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BAMPS, Bram; D'huys, Karlien; De Ketelaere, Bart; ADONS, Dimitri & PEETERS, Roos (2017) Evaluation of the Ultrasonic Sealing Performance of Flexible Films with Polyolefin Seal Layer. In: Unlocking the Full Potential of Packaging Across the Value-chain, School of engineering and management Vaud (HEIG-VD),p. 477-488.

Handle: http://hdl.handle.net/1942/28294

Evaluation of the ultrasonic sealing performance of flexible films with polyolefin seal layer

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Abstract: Recent studies have shown advantages of ultrasonic sealing over heat conductive sealing, namely sealing through contamination, decreased energy consumption, no need for thick peel layers. Up to now there is no efficient systematic methodology to determine the optimal settings for the ultrasonic sealing of flexible films. Besides that, almost no literature is available about the specific permeation of the seal area in a package.

In this study, seal strength, compaction and/or energy consumption are investigated as determining parameters in optimizing the ultrasonic sealing (USS) performance of flexible packaging films. These parameters are used to evaluate the ultrasonic seal performance of polyolefin monolayer and multilayer materials – 60 µm monolayer polyethylene (PE) and polypropylene (PP) polyolefin films, laminated films of 24 µm polyethylene terephthalate (PET) or oriented polyamide (OPA) with 40 µm PE seal layer. These materials are sealed with a 35 kHz ultrasonic sealer at 15 different settings of force, amplitude and time to calculate the optimum settings and to evaluate the ultrasonic seal performance. Besides that, the oxygen transmission rate (OTR) of ultrasonically sealed commercial pouches and flat films with and without the presence of a high barrier layer, ethylene vinyl alcohol (EVOH) or aluminum (Alu), in PET laminates with a PE sealing layer, is evaluated. This is measured with a Mocon OX-TRAN® device.

The optimization method is validated and ready for implementation. Metallocene linear low density polyethylene with C6 branches (mLLDPE-C6) and random copolymer polypropylene (raco PP) are selected as ultrasonic best performing polyolefin monolayers because of respectively mainly a lower energy consumption and a broader seal window compared to other tested films. PET was selected as the best performing laminate for ultrasonic sealing because of the lower energy consumption compared to OPA laminates. Ultrasonic seal strengths of OPA and PET laminates were similar. Strong ultrasonic seals have no impact on the OTR. This is illustrated by the OTR-results of high barrier PET/Alu/PE pouches at normal and extreme ultrasonic settings, respectively $0.66 \pm 1.56 \times 10^{-4}$ and $0.48 \pm 2.9 \times 10^{-4}$ cc/package.day.

Keywords: Seal permeation, Oxygen transmission rate, Ultrasonic sealing, flexible packaging, polyolefin seal layer, mono- and multilayer packaging material

1 Introduction

Packaging is a crucial step in the processing chain in the consumer goods industry. A proper closure of the package is essential to guarantee the safety and quality of the product throughout shelf-life. Flexible packages are more energy efficient compared to premade packages, such as glass and metal containers, because they are transported flat or in reel form and because the gross weight of packed product in non-plastic packaging and managing the resulting packaging waste requires a higher energy use [1].

The most common method used for achieving closure of a flexible packaging concept is heat sealing. Several technologies can be used to heat seal a package: conductive heating, impulse heating, hot air blast heating, ultrasonic heating, induction current heating, electrical field loss heating and hot wire heating. The heat conductive method is currently the most popular process[2]. Heat sealability of flexible films with polyolefin seal layers was extensively studied in the last decades [3,4]. Ultrasonic sealing of flexible films has been the subject of recent studies in which advantages over the more common heat conductive sealing (HCS) (sealing through powder contamination, decreased energy consumption, no need for thick peel layers) have been proven [5,6]. In these studies, the seal strength and/or energy derived from a T-peel test was considered as a measure of the ultrasonic seal performance[7,8].

In this paper, a methodology was developed to optimize the ultrasonic seal performance and to evaluate the ultrasonic seal performance of specific sealing materials in a monolayer and/or laminated flexible film structure. Ultimately, a method to measure seal permeability for oxygen gas in order to evaluate the barrier function of a seal was also demonstrated in this paper.

2 Methods & Materials

2.1 Methods

Equipment

Overall thickness is determined using a calibrated MTS MI20 with a repeatability of maximum 2.5 µm. The ultrasonic samples are prepared with a 35 kHz TSP750E-100-1 (Telsonic Ultrasonics) with a 75x5 mm sonotrode with a flat surface. The anvil has a semicilindric energy director with a radius of 2.5 mm. The heat conductive samples are prepared with a HSE3 Heat sealer (RDM Test Equipment) with 300x25 mm teflonized flat to flat jaws. The jaws are protected with silicon tape. All samples are tested with a 10 M universal testing machine (MTS) with a load cell of 2 kN.
The oxygen permeability for flat films and packages is measured respectively with Ox-Tran Model 702 and Ox-Tran 2/20 ML System (Mocon) according to ASTM F1927[9] and ASTM F1307[10].

Thickness

The thickness of the film was measured at 10 equally distributed locations over the film surface and the average value and standard deviation were recorded.

Ultrasonic Sealing

Ultrasonic seals were prepared at 15 different combinations of the seal time, seal force and seal amplitude. These 15 settings were selected according to a Box-Behnken experimental design in order to efficiently obtain as much information as possible on the effect of the parameters based on a limited amount of experiments.

Table 1: Box-Behnken design of applied ultrasonic parameter settings

Chronology of sealing	Seal time (ms)	Seal Force ($N.mm^{-1}$)	Seal amplitude
1	200	2	36
2	300	4	18
3	100	4	36
4	200	2	18
5	200	4	27
6	200	4	27
7	100	6	27
8	200	4	27
9	300	6	27
10	200	6	18
11	100	2	27
12	200	6	36
13	300	2	27
14	300	4	36
15	100	4	18

Directly after sealing, all ultrasonic seals were cooled down at 2 $N.mm^{-1}$ for 500 ms.

Heat Conductive Sealing

11 sealed samples are made by varying jaw temperature from 100 °C to 200 °C with a 10 °C interval while seal pressure and seal time is kept constant at respectively 2.46 $N.mm^{-1}$ and 1000 ms.

Seal Strength

After sealing, samples are conditioned at 23°C and 50% relative humidity for 24 hours. Samples are cut to a width of 15 mm and tested at 300 mm.min^{-1} at a clamp distance of 10 mm. The maximum value of strength (N) is divided with the seal width (15 mm) to obtain the seal strength (N.mm^{-1}), according to ASTM F88[11]. Three seals for each ultrasonic or heat conductive setting are measured, the average value and standard deviation is recorded.

Ultrasonic Seal Performance

Along with the seal strength, the seal compaction, the energy consumption of the sealing process and the size of the seal window were also taken into account to evaluate the ultrasonic seal performance. The seal window is the amount of sealed samples (seal strength > 0,07N.mm^{-1}) of the 15 ultrasonic samples of the experimental design.

The seal compaction (µm) is the travel distance of the horn in the sample. The energy consumption is the energy consumed by the ultrasonic sealing process. Both parameters are derived from the machine output. Figure 1 shows these parameters at good sealing conditions (27 µm, 200 ms, 4 N.mm^{-1}). First, power is built up so the vibrations can be maintained in the material, and at the same time seal compaction increased fast. In the last stage of the curve, there is a steady state condition in which the power decreases and seal compaction starts to stabilize.

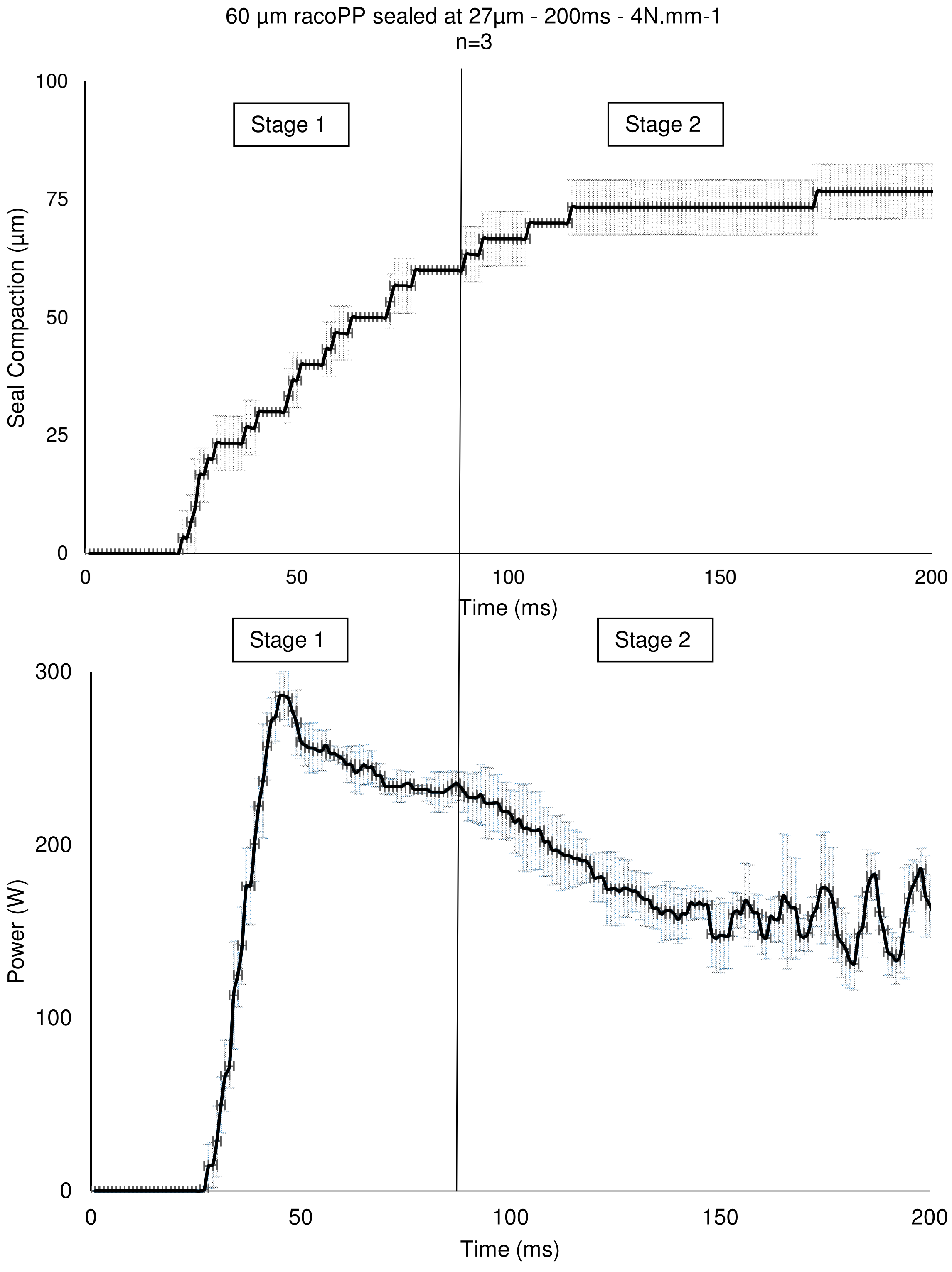

Figure 1: Output parameters (seal compaction and energy consumption) of random copolymer PP at 200ms_27µm_4N.mm^{-1} (n=3).

Seal optimization and validation

The sealing parameters can be optimized in order to achieve a maximal seal strength and/or in order to achieve a certain level of seal compaction or energy consumption. In this paper, the optimization methodology was firstly validated for obtaining the optimal sealing parameters for achieving the maximum seal strength. Next, the methodology was validated for obtaining the optimal sealing parameters that result in a compromise between seal strength and seal compaction. In these validation experiments seal strength desirability is a linear function from 0% (no strength) to 100% (maximum strength). In the second validation experiment seal compaction desirability is a linear function from 100% (no seal compaction) to 0% (50% compaction). The predicted seal strength and seal compaction are validated with the measured seal strength, according to ASTM-F88, and the measured (machine output) seal compaction.

Oxygen permeability

The oxygen permeability of flat films is tested according to ASTM F1927 with an external oxygen concentration of 100%, an external relative humidity of 50% and an external temperature of 23°C. The internal oxygen concentration, relative humidity and temperature were 0%, 0% and 23°C, respectively. The oxygen permeability of sealed packages is tested according to ASTM F1307 with an external conditioned atmosphere (21% oxygen, 50% relative humidity and 23°C) while internally oxygen concentration is 0%, relative humidity is 0% and temperature is 23°C.

The OTR of the flat film is recalculated according to the sealed package's dimensions and compared with the sealed package oxygen transmission.

Three commercial films are tested with the following oxygen barrier properties: one medium barrier film (PET/PE 12/60), one high barrier film (PET/PE-EVOH-PE 12/40) and one very high barrier film (PET/Alu/PE 12/9/75).The PET layer is in contact with the external atmosphere, while the PE seal layer is in contact with the internal atmosphere.

The dimensions of the sealed packages are shown in figure 2. The settings for heat conductive and ultrasonic sealing are respectively 1000ms_1N.mm^{-2}_150°C and 300ms_4N.mm^{-1}_36µm. Additionally two PET/Alu/PE pouches are sealed at extreme settings (until the film is cut through by the sonotrode) to check the oxygen permeability of extreme ultrasonic sealed packages. All seal settings give an average seal strength > 2 N.mm^{-1}.

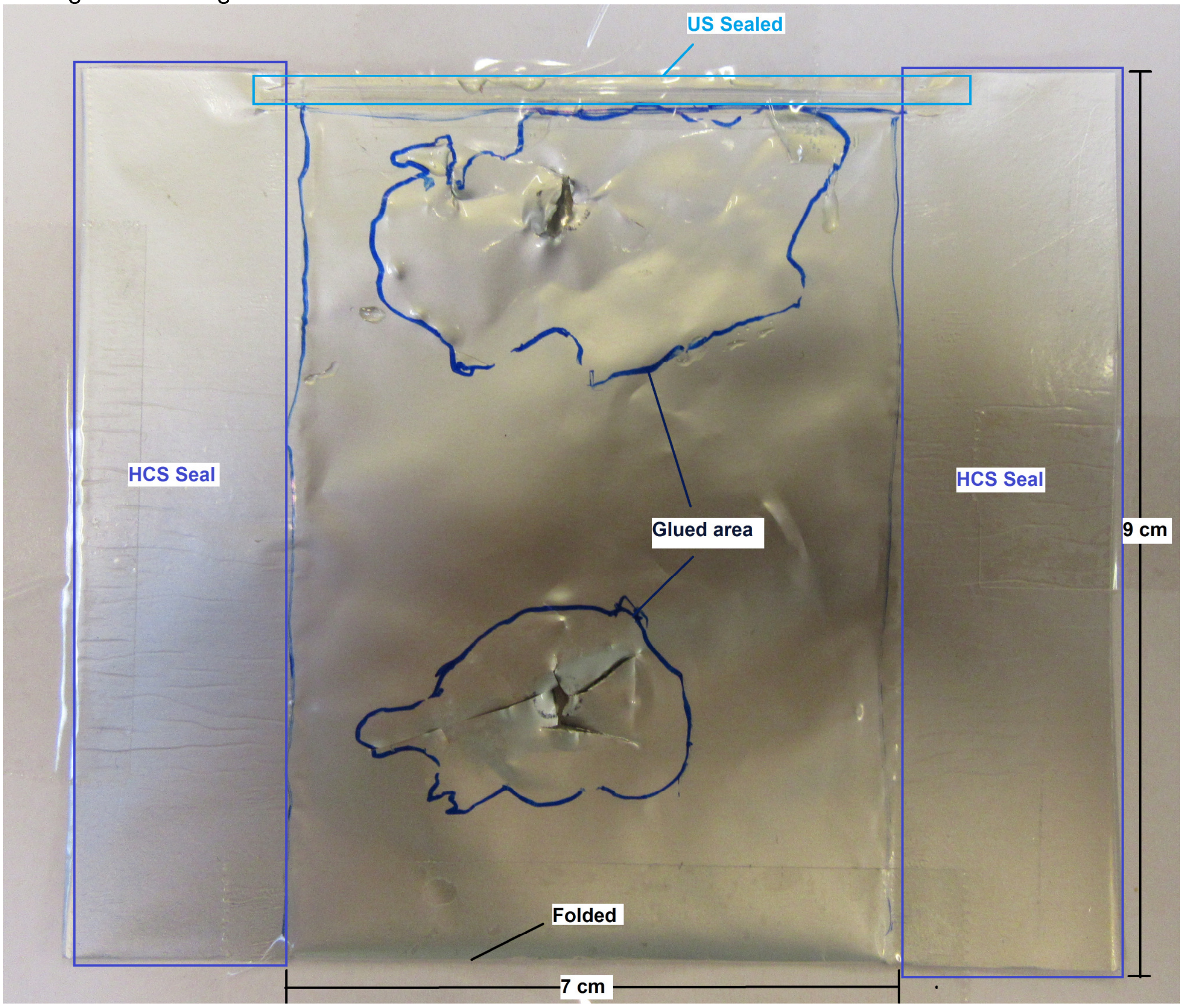

Figure 2: Dimensions of an ultrasonic and heat conductive sealed package

2.2 Materials

Flexible films

All films are listed in table 2.

Table 2: Composition, measured total thickness and production process of flexible films

Composition	Measured Total Thickness (µm) (n=10)	Production process
Monolayers		
LDPE 60	63 ± 2	Blown extrusion
mLLDPE-C6 60	64 ± 2	
LLDPE-C6 60	58 ± 3	
LLDPE-C4 60	63 ± 2	
Homopolymer PP (=homo PP) 60	61 ± 3	Cast extrusion
Random copolymer PP (=raco PP) 60	58 ± 3	
Plastic Laminates		
PET/LLDPE-C4 24/40	69 ± 1	Blown extrusion, corona pretreatment and lamination with Adcote 301/350
OPA/LLDPE-C4 23/40	69 ± 2	
PET/ mLLDPE-C6 24/40	69 ± 2	
OPA/mLLDPE-C6 23/40	71 ± 2	
PET/PE 12/60	76 ± 2	Commercial films, production process and specific composition is not known
PET/PE-EVOH-PE 12/40	55 ± 2	
PET/ALU/PE 12/9/75	105 ± 4	

3 Results

Ultrasonic seal optimization

Table 3 shows the predicted and the measured values for seal strength and seal compaction for two monolayer films: raco PP and mLLDPE-C6.

Table 3: Predicted and measured seal strength and seal compaction for random copolymer PP and mLLDPE-C6 monolayers sealed at optimum ultrasonic settings

Tested film (optimum ultrasonic settings)	Predicted seal strength ($N.mm^{-1}$)	Measured seal strength ($N.mm^{-1}$) n=9	Predicted seal compaction (µm)	Measured seal compaction (µm) n=3
Raco PP maximized seal strength (160ms_6N/mm_32µm)	1.54	1.56 ± 0.12	108	100 ± 10
mLLDPE-C6 maximized seal strength (200ms_5,7N/mm_30µm)	0.87	0.81 ± 0.17	65.8	60 ± 10
Raco PP Compromised seal strength and compaction (200ms_6,2N/mm_18µm)	1.41	1.44 ± 0.04	74	80 ± 10
mLLDPE-C6 Compromised seal strength and compaction (200ms_3,78N/mm_36µm)	0.72	0.88 ± 0.10	50	40 ± 10

The measured seal strengths are equal, with the exception of the seal strength of mLLDPE-C6 in the compromisation with compaction, as the predicted value while the measured distances are equal to the predicted values.

Ultrasonic seal performance

<u>*Monolayers*</u>

The results of the monolayers are shown in table 4 and figure 3.

Overall there are very few differences in seal performance of the PE monolayers, ultrasonic and heat conductive maximum seal strengths are similar. Metallocene LLDPE-C6 is selected as best performing ultrasonic PE monolayer because of the combination of a broad seal window, a good seal strength and low energy consumption.

The two PP films differ also very little in seal performance. Between the PE and the PP films there is a big difference in seal strength, especially for ultrasonic sealing. In contrary to heat conductive sealing, there is no need to insert more energy to PP than PE because of the differences in plateau initiation temperature (110-120°C and 130°C-160°C for respectively the tested PE and PP monolayers). Random copolymer actually has the broadest seal window (13/15) and reaches good seal strengths at low ultrasonic settings (low seal force, amplitude and seal time). This seal property is of interest for industrial purposes because of the increased process flexibility. Random copolymer PP is, although the seal strength is little lower, selected as best performing ultrasonic PP monolayer because of the broad seal window in comparison with homo-polymer PP.

Table 4: Comparison of size sealing window and max. seal strengths of polyolefin monolayers

	USS_{window}	$USS_{max\ parameters}$ ($N.mm^{-1}$)	$USS_{max\ Seal\ Strength}$ ($N.mm^{-1}$)	$HCS_{max\ parameters}$ ($N.mm^{-1}$)	$HCS_{max\ Seal\ Strength}$ ($N.mm^{-1}$)
LDPE	11/15	300ms_4N/mm_36µm	0.77 ± 0.01	1000ms_2,46N.mm^{-2}_130°C	0.81 ± 0.02
mLLDPE-C6	11/15	100ms_6N/mm_27µm	0.90 ± 0.03	1000ms_2,46N.mm^{-2}_120°C	0.86 ± 0.01
LLDPE-C6	11/15	100ms_6N/mm_27µm	0.89 ± 0.07	1000ms_2,46N.mm^{-2}_190°C	0.94 ± 0.05
LLDPE-C4	9/15	300ms_6N/mm_27µm	0.98 ± 0.06	1000ms_2,46N.mm^{-2}_170°C	0.90 ± 0.09
HomoPP	9/15	100ms_6N/mm_27µm	1.73 ± 0.09	1000ms_2,46N.mm^{-2}_160°C	1.59 ± 0.05
RacoPP	13/15	100ms_6N/mm_27µm	1.52 ± 0.07	1000ms_2,46N.mm^{-2}_160°C	1.24 ± 0.14

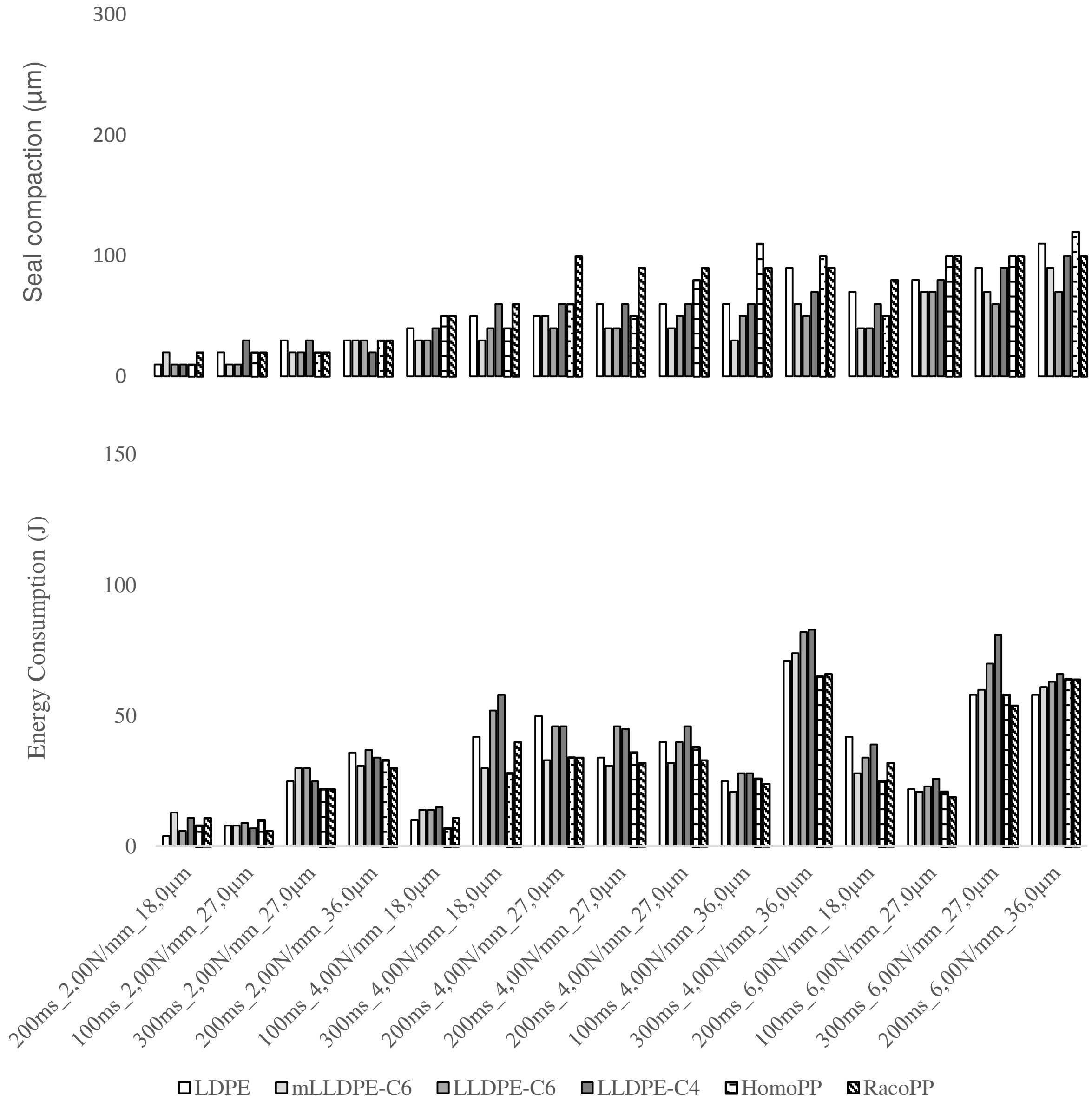

Figure 3: Comparison of seal compaction and energy consumption of polyolefin monolayers

Laminates

The results of the plastic laminates are shown in table 5 and figure 4.

All tested laminates have a broader ultrasonic seal window compared with the monolayers of similar thickness. (For LLDPE-C4: 14/15 with OPA and 11/15 with PET in comparison with 9/15 for the monolayer; For mLLDPE-C6: 13/15 with OPA and 15/15 with PET in comparison with 11/15 for the monolayer).

Compared with the monolayers the seal strength of the laminates is increased because of the lamination of a high strength outer layer of OPA or PET.

There is little difference in ultrasonic seal strength between both laminates. Compared with heat conductive strength, PET reaches a similar seal strength with ultrasonic sealing. For OPA the heat conductive seal strength is higher or equal to the ultrasonic seal strength, however deviations for OPA are high.

The seal compaction and energy consumption are shown in more detail in figure 4.
The OPA laminates have a slightly higher or equal seal compaction than PET in the US settings. The OPA laminates consume more energy than PET laminates with most of the US settings. Therefore PET is chosen as a better laminate layer for ultrasonic sealing.

Table 5: Comparison of size sealing window and max. seal strengths of plastic laminates

	USS_{window}	**$USS_{max\ parameters}$ ($N.mm^{-1}$)**	**$USS_{max\ Seal\ Strength}$ ($N.mm^{-1}$)**	**$HCS_{max\ parameters}$ ($N.mm^{-1}$)**	**$HCS_{max\ Seal\ Strength}$ ($N.mm^{-1}$)**
OPA/LLDPE-C4	14/15	200ms_6.00N/mm_36.0µm	1.98 ± 0.43	1000ms_2.46N.mm^{-2}_200°C	3.62 ± 0.17
OPA/mLLDPE-C6	13/15	300ms_6.00N/mm_27.0µm	2.94 ± 1.08	1000ms_2.46N.mm^{-2}_200°C	3.70 ± 0.47
PET/LLDPE-C4	11/15	300ms_4.00N/mm_36.0µm	2.73 ± 0.31	1000ms_2.46N.mm^{-2}_180°C	2.66 ± 0.06
PET/mLLDPE-C6	15/15	300ms_4.00N/mm_36.0µm	2.59 ± 0.06	1000ms_2.46N.mm^{-2}_200°C	2.81 ± 0.01

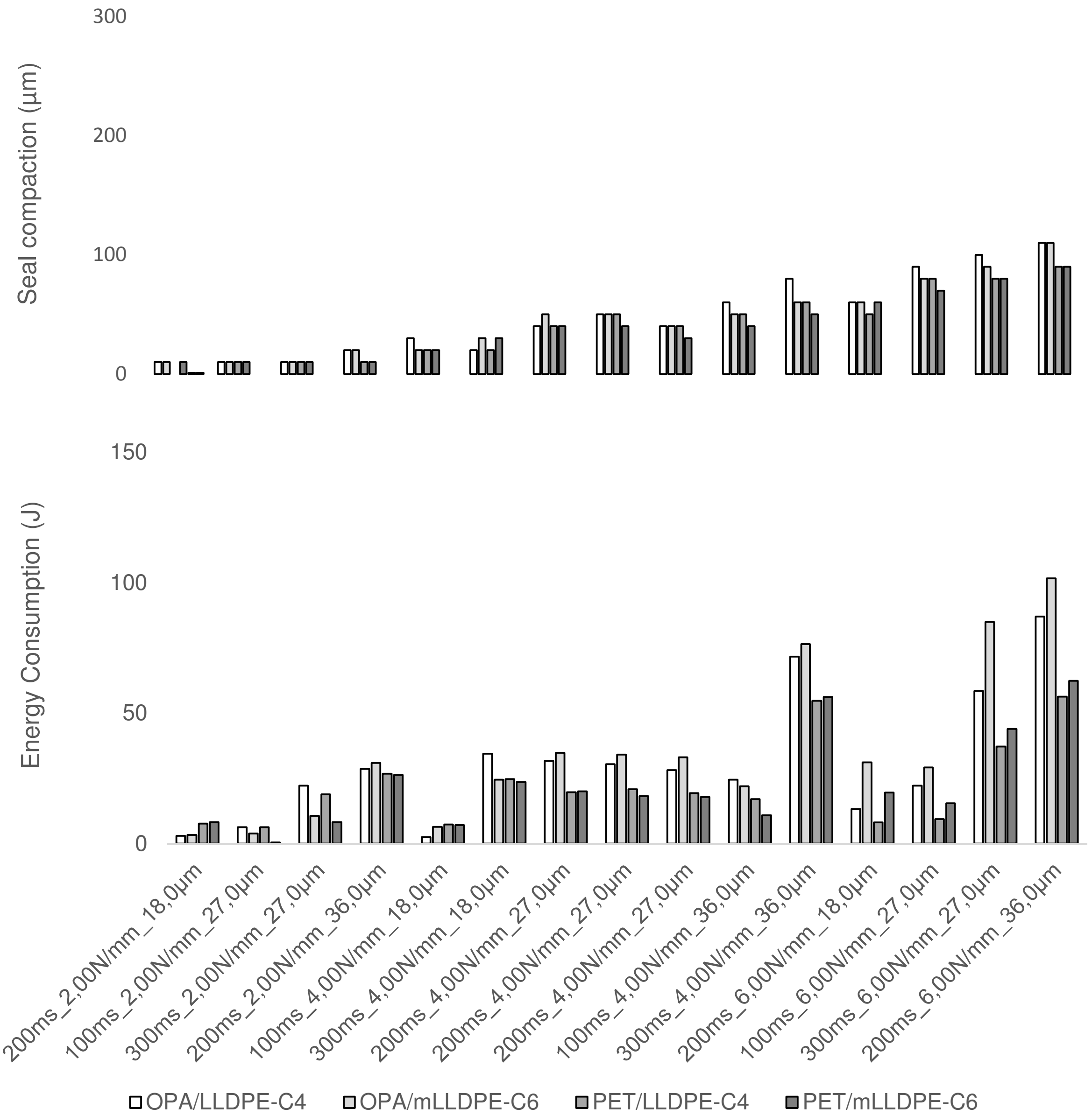

Figure 4: Comparison of seal compaction and energy consumption of plastic laminates

An interesting additional evaluation is the comparison of energy consumption of laminates over monolayers shown in table 6. Most of the laminates have a lower energy consumption than the monolayers. This could be a consequence of a better conduction of the ultrasonic vibrations by the stiffer laminates. To confirm this, however, further research is necessary.

Table 6: Comparison of proportion of the energy consumption of ultrasonic sealing of the laminated over the monolayer structure

	OPA/LLDPE-C4	**OPA/mLLDPE-C6**	**PET/LLDPE-C4**	**PET/mLLDPE-C6**
200ms_2,00N/mm_18,0µm	27%	25%	69%	64%
100ms_2,00N/mm_27,0µm	90%	49%	90%	6%
300ms_2,00N/mm_27,0µm	89%	36%	76%	28%
200ms_2,00N/mm_36,0µm	84%	100%	79%	85%
100ms_4,00N/mm_18,0µm	17%	46%	49%	50%

300ms_4,00N/mm_18,0µm	59%	82%	43%	79%
200ms_4,00N/mm_27,0µm	69%	105%	43%	61%
200ms_4,00N/mm_27,0µm	68%	110%	46%	59%
200ms_4,00N/mm_27,0µm	61%	103%	42%	56%
100ms_4,00N/mm_36,0µm	87%	105%	61%	52%
300ms_4,00N/mm_36,0µm	86%	103%	66%	76%
200ms_6,00N/mm_18,0µm	34%	111%	21%	70%
100ms_6,00N/mm_27,0µm	85%	139%	36%	73%
300ms_6,00N/mm_27,0µm	72%	141%	46%	73%
200ms_6,00N/mm_36,0µm	132%	166%	85%	102%

Oxygen transmission of sealed packages

Figure 5 compares the recalculated value OTR of the flat film with the sealed package oxygen transmission of each tested commercial film. There is no influence of ultrasonic (or heat conductive) seal permeation on the total package permeation with the high and very high barrier films. With the medium barrier film the oxygen permeation is slightly lower with the sealed packages as with the recalculated flat films. As an additional test, the PET/Alu/PE pouches are sealed at extreme ultrasonic settings to evaluate the influence of an ultrasonic cut though seal on the (high barrier) package permeation. The total package permeation of this cut through package is the very low value of $0.48 \pm 2.9 \times 10^{-4}$ cc/package.day.

Figure 5: Comparison of recalculated flat film and sealed package oxygen permeation

4 Conclusions

A method, with a Box-Behnken experimental design, is described to optimize ultrasonic seal strength. Expansion towards more seal performance parameters (e.g. energy consumption) is possible and subject for new studies.

The ultrasonic seal performance within one polyolefin group (PE or PP) is similar in many ways, a detailed analysis of parameters (seal strength, seal compaction, energy consumption, size seal window) is necessary to select a best material within one group. For PE, metallocene LLDPE-C6 is selected as best ultrasonic sealable monolayer because of the combination of a broad seal window, a good seal strength and a low energy consumption. Previous findings report on the good (ultrasonic) seal properties of metallocene catalysed PE [5, 12]. For PP, raco PP is selected as best ultrasonic sealable monolayer because it has the broadest seal window (13/15) of all tested monolayers in combination with good seal strength.

Laminated films have an increased ultrasonic seal performance compared to the monolayer materials. OPA and PET laminated PE films have an equal or broader seal window than monolayers PE with a similar total thickness. Most of the OPA and all of the PET laminated films have a lower energy consumption compared to the PE monolayers with similar thickness, when sealed at the same ultrasonic settings. PET is chosen as best laminate because the maximum ultrasonic and heat conductive strengths are similar and because the energy consumption is lower than the OPA laminated films for most of the tested ultrasonic settings (only low energy settings show no difference). As the choice of laminate layer has a big impact on the ultrasonic sealing performance, as seen during this research on two common laminate layers, more research is needed to optimize the laminate composition.

Ultrasonic sealing seems to have no effect on the oxygen transmission of packages, compared to recalculated flat film values. These results differ with recent findings on thermosealed packages [13], possibly because this research is done on laboratory made seals instead of industrial seals. However, more research is needed to explain the difference in oxygen permeation with the medium barrier film.

5 Acknowledgements

This research is done within the IWT-TETRA project “ULTRASEAL: The potential of ultrasonic sealing in packages” funded by VLAIO.

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