lacquers are a very thin alternative, from 1–10 μm, to extruded seal layers. The low amount of seal material can be beneficial to improve recyclability of multimaterial structures. The peel seals with acrylic heat seal lacquers with aluminium substrates are typical examples of adhesive peeling, for example, yoghurt cups with topfilm.

3.3 | Ionomer

3.3.1 | Chemical structure and morphology

Ionomers are produced by adding metal ions to poly(ethylene-co-methacrylic acid). Sodium (Na) and Zinc (Zn) ions are commonly used for packaging materials. The chemical structure is shown in Table 1.

The presence of positive ions partially neutralizes the acid groups in the polymer. Ionomers have ionic clusters, besides amorphous and crystalline regions, as shown in Figure 4.[7] The possibility to make ionic

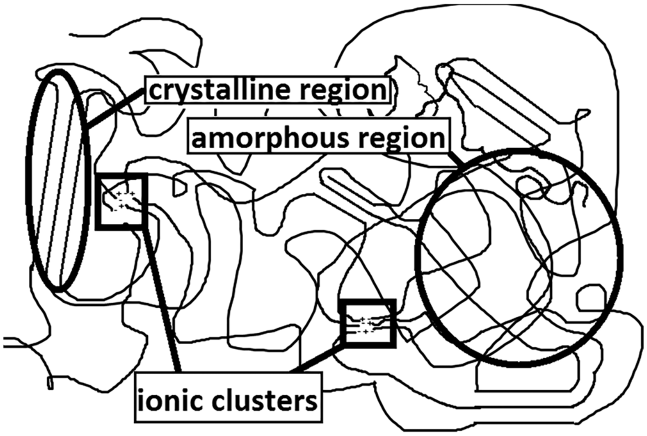


FIGURE 4 Schematic structure of different regions in ionomers.

interactions adds up to its strength potential, besides hydrogen bonding and chain entanglement.[6,95]

The crosslinks in these cluster are thermal reversible so it is still a thermoplastic polymer. The crosslinks restrict chain mobility.[100] Melt strength, which can be calculated after measuring the elongational load of molten polymer, increases, and MFI decreases if more ions are added.[7] A differentiation between ionomers can be made in acid content similar as with acrylic acid copolymers, in the amount of neutralization and in the type of metal ions.

3.3.2 | Seal characteristics

Ionomers can be combined with other types of polymers to achieve specific properties. For instance, when blended with PB, the resulting material can create a peelable seal layer.[83] Seal performance (hot tack strength, seal-through-contamination), oil and grease resistance, puncture and abrasion resistance of ionomers are described as interesting features to implement ionomers in flexible food packaging.[101,102]

A paper that evaluates hot tack performance of a commercial flexible film with ionomer seal layer reports a low initiation temperature of 90°C, a high peak value of 0.41 $\mathrm{N.mm^{-1}}$ and a broad processing window. Compared to mLLDPE-based seal layers, the ionomer film shows superior hot tack performance. The superior performance is however not reflected in the seal performance with solid contamination. The authors suggest a low effective seal layer thickness and an undesirable flow behaviour for this particular case with solid contamination as potential causes of the worse seal-through-contamination performance of the ionomer seal layer.[66] A low hot tack initiation temperature and broad hot tack processing window are also illustrated in another comparative seal study. Especially, the processing window of ionomers stands out. Ionomer films achieve strengths above 0.2 $\mathrm{N.mm^{-1}}$ in a broad temperature range above 120°C, while acrylic acid and mLLDPE films achieve these high values in more narrow processing windows of respectively 75°C and below 50°C. The same study reports a superior seal-through-fatty-contamination of ionomers, compared to mLLDPE.[95] Another study points out that ionomers have almost no squeezing-out behaviour during conductive sealing at temperatures below 150°C. Ionomers will rarely experience squeeze-out problems when seal temperature is optimized, a phenomenon attributed to high viscosity, caused by hydrogen bonds between acid groups and ionic interactions between metal cations and acid groups.[57]

4 | POLYESTERS

4.1 | Poly(ethylene terephthalate) (PET)

PET is a polyester, composed of a repeating sequence of terephthalate and ethylene groups as shown in Table 1. It is made in a condensation polymerization process of dimethyl terephthalate (DMT) or terephtalic acid (TPA) and ethylene glycol (EG). For packaging, it is a popular polymer because of its light weight, good appearance, mechanical and gas barrier properties. It was first developed in the 1940s. It is most famous as bottle material for carbonated drinks. PET is also used in sealable packaging concepts such as trays, cups and films.[10]

A differentiation is made between amorphous and crystalline PET (APET and CPET). The crystallization process is minimized to produce APET. Crystallization is influenced by structural factors such as MW, MW distribution and linearity of chain structure. A narrow MW distribution, high MW and linear chains are ideal to obtain high crystalline PET.[103] Crystallinity is however also influenced by extrinsic factors such as the temperature profile and stretching during production. Crystal growth rate decreases with increasing MW. At equal extrinsic crystallization conditions, high-molecular-weight samples can reach lower content of (incomplete) crystals if crystallization time would not be sufficient because of the decreased growth rate.[104]

APET can be sealed and is more soft, flexible, high transparent, glossy and has high impact strength. It is ductile at temperatures below its T_g, which is 67°C, such as room temperature. This is a result of the production process, where PET sheets are quenched against a cold roll after cast extrusion to limit the crystallization process, required to produce APET.[7] APET is more often used in packaging concepts, such as bottles, films and trays. In flexible films, APET is in competition with oriented polyamide, oriented PP and machine-direction oriented poly(ethylene) as outer layer material.[105] APET has poor heat sealability, as a result of insufficient flow behaviour, compared to polyolefins and ethylene copolymers, in the seal temperature range. APET can be sealed above T_g. Recrystallization during heating should be avoided to maintain mechanical and optical properties. APET is often laminated or coated with a better flowing seal material.[7]

CPET cannot be sealed as a result of crystalline structures that prevent chains to participate in the entanglement process. It is a strong, hard and stiff material associated with high temperature and solvent resistance. CPET has a T_g and T_m of, respectively, 81°C and 250°C. It is however not feasible to melt CPET; recrystallization results in a more brittle and less visually attractive material. CPET is used in rigid structures, such as trays and cups.[10]

A second differentiation of PET is made between PET and poly(ethylene glycol-co-1,4-cyclohexanedimethanol terephthalate) (PETG). **PETG** has limited application as seal material in flexible food packaging. It is produced by partially replacing ethylene glycol with 1,4-cyclohexane dimethanol (CHDM). PETG is an amorphous copolyester and is less likely to become brittle by heating, which makes it interesting in heating processes such as heat sealing. It is much softer, compared to APET, and has excellent oil resistance.[7]

4.1.1 | Seal characteristics

There is a very low number of seal studies available on the seal performance of flexible food packaging with PET seal layers. It is not a good practice to melt PET during sealing because of the brittleness of the cooled down material. The bad performing seal as a result of melting PET in seal layers is given as underlying cause of water vapour loss in a paper that studies the quality of rose snapper in ovenproof plastic films.[106] Another paper studies the rare topic of enhancement of seal performance with surface modification. Here, APET film is treated with atmospheric plasma, resulting in increased seal strength. Seal strength increased 25-fold from very low values without treatment, well below 0.03 $N.mm^{-1}$, to moderate values with treatment, between 0.4 and 0.5 $N.mm^{-1}$. Ageing decreases seal strengths of treated samples gradually. Plasma treatment increased surface roughness and added oxygen and nitrogen functional groups.[107] Another study illustrates the potential of laser technology to seal lidding film to a tray.[23] APET is high viscous compared to molten polyolefins during sealing. Low viscous behaviour is related with filling up channel leaks in packages or encapsulating contamination, referred to as 'caulkability'. The less caulkable APET is not suited as seal layer in pouches or in the presence with contamination. However, soft amorphous seal layers (e.g., PETG) can be used to increase caulkability. This is highlighted in a seal-through-contamination study with a lidding film that contains a PETG seal layer. This caulking behaviour can however also be partially attributed by the soft layers, containing PE, above the seal layer.[108] Because of the lack of publications, insights on the influence of material properties on seal performance is rather limited.

4.2 | Poly(lactic acid) (PLA)

Polylactic acid is a biodegradable material that can be produced from renewable resources. It is produced by condensation of D or L-lactic acid and lactide. The structure is shown in Table 1. PDLA and PLLA are transparent and semicrystalline materials with a T_m between 170°C and 180°C and a T_g around 55–60°C, and crystallinity around 35%, being strong and stiff at room temperature. Without modification, PLA is a very brittle material. However, these weaknesses can be overcome by blending with other polymers, such as poly(butylene adipate-co-terephthalate) (PBAT), poly(caprolactone) (PCL) or starch and/or adding additives.

4.2.1 | Seal characteristics

Blends

In recent studies, the influence of **PLA:PBAT** blended films with and without chain extenders on the heat conductive seal strength is evaluated. Maximal seal strength is achieved with a blend of PLA:PBAT and chain extender in a 40:60:0.15 blend ratio.[109] In another study, seal strengths of 0.5–0.7 $N.mm^{-1}$ at a broad range of interfacial seal temperatures between 76°C and 105°C and low haze (<4%) are achieved with PLA:PBAT blended film in a blend ratio of 80:20.[110] Another study reports a hot tack initiation temperature of 75°C for films of 100% PLA or a PLA and PBAT blend, with respective peak hot tack strengths of 0.33 and 0.11 $N.mm^{-1}$.[111] In a study that evaluates seal performance of PLA:PBAT blended film, a decreased seal initiation of 20°C, compared to pure PLA film, is reported. The underlying cause of this shift is related to the crystalline structure of PBAT. A gradual decrease of PBAT crystallinity correlates with the shift in seal initiation temperature.[112]

In another study, seal performance of blended films with different ratios of **PLA and PCL** is compared with pure PLA and PCL reference films. Increasing PCL content decreases seal and hot tack initiation temperatures. Hot tack initiation temperature decreases 15°C with 20% PCL, compared to pure PLA, that initiates at 85°C. Another 10°C decrease is realized with 40% PCL. Besides the advantage of decreasing seal initiation temperature, peak seal and hot tack strength slightly increases at high PCL content. The underlying causes of improved seal performance are a decreasing T_m, as evidenced by DSC, at increasing PCL content, and higher mobility of polymeric long chains, causing higher interdiffusion. The authors additionally attribute the strong seals in the melt phase to polar interactions, arising from the nature of polyesters.[93] The slightly increased seal strength with rising PCL content in a PLA:PCL blend is also confirmed in another study with 10% PCL. In the same study, however, seal strength decreases at PCL content of 20% and 25%.[113]

The influence of blend ratio of **PLA:starch** on seal strength is subject of another study. Seal strength of PLA blends with 5 and 10 weight percent starch are in general slightly decreased compared to pure PLA at considered seal temperatures and times of, respectively, 90–100°C and 0.5–2.0 s.[114]

In another study, hot tack performance of blended films with PLA and **nanoclay** is evaluated. Sealing initiates around 80°C. A peak value, just below 0.6 $N.mm^{-1}$, is achieved at 135°C with pure PLA samples. Samples with 2% and 4% clay content achieve increased peak values, up to 1.0 $N.mm^{-1}$, at the same temperature.[115]

The abovementioned studies use conductive heat sealing; however, some other studies investigate **alternative technologies to seal PLA**. Ultrasonic seal technology is used to evaluate the seal performance of plasticized PLA. High-molecular-weight poly(ethylene glycol) (PEG) is used as plasticizer to decrease overall brittleness. Films are produced by cast extrusion to a thickness of 50 μm. All films, with and without plasticizer, are heat sealable with ultrasonic technology. The addition of plasticizer improves seal performance by broadening the processing window.[116] Impulse and ultrasonic seal performance are also subject of another study on PLA films. Impulse sealing produces strong seals at low seal times, down to 0.5 s, while ultrasonic sealing is able to produce strong seals at lower seal times, down to 0.15 s.[117] A final study uses laser technology to optimize seal settings of coffee capsules of PLA, with flexible lidding film, sealed to a cup.[118]

Finally, other aspects are investigated using PLA, such as the influence of **crimp**, the effect of a **heat-treatment** after production and the potential effect on the **chemical structure**. PLA achieves increased seal strength at crimp angles below 90°C, although small fractures are caused by these narrow angles. The study concludes that crimp geometry is a secondary factor, compared to seal temperature.[119] In another study, the impact of post-production heat treatment on heat seal performance is evaluated. The goal is to improve the water barrier, by an alteration of the polymer matrix between 100°C and 150°C. As this kind of heat treatments between 100°C and 130°C with long duration times increases crystallinity, higher seal temperatures are required to produce strong seals. At treatment temperatures of 140–150°C, PLA remains amorphous, and seal temperatures are only slightly elevated, compared to untreated PLA.[120] Finally, it is shown that the chemical structure and molar mass of PLA, before and after conductive heating, are not changed, based on nuclear magnetic resonance spectra and gel permeation chromatography.[121]

It can be concluded that with the available studies on seal performance of PLA, improvement is possible by blending other components, such as PBAT, PCL or starch. Several technologies, such as heat conduction, ultrasonic, impulse and laser, are available to seal PLA properly. It is currently used as an emerging seal material in many applications, and the trend is positive as there is a growing demand for flexible bioplastic packaging.[122]

4.3 | Poly(butylene succinate) (PBS)

PBS is a biodegradable material. Depending on the resources of monomers, it can be fossil and/or biobased. The structure is shown in Table 1.

Because of the long alkyl chains, PBS is a rather soft material. It is a semicrystalline hazy material with a T_g around −32°C, a T_m of around 115°C and a tensile strength of 30–35 MPa. These properties are comparable with polyolefins, and thus, this material can be seen as a biobased and biodegradable alternative.[7,123,124] The same machinery can be considered for monofilament extrusion, blown extrusion and injection moulding as for conventional thermoplastics processing.[125] Because of food contact approval and good sealability, this material could be used as seal material at the inner side of food packaging. However, there are very few studies available on the seal performance of PBS packaging films. Seal initiation temperatures around 80–100°C are found with thin films, sealed for 1.0 s.[111,126] A peak hot tack strength of 0.40 $N.mm^{-1}$ can be achieved.[111] Another study reports that PBS films can be sealed at the same settings as films with PE seal layers. Only one setting is considered in that study, and samples are sealed at 140°C and 0.8 s.[127] Properties can be modified by blending, adding fillers and copolymerization, among others.[128–131]

4.4 | Polyhydroxyalkanoates (PHA)

PHAs are a family of biodegradable polyesters that can be produced and degraded by a broad range of microorganism species. The properties of PHAs range from brittle wax-like to plastic behaviour and are related with the chemical structure. Poly(hydroxybutyrate) (PHB) for example is a highly crystalline (up to 70%) and stiff, but brittle, polymer with high T_m of 175°C. Copolymerization and the presence of long side chains can disrupt the crystal structure and decrease T_m and Young's modulus. Poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV) and poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (PHBHHx) are promising copolymers with decreased T_m and Young's moduli.[8] The chemical structures of both polymers are illustrated in Table 1. A case is mentioned where T_m is decreased from 175°C to 96°C by incorporating 34 mol% hydroxyvalerate units.[8] Table 4 shows the chemical composition of the PHA's, considered in food packaging.

Seal research on PHAs is very rare. One study shows that the seal initiation temperature of a film with PHBV in the seal layer is 40–50°C higher, compared to polyolefins. This corresponds closely with the difference in T_m. Heat sealing can be achieved in a temperature window between 190°C and 230°C, at a seal pressure of 275 kPa and a seal time between 1 and 2 s.[132] In one of our previous studies, a very thick PHBV film of 264 μm, blended with PBAT, is heat sealed. Seal initiation temperature, peak hot tack temperature and peak hot tack strength are achieved with respective values of 185°C, 135°C and 0.37 $N.mm^{-1}$.[111] With a 3:1 PLA:PHBV blended seal layer, very low seal strengths around 0.01 $N.mm^{-1}$ are reported.[133] Sealed films with poly(3-hydroxybutyrate-co-4-hydroxybutyrate) are used in a shelf life study, but no information is shared on the seal performance and settings.[134] Another study evaluates chemical structure and molar mass of PHBV, before and after conductive heating, but has not found any changes.[121]

4.5 | Other polyesters

PCL and PBAT are biodegradable polyesters that are used as blend component in biodegradable packaging. Both polymers can be used to increase toughness and/or to decrease seal initiation temperature of biodegradable seal layers. The influence of these components is described above in relation to PLA.[93,109–113]

In a rare seal study, a series of poly(hexylene 2,5-thiophenedicarboxylate-cobis(2-hydroxyethoxybenzene) (PTBH) copolyesters are synthesized and compared on seal strength. Ranges of T_g and T_m vary between 25–112°C and 160–200°C, respectively. Seal initiation temperatures between 117°C and 120°C are reported. Seal strength is correlated with MW. Seal strengths values between 0.3 and 1.0 $N.mm^{-1}$ can be achieved at seal temperatures between 117°C and 140°C, at constant seal time of 1.0 s.[135]

TABLE 4 Chemical composition of polyhydroxyalkanoates considered in food packaging.

	R1	R2	X
PHB	$-CH_3$	$-CH_3$	1
PHV	$-CH_2-CH_3$	$-CH_2-CH_3$	1
PHBV	$-CH_3$	$-CH_2-CH_3$	1
PHHx	$-CH_2-CH_2-CH_3$	$-CH_2-CH_2-CH_3$	1
PHBHHx	$-CH_3$	$- CH_2-CH_2-CH_3$	1

5 | OTHER HEAT SEALABLE POLYMERS

This section describes non-polyester biodegradable polymers: polysaccharides and proteins.

5.1 | Polysaccharides

5.1.1 | Starch

Starch is a semicrystalline polysaccharide that can be obtained from renewable sources. Starch-based films have an amylose and amylopectin fraction. Increasing amylose content improves the crystallinity and the mechanical and barrier properties, because of the morphology and interchain bonds between the molecules. Amylose is a linear long chain with interchain hydrogen bonds, resulting in a more dense and stronger structure. Films with increased amylopectin content are less crystalline because of the branched structure, caused by additional α-1,6-glycosidic bonds, and the smaller amylopectin chains.[136] Both structures are shown in Table 1. The amylose content depends on the botanical origin. The amylose contents range between 15% in cassava, 15–30% in water chestnut and sweet potato and around 30% in mung bean.[137] In its natural state, polymer degradation occurs before melting. Therefore, it's necessary to use blends and/or to add plasticizers to make starch films heat sealable.

A first parameter is the influence of the **moisture content** in relation to the seal strength and crystallinity. Cassava starch chains are mobile, resulting in a low seal initiation temperature, compared to mung bean starch. Water acts as plasticizer; therefore, high relative humidity decreases seal initiation temperature.[137] The influence of the film production process, mainly focussing on the amount and composition of plasticizers, such as glycerol and sorbitol, on seal performance, checked at jaw temperatures in the range of 50–160°C, is subject in various studies. Plasticizing is needed to toughen the film and avoid brittle behaviour of sealed samples.[138–141] Seal strength increases at high plasticizer concentrations. Seal strengths between 0.1 and 0.5 $\mathrm{N.mm^{-1}}$ are reported.[138,141] Seal initiation temperature can be correlated with the first endotherm in a DSC thermogram.[138,141] Another study evaluates the influence of hydrothermal treatment, resulting in irreversible modifications that can improve the film properties. Heat moisture and annealing treatment slightly increased seal strength.[142]

There are other studies on the influences of potential reinforcements in biodegradable films, such as **blends** with cellulose, chitosan, bagasse particles, natural fibres, graphene oxide and/or **nanoparticles**.[143–153] Pure corn starch and its blends with amylose, cellulose and methylcellulose achieve maximum seal strength values between 0.2 and 0.4 $\mathrm{N.mm^{-1}}$ at a jaw temperature of 166°C, using an impulse sealer.[143] In a study with cassava starch, reinforced with cassava bagasse particles, incorporation of medium sized particles, with a size between 53 and 250 µm, using a vibratory sieve to separate different particle sizes, led to increased seal strength, compared to neat samples or samples with incorporation of small or larger particles.[144] In a study that blended cassava starch and cellulose fibres to produce sealed bags, optimal seal strength is achieved at 150°C and a contact time of 24 s.[145] Another study on a similar film of blended cassava starch and cassava fibres achieved relatively low maximum seal strength, around 0.04 $\mathrm{N.mm^{-1}}$.[144] Seal strength of films, using an impulse sealer, of mung-bean starch, crosslinked with citric acid, are maximum, achieving a value between 0.4–0.5 $\mathrm{N.mm^{-1}}$, with 3% citric acid content.[147] In blends of corn starch and chitosan, another polysaccharide with good film forming ability, seal strength increases with the proportion of corn starch.[153] Another study checked the seal performance of a blend of potato starch, chitosan and graphene oxide. A maximum seal strength of 0.17 $\mathrm{N.mm^{-1}}$ is achieved with a blend ratio of 75:25 starch:chitosan. Incorporation of graphene oxide decreases seal strength down to 0.04 $\mathrm{N.mm^{-1}}$.[151]

The presence of **nanoparticles** can increase the seal strength of starch films, which is demonstrated for nanoclay and ZnO nanorods. With nanoclay, seal strength is correlated with nanoclay content, achieving a maximum value at maximum nanoclay content of 5%, while in the study on ZnO nanorods, only the sample with a ZnO content of 1% achieved an increased seal strength. In both cases, maximum values around 0.5–0.6 $\mathrm{N.mm^{-1}}$ can be obtained.[148,150] Another study highlights the influence of the botanical source of starch, related to the presence of nanoclay or nanosilicondioxide particles. Tapioca starch achieves the strongest increase of seal strength by incorporation of these nanoparticles, while the results with sago, potato and mung bean starch are almost equal to the seal strength the neat samples.[152] In another study on the influence of green synthesized silver nanoparticles, heat sealability is not affected by the presence of these particles.[149]

In addition to blending with other biopolymers, starch is blended as minor component with LDPE to increase the degradability of the overall film. One study reports that heat sealing is not affected by incorporation of corn starch, considering elongation as quality criterium,[154] while another study incorporating potato starch, glycerine and itaconic acid reports a reduction in seal strength. Temperature needs to be increased to achieve sufficient seal strengths.[155]

Starch is an emerging polymer in heat seal layers. It can be obtained from various botanical sources, and plasticizer is used to achieve a well performing seal. The influences of blends with other materials, mainly biodegradable reinforcements, are subject of various studies that highlight the potential of forming a seal of moderate strength, up to 0.6 $\mathrm{N.mm^{-1}}$.

5.1.2 | Cellulose, chitosan and other polysaccharides

Cellulose is a semicrystalline polysaccharide that can be obtained from renewable resources. It is mainly used in food packaging as a structural component because of its low cost, thermal resistance, mechanical potential and biodegradability. It is not sealable in its natural state.[156,157] Derivatives, such as cellulose ester, and/or blends, with poly(vinylalcohol), other polysaccharides or proteins, are used in the few available studies to heat seal.[158–163] Zero to very low strengths are reported, up to 0.12 N.mm^{-1}, of seals with cellulose esters.[159] Another study relates the fusion endotherm around 229°C in a DSC thermogram with the heat sealability of cellulose acetate film.[160] Seals of blended films of soy protein isolate (SPI) and carboxymethyl cellulose (CMC) initiate at 180°C and achieve plateau levels around 220°C. There is a positive correlation between seal strength and carboxymethylcellulose content. However, maximum seal strengths, with a value up to 0.2 N.mm^{-1} for blend ratios of 50:50 SPI:CMC, are rather low.[161] Another blend with κ-carrageenan, hydroxypropyl methylcellulose and the extract of *Prunus maackii* pomace, achieves seal strengths, using an impulse sealer, up to 1.1 N.mm^{-1}, depending on the blend ratio.[163]

Chitosan is a semicrystalline polysaccharide that can be produced by deacetylation of chitin, which is the most biosynthesized polymer after cellulose and lignin. Chitosan degrades before it melts, very similar to starch and cellulose. Therefore, it is not sealable, and seal studies use chitosan blends and/or add plasticizers to produce sealable packaging films.[164]

Seal strength of pure chitosan is compared with plasticized chitosan, by adding glycerol, and a chitosan-glycerol-gelatin blend. Additionally, CO_2 treatment of film forming solutions is evaluated. This treatment decreases seal strength. The authors suggest weaker hydrogen bonds as underlying cause of this decrease. Pure chitosan film has a seal strength below 0.01 N.mm^{-1}, and plasticization increases seal strength up to 0.07 N.mm^{-1}. A maximum but still relatively low value of 0.11 N.mm^{-1} is achieved with the blended film. Seal strength improvement is related to enhanced molecular diffusion and stabilization by hydrogen bonding.[165,166] Two studies use chitosan as minor component in blends with PLA. In one of these studies, a decreased seal strength is caused by chitosan addition, related with the difference in hydrophobicity of both polymers.[167] The other study adds polyphenol:chitosan blends in a PLA matrix and observes decreased seal strengths with higher amounts of chitosan in blends. Unevenly distributed fine particles on the surface are suggested as underlying cause of reduced seal strength.[168] In blends of chitosan and LDPE, seal strength decreases with higher amounts of chitosan. This decrease is related with the difference of hydrophobicity between both polymers.[169]

Soybean polysaccharide and cassia-gum are examples of other polysaccharides that can be used in combination with plasticizers, mainly glycerol, sometimes combined with sorbitol, to produce heat sealable packaging films.[170–173] Seal strength of plasticized soybean polysaccharide films increases with higher amounts of nanorod-rich ZnO, up to 0.8 N.mm^{-1} with 4% nanorod content, related with increased moisture content, resulting in more flexible and better heat seal processable films.[171] Another study reports a slight and insignificant increase of seal strength in soybean polysaccharide films with higher amounts of halloysite nanoclay (HNC), up to 0.8 N.mm^{-1} with 5% HNC. Seal strength results are also similarly related with moisture content.[173] Seal strength of plasticized cassia gum films increases if 2% or 4% of carboxylated cellulose nano crystal whisker (C-CNCW) are added, up to a relative high value of 2.2 N.mm^{-1} with 4% C-CNCW, but decreases with the addition of 6% C-CNCW. The underlying causes of the increase and decreases are, respectively, hydrogen bonds between cassia gum, glycerol and C-CNCW and heterogeneous dispersion at 6% C-CNCW.[173]

5.2 | Proteins

Most studies on protein-based seal layers use gelatin as main component. **Gelatin** is a protein fragment that is produced by partial or full hydrolysis of collagen. Gelatin has film forming capacity, mainly attributed to the presence of hydrogen bonds. However, gelatin film is very brittle and moisture sensitive. Therefore, packaging performance of blended films and addition of plasticizer are evaluated.[174] Some studies show thermal transitions of these films, obtained by DSC. However, not all of these studies relate thermal transitions with seal performance. One study shows T_g of gelatin films with or without the addition of ZnO nanorods, up to 5% content, between 59°C and 63°C. However, no information is shared on seal temperature. The maximum seal strength of the neat gelatin film is close to 1 N.mm^{-1} and can be increased up to 1.4 N.mm^{-1} with the addition of 1% ZnO nanorods. The increase is related to improvement of hydrogen and other bonds at the surface. The reduction of seal strength at increasing content of ZnO nanorods is related with reduced moisture content and the resulting reduced flexibility.[148] Another study reports T_g, obtained with a dynamic mechanical thermal analyser, of plasticized bovine gelatin films, with and without the addition of curry powder to flavour oven bags. T_g increases from −39.2°C, without curry, up to −31.7°C if 4% curry powder is incorporated. The authors suggest that the cause of this increase is related to the plasticizing effect of water, in decreased moisture content. This information is not related with thermal sealing behaviour. Very low seal strengths are reported, around 0.1–0.2 N.mm^{-1}, with no significant effect of curry powder addition.[175] DSC thermograms are shared of blended films of porcine skin gelatin and soy bean polysaccharide. T_g varies between 49.2°C for soy bean polysaccharide and 54.4°C for gelatin films. Seals of pure and blended films are made at 150°C and 1.5 s. Seal strength increases with higher amounts of gelatin until a plateau value, around 0.6 N.mm^{-1}, is reached at 60% gelatin content.[176] T_g and T_m are obtained by DSC in a study on plasticized fish gelatin films with and without incorporation of 0.2% epigallocatechin gallate (EGSG), a flavonol that can prevent lipid oxidation. For neat gelatin film, T_g and T_m of, respectively, 45.54°C and 129.78°C are reported, whereas the film with EGSG has a T_g and T_m of, respectively, 51.86°C and 129.76°C. The authors suggest that gelatin chain mobility is restricted by adding EGSG because of the facilitation of protein–protein interactions in the film matrix. Maximum strength of samples, sealed well above the melting point, at 150°C and 1.25 s, increases with higher amounts of EGCS, from 0.46 N.mm^{-1} with neat gelatin film up to 0.63 N.mm^{-1} with incorporation of 0.2% EGCS. The high amount of hydroxyl groups in EGCS and the resulting interactions via hydrogen bonding in the seal area are suggested as underlying cause of increased seal strength.[177] A melting temperature of 94°C is reported of a dried coating blend of gelatin, glycerol and lipid. Seals, produced at 90°C, achieve maximum strength with 7.5% glycerol and 1% lipid content.[178]

In another study of the same authors, the influence of seal settings of a blended film with **pig skin gelatin**, acetic acid esters and glycerol is evaluated. Temperature is varied between 70°C and 110°C but has a minor positive effect on seal strength. Maximum seal strength varies between very low values in the experimental design, from 0.02 to 0.04 $N.mm^{-1}$. Seal pressure affects seal strength the most, in a negative way. There is no effect of seal time.[179] The influence of blend ratio in films with bovine gelatin, glycerol and epoxidized soy bean oil (ESBO) on seal strength is studied. Seal strength increased significantly by replacing glycerol partially with ESBO, from 0.26 $N.mm^{-1}$ with 40% glycerol and 0% ESBO, up to 0.69 $N.mm^{-1}$ with 20% glycerol and 20% ESBO. The increase in seal strength is related with increased rigidity because the plasticization effect attenuates when glycerol content decreases.[180] A seal study on blends of gelatin and konjac glucomannan (KGM), which is a polysaccharide, shows a maximum seal strength, sealed at temperature and time of 140°C and 1 s, at 40% KGM content, below 0.1 $N.mm^{-1}$. This value is relatively low but a remarkable increase, related to intermolecular interactions between KGM and gelatin, compared to films with 0 or 100% KGM.[181] The influence of nanoparticle content on the seal strength of plasticized bovine skin gelatin films is subject of another study. Addition of HNC or nano-SiO2 decreases seal strength, from 0.7–0.8 $N.mm^{-1}$ at 0% content down to 0.3–0.4 $N.mm^{-1}$ at 5% content. The authors suggest a tendency to block interactions between melted surfaces as underlying cause of the decrease.[182] Another study evaluates the influence of blend ratio and pH on seal strength of a blended film of chicken protein isolate (CPI) and fish skin gelatin. CPI is not sealable because of the restricted mobility, caused by disulphide bonds. Seal strength increases with higher gelatin content, up to 0.25 $N.mm^{-1}$ for 100% gelatin content. The increase is related with a higher intermolecular interdiffusion, an increased degree of entanglement and more interfacial interactions. A pH 3 of the film forming solution increases the seal strength. The decreased strength at pH 11 is related with the potential formation of Maillard reactions between amino groups of proteins and carboxyl groups of lipid oxidation products at alkaline conditions during film production, resulting in cross linking and reduced molecular interdiffusion, entanglement and interaction across the seal surface.[183] The Maillard reaction is purposely applied in a study to modify blended film, containing fish skin gelatin, L-arabinose and plasticizers (sorbitol and glycerol). A design-of-experiment approach is followed to optimize seal strength with a vacuum packaging machine, prior to shelf life tests with milk powder. An optimal seal strength, slightly below 0.5 $N.mm^{-1}$, is achieved at vacuum, heat sealing and cooling times of, respectively, 6.89, 2.64 and 3.06 s.[184] Other shelf life studies report heat sealability of packaging films with gelatin-based seal layers without sharing details on seal performance.[185,186]

In addition to gelation, **SPI** can also be used in sealable films for food industry. Plasticizing and/or blending is needed to improve heat sealability.[175,183,187,188] Similar to the use of polysaccharides, it is important that sealing occurs without degradation. Two studies report melting temperatures of films blends of SPI, poly(vinyl alcohol) (PVA) and glycerol, below 170°C. The influence of jaw temperature is related to the melting point. Strong seals are obtained above a jaw temperature of 220°C. Seal strength increases with higher amounts of PVA, up to a value around 1.4 $N.mm^{-1}$ in films with 30% PVA. The seal principle of the blend is based on entanglement of long-chain molecules, equal to other seal polymers, and additional hydrogen bonds.[187,188] A seal study on plasticized whey protein isolate films compares DSC-thermograms of films with sorbitol and glycerol. The onset melting temperatures with glycerol, from 108°C to 122°C, are slightly lower than those with sorbitol, around 126–127°C. These values are related with the maximum seal strength. Maximum values, around 0.3 $N.mm^{-1}$, are obtained at 110°C for glycerol and 130°C for sorbitol.[189] DSC thermograms of films, composed of SPI, glycerol and the incorporation of diatomite and/or thymol, as antimicrobial additives, exhibit melting endotherms with peak values between 116.2°C and 121.1°C. Seal strength is optimized; a relatively low maximum value around 0.16 $N.mm^{-1}$ can be obtained at jaw temperature of 139.5°C, seal time of 2.5 s and seal pressure of 0.48 $N.mm^{-2}$.[190] Another study measures glass transition temperatures of SPI, plasticized with glycerol, around −20°C, but no relation is made between thermal properties and seal behaviour. Very low seal strengths, below 0.1 $N.mm^{-1}$, are reported.[191] In a rare seal layer blend composition of soy protein, CMC, polycarboxylic salt and glycerol, maximal seal strength values, between 1.1 and 1.6 $N.mm^{-1}$, can be obtained between 110°C and 130°C.[192] Another rare seal layer blend composition, with semolina flour, with a respective protein and gluten content of 14.2% and 18.5%, up to 5% kaolin nanoclay, as potential improvement of the physicochemical properties, and the plasticizers glycerol and sorbitol, is evaluated on seal strength. A maximum seal strength, slightly below 0.4 $N.mm^{-1}$, can be obtained at 2% kaolin clay content. The enhancement at 2% is related with hydrogen and other bonds on the surface. The decrease at 3–5% kaolin content is related with the reduction of moisture content and resulting film flexibility.[193] Another study finds a narrow optimal jaw temperature range, between 120°C and 130°C, to seal corn zein films. In this range, a seal strength of 0.3 $N.mm^{-1}$ can be obtained.[194]

Concluding for non-polyester biodegradable materials, such as polysaccharides and proteins, plasticization and/or blends with other heat sealable materials are needed to produce well sealable materials for food applications. The seal principle of these materials is, similar to polyolefins and polyesters, mainly based on entangling long chains. Increasing the mobility of the polymer chains, by increasing the moisture content or plasticizer content can improve the seal performance. In addition, further improvements are possible by blending other polymers and/or by adding nanoparticles.

6 | ADDITIVES

Additives and processing aids are organic or inorganic molecules that are added in small amounts to the polymer matrix to tailor the

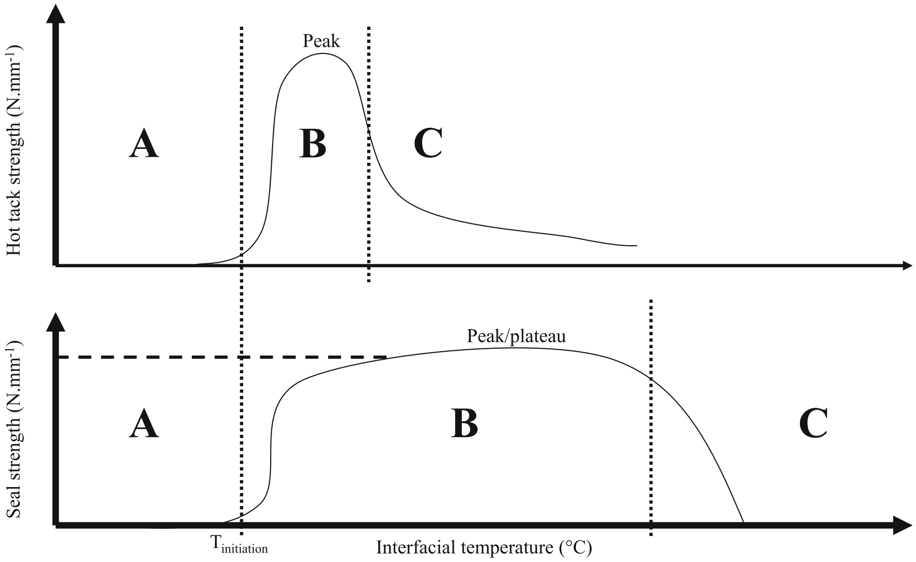


FIGURE 5 Simplified representation of temperature dependence of hot tack and seal strengths of thermoplastic films: low values below seal initiation temperature (A), high values in process window (B) and decreased values at high temperature (C).

properties of packaging films and/or improve the processing. Stabilizers, modifiers, such as pigments, opacifiers, slip agents, antiblock, chill roll release, lubricants, antistats, process aids, nucleating agents, clarifying agents, antifog, tackifiers, tougheners and fillers are well known examples.[7]

As described in the former section, **plasticizers**, such as sorbitol and glycerol, are critical to produce flexible and sealable polysaccharide and/or protein films. Plasticization is required to mobilize the chains sufficiently, required to entangle during sealing. Few seal studies on other polymers describe the influence of plasticizer. One study describes the influence of poly(ethylene glycol) (PEG) plasticizer, on the ultrasonic seal performance of PLA. It broadens the seal processing window. The minimum amplitude to seal decreases from 18 to 13 μm by adding PEG, compared to pure PLA. The maximum amplitude remains 32 μm for plasticized PLA.[116] Isophtalic acid (IPA) is another example of a plasticizer that is used in a seal study. Addition of IPA decreases crystallinity and T_m of polyesters that is critical to increase the seal performance. IPA can also make the whole composition dielectric sealable, a technology with currently no applications in flexible food packaging.[195]

Tackifiers can change the viscoelasticity of the seal polymer and thus impact seal performance. **Lubricants** can increase intermolecular slippage and thus decrease adhesion. **Slip agents**, like fatty acid amides, bloom to the surface, where sealing occurs, and can interfere sealing if too much is used. Lubricants and processing aids, like silicon oils, can interfere as well by contaminating the seal interface.[6]

Other interesting additives include **fillers**. Different fillers, such as calcium carbonate, talc, mica and glass fibre, have been evaluated on the ultrasonic weld performance of PP. Though the samples were injection moulded, which deviates from the thin packaging films described above, it gives a first indication of the influence of fillers on seal performance. Weld strength, expressed in N.mm^{-2}, and elongation decrease by adding filler and by increasing the filler concentration. This is attributed to the prevention of PP bonding. At extreme filler concentrations of 40%, maximum weld strength decreased to 18, 10 and 6 N.mm^{-2} for calcium carbonate, mica and talc, while unfilled PP achieved 23 N.mm^{-2}.[196] Reduced seal strength, caused by blocking interfacial interactions through the addition of non-sealable particles, is described in another study.[182] A recent study on seal performance of PP and LDPE films with talc fillers finds an increase of hot tack and seal strength with talc filled samples. Increased performance is related with (i) the enhancement of free volume in the polymer matrix, resulting in improved diffusion; (ii) increased compatibility between different polymer types, caused by the high surface interactions of filler and both polymers; and (iii) improved heat transfer because of the high thermal conductivity of talc, compared to PE and PP. They show that the seal initiation temperature slightly increases if talc is incorporated, which is caused by a different wetting behaviour and a decrease of the polymer–polymer contact ratio at the seal interface.[197] Increased thermal conductivity is also reported in a study on the influence of clay nanoparticles. Peelable seals can be obtained over a broad jaw temperature process window, over 100°C at 3% organoclay content with partially exfoliated particles, as alternative to PB-based cohesive peel systems.[198] The possibility to produce broad-range peealable seals by adding nanofiller is also described in another study.[199] With the potential impact on surface chemistry and crystallinity, among other aspects, additives can impact seal performance. This is uncharted territory in open literature.

7 | CONCLUSION

Seal performance of thermoplastics is mainly evaluated in literature with hot tack and seal strength experiments. The dependence of strength with temperature is related with thermal transitions, mainly obtained by DSC.

Figure 5 is a simplified representation of the temperature dependence of hot tack and seal strengths of thermoplastic films. These values cannot be compared in one graph because of the different process parameters, such as cooling time and grip separation rate.

Absolute values of hot tack strength are typically much lower than seal strength, mainly as a result of the influence of temperature on the seal and mechanical strength of thermoplastic film. However, Figure 5 illustrates strength values of equal relative magnitude, to increase readability. Temperatures in areas A and C are not appropriate for sealing, resulting in low strengths. Area B is a temperature range that can be considered for sealing. The intersection between areas A and B is determined by polymer diffusion. The intersection shifts to the left if the mobility of the polymer chains at the seal surface is high. This is related to the thermal transitions of thermoplastics: T_g for amorphous polymers and T_m for semicrystalline polymers. Copolymerization, addition of plasticizer and/or blending, can push seal initiation to lower temperatures.

Area B corresponds with the process window with moderate to peak strength values. In general, this window is narrow for hot tack and wide for seal strength.

For hot tack, the intersection between areas B and C is determined by melt strength. Melt strength is negatively correlated with temperature; therefore, hot tack strength decreases at increasing temperatures (right side of area B). Melt strength is relatively low for polymers without additional chemical forces, such as hydrogen bonds, polar and/or ionic interactions. These interactions result in high melt strength and wide hot tack processing windows, which is desired to ensure seal quality when it becomes pressurized while it is still hot. PE and ionomer are examples of polymers with respective low and high melt strengths, characterized by, respectively, narrow and wide hot tack processing windows.

Melt strength is not relevant in seal strength tests because the seal is cooled down. Therefore, the process window is broader than hot tack, and the intersection between areas B and C is determined by other properties. Thermal stability and squeezing-out of seal material are examples of limitations for seal strength. In a multilayer, degradation and/or melting of other layers is also of critical importance.

Peak values of hot tack and seal strength can be increased by specific modifications of the seal material, for example, by (i) increasing the crystallinity; (ii) increasing the diffusion and entanglement density of long linear chains across the interface; (iii) introducing extra chemical forces, such as hydrogen bonds, polar and/or ionic interactions. The latter option is more important for hot tack because these forces are strongly related with melt strength. High strength is desired to ensure seal quality when it becomes pressurized. In peel applications, low seal strengths are desired for optimal convenience.

Nevertheless, some of the above described enhancements are counteracting; for example, increased crystallinity can restrict chain mobility and thus increase the seal initiation temperature, but on the other hand, it can also increase peak strength values.

Besides the relations of strength and temperature illustrated in Figure 3, rheology is also crucial in optimizing seal performance. Low viscosity is needed to fill up gaps at wrinkles, gusset areas and/or encapsulate contamination.

The seal failure mechanism, such as the presence of angel hair during cohesive peeling, delamination and material break, is another example of an important seal characteristic.

In addition to optimizing seal strength and seal temperature based on profound insight in material properties, transparency and gloss of seal materials are visual characteristics that play an important role in material selection for food packaging, because the seal material is in general omnipresent over the full packaging surface. Other important criteria in selecting seal materials are cost, availability, food contact, coefficient of friction, hydrophobicity, end-of-life options and processability.

In conclusion, there is currently no seal material available showing optimal performance across all criteria for all food packaging applications. Depending on the application, a pragmatic selection must be made by (i) identifying and hierarchizing important criteria; and (ii) matching an appropriate seal material with these criteria. The analysis of the current available knowledge and insights in seal performance of polyolefines, ethylene copolymers, polyesters and other heat sealable polymers, which is provided in this review, can support scientists and different stakeholders in the food and packaging industry to improve and optimize the production and the efficiency of the seal process of sealable materials in flexible packaging.

DATA AVAILABILITY STATEMENT

Data sharing is not applicable to this article as no new data were created or analysed in this study.

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