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Ink Migration Barrier and Direct Food Contact Heat Sealing Lacquer Design for Aluminum / Pet Blister Structure

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Abstract: The packaging industry as a pioneer in waste production is looking for environmentally friendly and green solutions in these days. These factors are driving the development of packaging and forcing the industry to invest in research and development to make flexible packaging more sustainable than the existing packaging options. In this review, the factors of main ingredient's selection on migration barrier properties have been investigated within the new designed heat-sealing lacquer. The new heat-sealing lacquer design has been carried out, which will ensure thermal sealing in Aluminum/Pet Blister Packaging structure, suitable for direct contact food, and at the same time prevent the migration of printed ink to the secondary packaging layer, Pet film side. Because of laboratory tests, the effects and results of heat-sealing lacquer formulations have been assessed and prevented the negative ink migration tendencies in existing heat-seal lacquers for blister packaging structure. That would be possible to use the obtained PET film by recycling processes as a green solution to flexible packaging sector. In this way, Pet films can be separated and used in recycling processes when blister packaging printed with new designed heat-sealing lacquer.

Keywords: Blister packaging, direct food contact, heat-seal lacquer, migration, recycling process.

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1. INTRODUCTION

In our daily lives, flexible packaging plays an important role in the preservation, storage, distribution and marketing of food and derivatives. Packaging materials as a principal function is to keep food safe. It protects food from any damage during the transfer to consumer. The other important function is to attract the consumer's attention. Therefore, the design of packaging and the inks that are applied on packaging are important phenomena on the perspectives of marketing. Therefore, the widespread use of plastic based, flexible packaging materials have caused about the waste problem for environment in the last 50 years (TurkChem, 2020). Turkish Plastics Industry published a report on Turkey's environment issues in 2020, report is pointing to 9.54 million tons of plastic are produced in Turkey market in a year. Packaging material wastes constitute 41.9% by weight of the plastic consumed plastic amount (Plastics Industry Report, 2020). Despite this high rate, just 4.46% of these consumed plastic wastes can be collected in our country for recycling (Pagder, 2020). Based on literature survey and on author's knowledge at the situation in the world, the market research shows that 146 million tons thousand tons of plastic packaging is consumed to waste in world in 2015. Against that huge consumption, only approximately 29.2 million tons of this is collected for recycling in 2016 (Ritchie, 2018).

Figure 1 shows that packaging wastes are dominant for the waste generation globally accounting for 47 percent between 2015 and 2021. The other sectors are others, textile, personal care, storage wastes, construction, and electrical equipment, respectively. Figure 2 represents the waste management percentages in world again between years 2015 and 2021. It can be understood that only 19% of these collected packaging wastes are managed as suitable for recycling. A large portion of these wastes is going to landfilling or the natural environment and incineration (Ritchie, 2018).

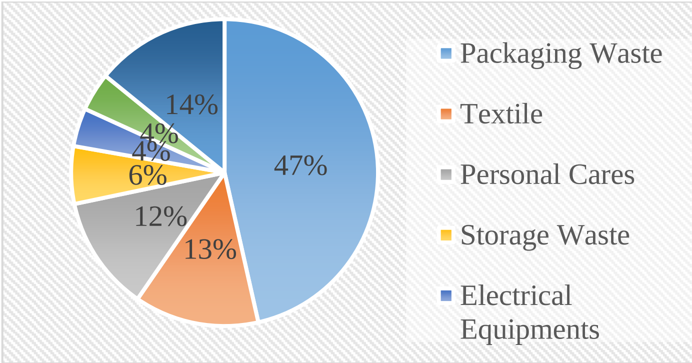

Figure 1: Waste generation in world between 2015 and 2021(Ritchie, 2018)

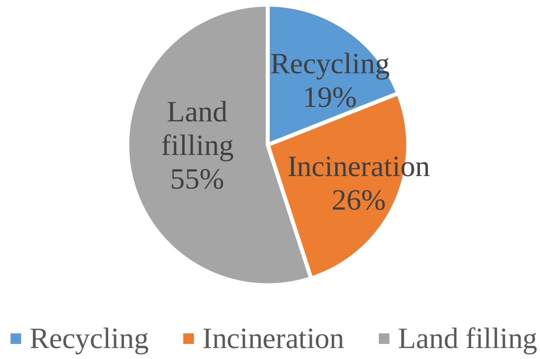

Figure 2: Waste management in world between 2015 and 2021 (Ritchie, 2021)

So that, recycling of these packaging wastes is a necessity. The increasing awareness of environmental protection and economic consideration direct us to regain these values, which are classified as a waste. These waste materials are our valuable raw materials stocks and energy stocks, which can be recyclable.

The packaging industry is seeking for innovative environmental solutions to avoid depleting raw material reserves. The environmental pollution can be decreased by the help of new environmentally designs for packaging sector. The main result of that small amount of recycling rate can cause the impurities on consumed packaging materials. However, the inks and paints applied on packaging materials cause a migration that pollutes flexible packaging materials. As a result, consumed packaging material will not be suitable for recycling processes due to stain and impurities on it. A study by Pinter et al. also showed that contamination issues affect the quality of the recycled material in terms of opacity and densitometric color properties. Blushing and staining by any ink are considered as unrequested parameters for mechanical recycling. Because of these impurities in recycling processes, the recycling rates and repetition are reducing. Today, PET packages constitute the biggest consumption amount plastic wastes. Therefore, blister packaging has experienced noticeable growth over the past years. The blister packaging market was valued at USD 21.3 billion in 2020. Blister Packaging is projected to be worth USD 38.17 billion by 2027. The compound annual growth rate (CAGR) of 7.2% during the forecast period (2021 - 2027) (Blister Packaging Market Report, 2021). Thereof, recycling of PET wastes has become an important phenomenon because of increasing awareness of environmental protection, economic requirements, and raw material depletions. According to market research and available data, we can understand that PET is one of the most common thermoplastic polymers for packaging industry. Again, in 2021, Pinter et al, considered that PET would be the most recyclable food-packaging plastic. The use of this material is expected to increase. The reason behind is to ability of absorption of post-consumer contaminations at lower levels, when we evaluate with other plastics like polyethylene and polypropylene. That chemical characteristic of PET makes it more acceptable for the recycling process in terms of high recycling rate and percent of usage values (Pudack et al., 2019).

These huge consumptions and forecast amounts again direct us to the necessity of recycling. The widespread use, chemical and commercial properties need to be evaluated in terms of the environment. Therefore, every product that is placed on the market should include some thought to its end-of-life and how its associated packaging will be disposed of and recycled. The biggest parameter preventing this recycling of PET is that the material is contaminated with ink.

The aim of this study is to design a heat-sealing lacquer, which will prevent contamination of ink to PET material used in blister packaging. The new heat-sealing lacquer design prevents the transfer of ink to secondary substrate, PET material after sealing. The migration of ink from aluminum material to PET film because of the heat-sealing process. The newly designed formulation of heat-sealing lacquer prevents ink migration that pollutes flexible packaging materials. With the help of a new design heat-sealing lacquer, the obtained PET film after separation processes can be suitable to reprocess in recycling processes for sustainability. The new heat-sealing lacquer is also designed to allow recycling and at the same time, it will be suitable for direct food contact (DFC) product status. The improved packaging will provide food safety and help reduce the carbon footprint by the help of recycling of packaging material. Thereby, significantly contributing to the sustainability of the recycling of PET can be available.

2. EXPERIMENTAL SECTION

2.1. Packaging Materials

The blister packs are combined with two different films. The primary material is aluminum film which the design to be printed with ink and the secondary (other) material is PET film. Both materials are adhered with thermally to create blister packaging (Allen, 2010).

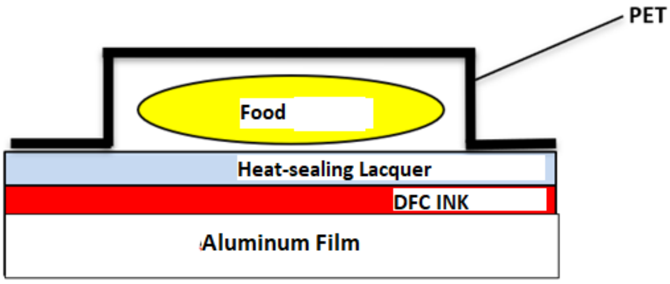

Figure 3: Blister Packaging Structure.

The heat-sealing lacquer and the printing ink are two main components that accompany these two major layers. The first component ink supply packaging's visual design, aesthetic, and instructive aspects. Another component, heat-sealing lacquer is applied for giving adhesion between the two halves of the blister. Figure 3 shows the direction of heat seal lacquer application side and structure of the blister packaging. Heat seal lacquer applied on the primary material, which is aluminum foil on the top of ink printed designs. This application provides sealing between primary material and the secondary material, which is PET film by the help of heat-sealing lacquer. Sealing of flexible packaging film can be closed by the most common method which has been done by seal bar (seal jaw) sealing in the food industry (Ilhan et al., 2021).

2.2. Methods

After detailed literature and market research related the DFC HSL groups done, the formulations of heat-sealant lacquer have been developed with various resin groups belongs to variable producer which learnt form literate survey and old experiences on heat-sealing lacquers. The heat-sealing lacquer designed in this study prevents the migration of ink to the PET film while providing thermal sealing in blister packaging applications with the help of compatible resin and additive selection. Thus, PET film is not contaminated and stained with ink. It is possible to use the obtained PET film by recycling as pure material source again. Therefore, heat-sealing lacquer allows excellent thermal sealing of used materials. Therefore, as a primary packaging level based on the product's proximity and intended usage of blister packs are in direct contact with the food products (Lalpuria et al., 2012).

Figure 4: Current Heat- Sealing Lacquer Applied PE Film.

Figure 5: New Heat- Sealing Lacquer Applied PET Film.

In Figure 4 shows that when the blister packaging, which applied with current heat-sealing lacquer, separated, the ink migrates from aluminum material to PET. Since the PET film is contaminated with ink, it cannot be recycled. However, with the new study, as seen in Figure 5, there is no ink contamination have been seen on separate PET film. When the PET film is separated from the aluminum after the heat-sealing process, the PET film stays clean which is suitable for recycling.

Experimental studies were first named ADHS coding system to understand the effect of resin types initially. Then, within selected resins, HSL formulation studies were followed to heat seal formulation roofs to find the best match with the desired design parameters. All the test measurements were carried out at Sun Chemical facilities.

In laboratory tests, the selection of resin types has been evaluated according to their following fundamental requirements initially.

- Adhesion
- Heat sealing strength
- Ink migration properties on PET, after separation of structure

In all formulations, the solid contents of variable resins were set to 25% value for fair consideration of resin selections. Out of 28 commonly used resin types, 8 of them were chosen according to the performances below. The new formulations of heat-sealing lacquer have been developed with these selected resin groups.

- Film-forming properties
- Adhesion
- Sealing strength
- Tear resistance
- Resistance properties (heat, mechanical and chemical)
- Solvent release
- Anti-blocking property
- Stability

In addition, non-migratory resin type for protecting ink contamination to PET side was the most important phenomena for these selections. Extra performance and resistance properties that could be obtained with the use of more than one resin were evaluated. Thus, co-binder usage was evaluated; the compatibility of resins was tested. As a second step of study, additive usage in formulations gives other wanted properties of formulations. Additives alter the lacquer physical properties to boost different situations, like enhancing the flexibility of the printed film, increasing mechanicals resistance such scrub, rub off. Additionally, they can promote adhesion, surface properties and other required special effects.

Formulation structures that would not cause the migration of ink and any ink stain to the PET film side after separation of blister pack while providing thermal sealing in blister packaging applications were tested.

3. RESULTS AND DISCUSSION

3.1. Adhesion Control and Viscosity Measurements

Adhesion as a control parameter is the primary requirement as mentioned before for designed lacquer. Therefore, the resistance of the coated substrate to mechanical and chemical deformations, which can cause the removal from that dried, solid coated substrate in terms area, were measured for that fundamental purpose. Also, for multicoating systems, as in that study the term of inter-coating adhesion force refers to the resistance of a heat-sealing lacquer coating layer from the bottom ink coating layer.

Table 1: Viscosity and Dilution Rates of Formulations with Common Solvents Used.

Formulation No	Initial Viscosity (F4,@ 20^{0}C)	%Dilution Rate with ETAC to V=18''	% Dilution Rate with ETOH to V=18''	% Dilution Rate with ETOH:Ethoxy Propanol (1:1) to V=18''
HSL1	26	12	9	11
HSL2	42	36	23	29
HSL3	13	0	0	0
HSL4	74	60	50	60
HSL5	45	35	33	35
HSL6 (Final formulation)	60	46	42	46

Table 1 shows initial viscosity values of studied and existing-conventional lacquers. There are also percent of dilution rates are tabulated with different solvents to adjust 18 seconds printing viscosity, which is appropriate gravure printing. Final formulation HSL6 is compatible with all solvents as a good opportunity for variable applications.

3.2. Heat Sealing Strength

Heat-sealing strength was the other important parameter for this study. The sealing quality and integrity are essential requirements for final design of blister packaging. Evaluations of sealing strength and adequate final sealing property were done at first varnish's formulations with variable resin types. All prepared drawdowns of ADHS varnishes, sealed with heat sealing device at conditions were 1 bar during 1 second, and 180 ºC temperature as a condition 1 parameters. After heat-sealing process, the sealing strength measured in terms Newtons with Loyd device.

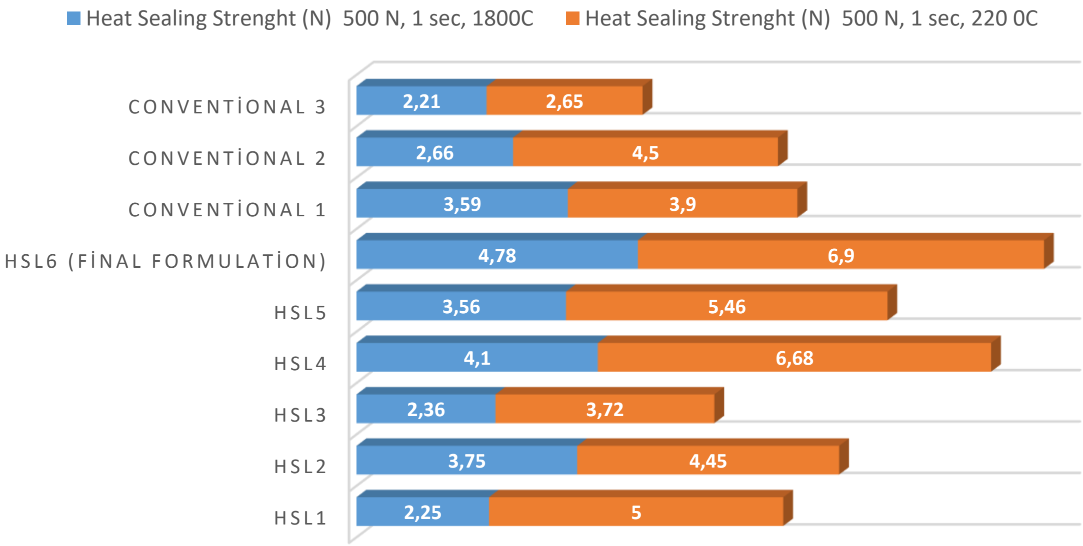

Figure 6: HS Strengths in Newton at sealing condition 1 and 2.

From Figure 6, it was seen that at both temperature conditions HSL 6 formulation has higher bond strength than other studied and existing, conventional heat seal lacquers. In addition, high temperature effect better bond strength was observed as expected.

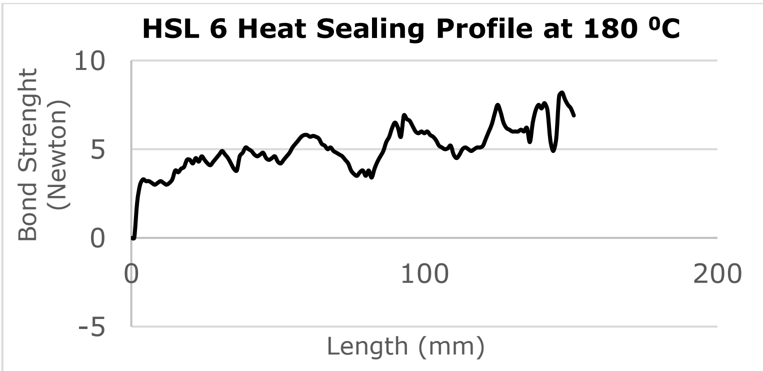

Figure 7: HSL 6 HS Profile at 180 °C.

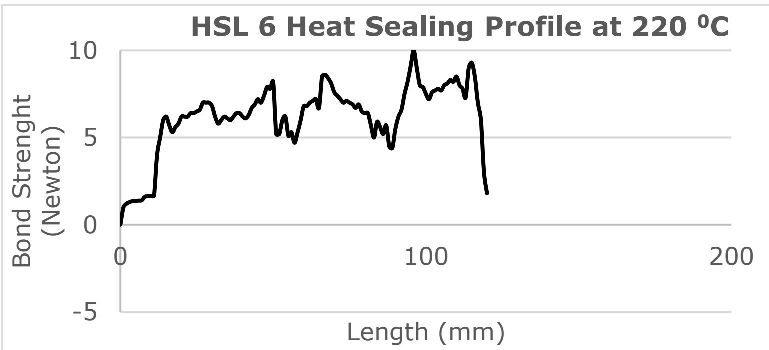

Figure 8: HSL 6 HS Profile at 220 °C.

In addition, Figure 7 and Figure 8 show the heat-sealing strength profile of HSL 6 at sealing condition 1 and condition 2. In figure's X-axis, the load results plotted on the graph in terms of newton and Y-axis shows the length of measured printed the strips with printed full structure of design. In Figure 8, film tearing was observed at 220^{0}C temperature sealed strip, while measuring the sealing force with the device. Therefore, that indicates quite a high enough bond strength that tears the aluminum film.

3.3. Migration and Rub-Off Test Results

The migration and rub-off were the main issue for this study. With the currently used heat-sealing lacquer design, the ink, which applied to main surface, aluminum side of blister, migrates to the secondary material (PET) surface. Because of that migration, the PET film was stained as seen below figure, which evaluated as one grade. Therefore, both surfaces of packaging are damaged by ink. Both surfaces cannot be recycled in this circumstance. Within the new design parameters, all ADHS varnishes were tested to find non-migratory resin types. Test results were rated between 1 and 5 scale by visual controls in Figure 9.

In results, grade 1 represents complete ink migration to PET side. Grade 5 represents the clear PET, no ink migration in Figure 9 after heat-sealing and cooling steps of prints. PET and aluminum were necessary to peel from each other. Then, PET film controlled for any removal, migration of ink.

Figure 9: Migration and Rub-Off on PET film.

3.4. Fourier Transform Infrared Spectroscopy (FTIR) Results

Migration and rub-off results were measured by FTIR for the HSL formulations. In FTIR tests, the peeled PET films after heat sealing and cooling processes were analyzed to be sure that there was no residual from the ink contamination, which indistinguishable by the visual tests on PET film side. FTIR analyses of four samples were performed with wave numbers between 4000 and 600 cm-1 on PET peeled film. Fourier Transform Infrared Spectroscopy was used to identify the surface of peeled PET film and neat PET film by using infrared light. Therefore, the result of FTIR analysis observed functional groups, characteristic peaks and impurities of the samples compared to neat, unprinted PET film. The sample results were interpreted as no migration when parallel to the scanned wavelength and peaks for the neat PET film surface. There were three trials done in that step as an elimination of all laboratory test results. The below characteristics bands show these three peeled samples in red, blue, and green colors, respectively. In addition, the unprinted PET sample was represented in purple color. From the fingerprints of these four results, we can say that the green and purple peaks overlapped. The other two characteristics bands had represented different peak and small vibrations due to absorption increasement and different chemicals migration from applied ink and HSL formulations of samples.

FTIR was used to check the efficiency of the final design HSL 6, so from the result of spectrum, the green colored transmittance bond shows the same functional groups present on neat PET film. The spectrum of transmittance bond of HSL 6 and pure PET film showed higher than 95 % similarities with their transmittance bon with same vibrations as seen Figure 10.

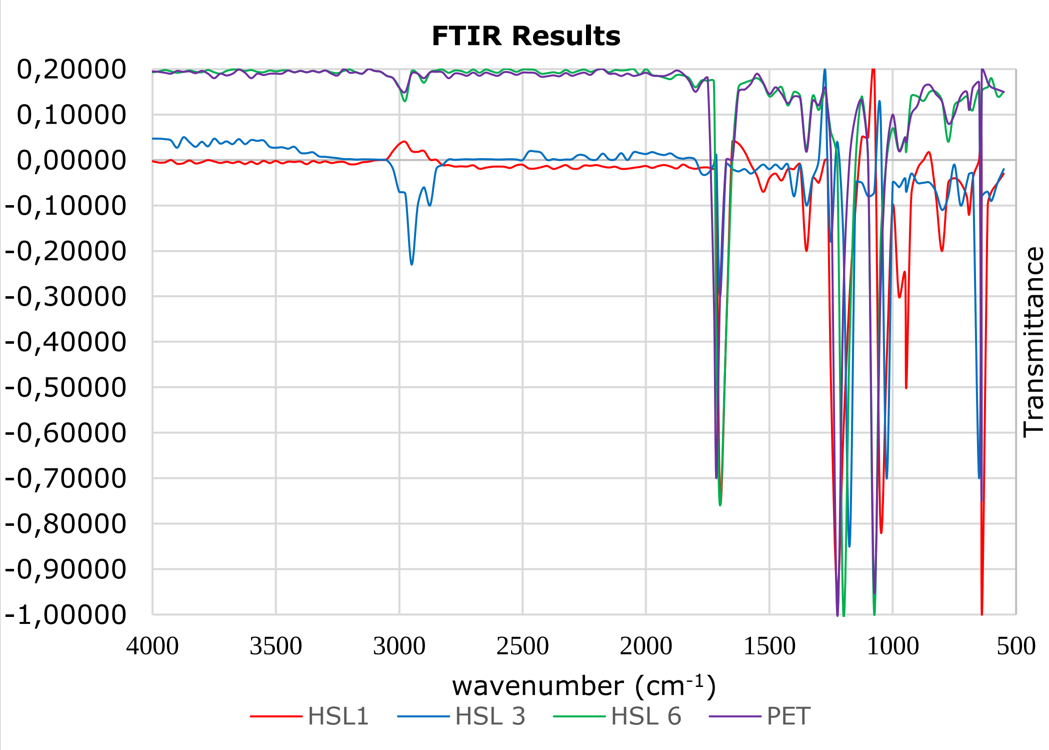

Figure 10: FTIR Result of HSL Formulations.

3.5. Coefficient of Friction (CoF) Control Results

Formulations HSL 5 and HSL 6 were tested to get the correct coefficient of friction levels to suit the prin-ting process requirements. The amount and types of used wax particles were adjusted for this purpose. In addition, the conventional lacquers CoF values were compared to new design because of their old industrial trials based on the positive feedback. As a target value, CoF set to between 0.25 and 0.50. Therefore, The CoF test results were shown in Table 2, HSL 6 formulation measured in the target value ranges and parallel results with respect to conventional lacquers in face-to-face and face to metal test methods with successfully.

Table 2: CoF Test Results.

Formulation No	COF	
	Face to Face	**Face to Metal**
HSL6	0.45	0.25
HSL5	1.92	1

3.6. Block Resistance Test Results

When printing processes on non-absorbent, flexible materials like aluminum, the designed lacquer with a strong adhesion, elasticity and sealing properties it is getting very hard to balance the drying and anti-blockage properties at the same time. The usage of appropriate wax compounds and additives to lacquer formulations gives positive effect also in terms of blocking properties as well as the tendency to migration, slip and scratching resistances (Leach at al., 1993). Furthermore, efficient drying conditions controlled to ensure that applied lacquer set to avoid blocking issues in the printing process indispensably. Figure 11 shows block test result of HSL formulations at different conditions. From drawdowns it was seen that some block and migration problems on HSL 4. Some part of coated ink migrates to other side of aluminum film. In addition, increased test temperature caused more block and migration tendency for HSL 4. Therefore, the HSL 4 formulation was eliminated because of the block resistance test by visually.

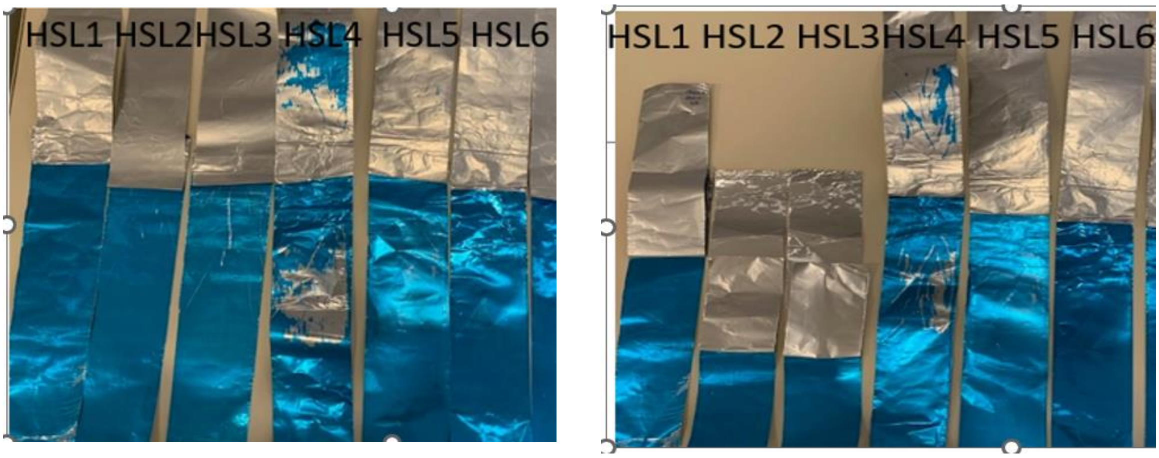

Figure 11: Block Test Result of Drawdowns at conditions 45 0C, 3 days, 45 kg & 20-25 ^{0}C, 1 week, 10 tons.

Table 3 was tabulated according to block formation of drawdowns under both test conditions. Ass seen table HSL 6 and conventional heat-sealing lacquers had been evaluated 0 which means no formation of blocking. The aluminum film can be peeled off over the coating surface easily, Other HSL formulation could not passed this test.

Table 3: Numerical Block Test Results.

	Block formation: 5 (high blocking) - 0 (no block formation)					
Conditions:	**HSL 1**	**HSL 2**	**HSL 3**	**HSL 4**	**HSL 5**	**HSL 6**
45^{0}C, 3 days, 45 kg	2	0	0	5	2	0
20-25^{0}C, 1 week, 10 tons	4	3	2	5	4	0

3.7. Solvent Retention Test Results

The presence of residual solvents in packaging changes the taste and odor of the finished products as the first to notice. Residual solvent prevents the packaging from closing by sealed securely. The formulation of lacquer designs is taken to ensure and tested in term of solvent release properties. In efficient drying or inappropriate resin and solvent selection in formulation structure cause blocking issues in the printed reels. The printed substrates that come in contact with food need to be controlled according to strict regulations and standards. Therefore, retained total solvents amount was limited with the 10 mg/m2 for various converters as a threshold in food packaging. By nature of solvent-based lacquers, a gravure printed substrates still contains some amount of solvent after drying processes. When consideration is done for, the commonly used threshold values for residual solvent amount the formulations to be compatible low-odor and solvent retention grades. Figure 12 shows the graphical representation of HSL 6 formulation and conventional heat-sealing lacquer in the interval of different drying conditions to understand the solvent retention profile of final prints. From the graph, we can understand that HSL 6 releases the solvent with a steeper acceleration and similarity in terms of drying and retention profile with conventional one. In addition, in Table 4 the total solvent residual amounts are shown in terms of mg/m^2.

New design HSL 6 show lower solvent residual amounts in certain drying conditions compared to industrial threshold for solvent residual at 10 sec., 80 °C and 5 min., 20 °C drying conditions which is lower values of currently used conventional heat-seal lacquer.

Table 4: Residual solvent amounts [mg/m^2] for different drying conditions.

Drying Conditions	Conventional 1	HSL 6 (Final Formulation)
30 sec, 20 ^{0}C	15.5	18.3
10 sec, 80 ^{0}C	6.01	4.4
5 min, 20 ^{0}C	2.54	3.05

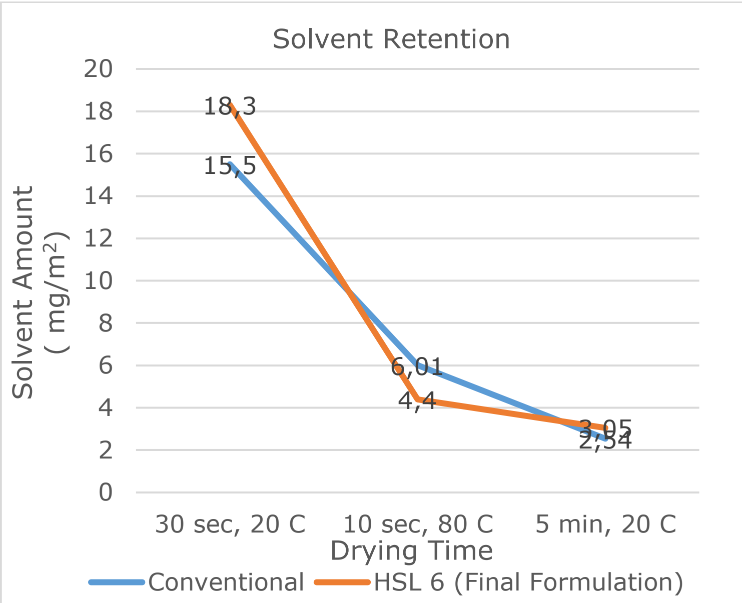

Figure 12: Residual solvent amounts [mg/m^2].

4. CONCLUSION

The study dealt with the heat-sealing lacquer which has ink migration barrier property and applicable for direct food contact for Aluminum / PET Blister packaging to used resin and additive types in formulation. With the help of these selected components, finished packaging material PET film will be suitable for recycling. The heat-sealing lacquer to be designed has been provided thermal sealing and other technical properties in blister packaging applications as like as currently used conventional lacquers, which are not recyclable due to migration issue. Since the blister, packages made with the newly designed heat-sealing lacquer observed that prevents ink contamination to the PET film side.

Therefore, the PET film is suitable for use in recycling processes. Thus, the biggest consumed plastic waste of PET package is possible to use the obtained PET film by recycling as pure material source again when the new designed heat-sealing lacquer is used. In addition, recycled PET can be used as a resource again in raw material reserves.

Environmental pollution will be decreased; new environmental solutions will be presented to the packaging industry. Environmental protection and economic saving can be done. According to laboratory tests and literature surveys, elastomeric PU was chosen as the main resin in the new design. Good sealing performance was achieved by using PVB resin with Elastomeric PU resin.

1- 5 % amount of CAB resin gives excellent anti-blocking and release property to heat-sealing lacquer formulation. On the other hand, it is experienced that adhesion promoter usage created migration tendency of ink to PET side.

1 - 3 % PE wax usage improved rub, mar, and abrasion resistance of final formulation also, decreased FTF CoF value from 1.92 to 0.45, which is suitable for printing process.

The new formulation has higher heat-sealing bond strength than existing conventional heat-sealing lacquers, which are not recyclable.
Therefore, according to that observation the factors of resin and additive selection on migration properties of heat-sealing lacquer for Aluminum / Pet Blister structure was completed successfully with the laboratory results and industrial trial.

In addition, another valuable output is this requirement carried out by using the resources of our country. The most importantly, the printed blister packages with this new designed heat-sealing lacquer can now be recycled and gain re-value as a raw material and energy stock in comply with the circular economy plan.

5. CONFLICT OF INTEREST

The authors declare no conflict of interest.

6. ACKNOWLEDGMENTS

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